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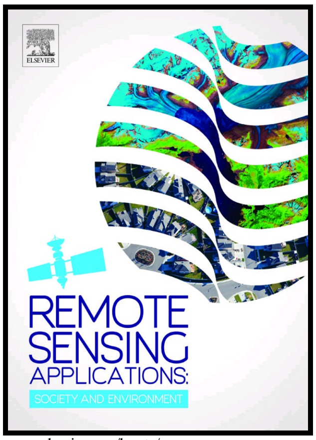

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Measuring Landfill Methane Emissions using Satellite and Ground data

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Abstract

Landfill methane emissions (LME) vary in short periods depending upon the meteorological and atmospheric conditions. In this paper, coupling the Atmospheric InfraRed Sounder (AIRS) with the tracer dilution method (TDM) is proposed during unmeasured emission days to have a better annual estimation of the LME. Some assumptions were made to develop this proposed model. The atmospheric model Advanced Regional Prediction System (ARPS) was employed to evaluate assumptions made during emission estimation using the proposed technique. Methane emissions of a landfill for 13 days during 2011-2013 were measured by the TDM and filtered to remove unreliable data. Then, the filtered data was employed to train the proposed linear regression model to estimate methane emissions. Daytime methane vertical profile concentrations (DMVPC) and nighttime methane vertical profile concentrations (NMVPC) were utilized to study correlations between ground field and satellite measurements for model training. Because field measurements were carried out around noon times, the DMVPC data showed a stronger correlation. Finally, both the TDM interpolation, which is the (normal approach for annual emission estimation) and a coupled of remote sensing (RS) and the TDM technique were utilized to estimate annual LME. The results revealed that interpolating TDM measurements with wide gaps underestimates the LME by about 13% compared to this new RS- field technique, which produces a higher estimation of LME.

Keywords: Landfill, Satellite, Methane emission, Remote sensing, Atmospheric modeling, Tracer dilution method.

Introduction

Methane is considered the second strongest contributors to global warming after carbon dioxide (Lemke et al. 2007). Methane emission from biological decompositions of buried waste in landfills is known as an important source of anthropogenic methane emissions (Spokas et al. 2015). To develop or evaluate the measures that control LME, and to estimate LME for greenhouse gas inventories, it is necessary to first quantify LME.

LME vary annually depending upon waste disposal rates, type of sources, meteorological and atmospheric conditions (Foster-Wittig et al. 2015). It also has a strong seasonal dependence due to variations in soil temperature and moisture that highly influence microbial activities and gas transport in soils (Stern et al. 2007; Wang et al. 2011). LME are also found to be highly dependent on the barometric pressure for a period of several days. For example, Czepiel et al. (2003) measured methane emissions from a landfill and found that LME increased by a factor of five when the barometric pressure dropped approximately 15 mb over a period of one month. Xu et al. (2014) estimated methane emissions from part of a landfill and attributed 35-fold variations in LME to changes in barometric pressure changes during a two-day period. Along with annual and seasonal variations, LME also show strong diurnal and subdiurnal variations. LME might also be affected by changes in surface winds. Delkash et al. (2016) applied tracer dilution method (TDM) to measure the methane emissions from a landfill. Aided a set of numerical simulations, they found significant changes in landfill methane fluxes due to changes in surface wind speeds and directions. All these studies indicate that LME have strong temporal variations. Therefore, a quantification method that can capture all these variations is needed.

Several techniques have been developed to quantify LME including: static and dynamic flux chambers (Bogner et al. 1997), vertical radial plume mapping (Thoma et al. 2009), eddy-covariance (Xu et al. 2014), differential absorption LiDAR (DiAL) (Robinson et al. 2011), inverse plume modeling (Zhu et al. 2013), and TDM (Delkash et al. 2016; Foster-Wittig et al. 2015). All these techniques can only be used for short periods (e.g. the TDM, flux chamber) or limited footprints (e.g. the eddy covariance technique) due to various limitations especially from logistic. To estimate annual emissions, a linear piecewise interpolation is usually adopted for daily emissions to fill the gaps during two consecutive measurements. The accuracy of this approach is often compromised by temporal emission variations due to barometric pressure, wind speed, soil moisture and soil temperature changes that were noted above. Therefore, emission measurements can be inaccurate with uncertainties greater than 200% compared to other methane emission sources (IPCC, 2006). Thus, a continuous estimation of LME is required for a better and more reliable annual estimation.

Remote sensing (RS) techniques have emerged as an important tool for qualitative and quantitative evaluations of land, ocean, biophysical, and atmospheric parameters and to understand the coupling between land, ocean and atmosphere (Singh et al. 2010; Singh et al. 2007). RS techniques using multi-sensors are commonly used in qualitative change detection. Quantitative evaluations require detailed validations of RS data with ground observations. One of the applications of the RS techniques in environmental engineering is quantification of gas emissions. Satellite technology has provided information about spatial and temporal evaluation of methane concentrations globally and source emissions are estimated by an inverse modeling approach. Recently, number of satellite based studies have been carried out and methane concentrations have been mapped from different regions using SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY) and AIRS data (Prasad et al. 2014; Rajab et al. 2011; Uspensky et al. 2011; Xiong et al. 2010). Kort et al. (2014) observed very high methane emissions from the Four Corners of the US using satellite data.

Although satellites have been widely used to estimate gas emissions from different sources, they have not been applied to estimate LME. This technique requires several daily field measurements to train a linear model. As note before, field measurements are costly and are carried out sparsely. The application of satellite images in estimating LME was pursued. The present study is the first application of satellite images in estimating landfill emissions and is important in estimating annual LME, especially when continuous ground data is lacking.

In this paper, a regression model was considered to retrieve methane concentrations from AIRS data for accurate estimations of annual LME. AIRS on EOS/Aqua platform was launched as a thermal infrared sounder in the year 2002 to improve weather forecasting and provide methane products. This satellite gives reliable results for methane concentrations. The accuracy of AIRS products for methane data has been validated using aircraft campaigns (RMSE less than 1.5%) (Xiong et al. 2010). This study compared methane concentrations retrieved from the satellite with ground observations and study the capability of the RS technique to use as a reasonably fast and economical tool in providing a better estimation of annual LME. We have further studied correlation between methane concentrations obtained from AIRS data and the measured LME at Turkey Run landfill, Georgia (USA). The accuracy of the proposed technique was examined using correlations between barometric pressure and LME, which are well-understood.

2- Materials and Methods

2-1 Theory

This paper suggests a new methodology for measuring methane emissions from landfills. This method correlates methane emission variations with variations in methane concentration of the area where the landfill is located. Signals of landfill emission variations are separated from noise from other emission sources. In order to differentiate landfill emission signals from other noise, some assumptions are made. First, LME signals should be detectable from the background, which means LME must be higher than emissions of other sources. Second, all methane sources except the landfill are assumed to have relatively steady emission rates. This assumption associates all variations in methane concentrations that are detected by AIRS with variations in landfill emissions (http://ghgdata.epa.gov/ghgp/main.do).

To develop the proposed method, the mass balance concept is considered. A set of tiles of AIRS images around the landfill is required. Under daytime convective conditions, each AIRS cell is assumed to be a completely stirred tank reactor (CSTR), such that methane is considered well-mixed at the scale of the cell (roughly 1^o x 1^o), see the schematic in Fig 1. Under this assumption, effluent methane concentration leaving the cell is equal to that at the center of the cell, as measured by AIRS. Moreover, a quasi-steady state mass transfer leading to non-accumulative conditions is assumed to remove short-term temporal variability in methane concentrations. Under these two assumptions, the mass balance equation for methane in the landfill cell is given in 2D form in Eq. 1 for simplicity.

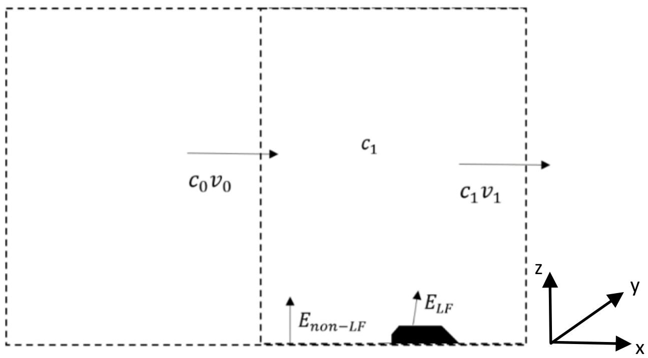


Figure 1- Schematic view of the proposed CSTR model for methane transport.

$$\underbrace{\int_0^H \int_0^{L_y} c_0 v_0 \, dy \, dz}_{Inflow\ mass\ flux} + \underbrace{E_{LF} + E_{non-LF}}_{Sources} = \underbrace{\int_0^H \int_0^{L_y} c_1 v_1 \, dy \, dz}_{Outflow\ mass\ flux} \quad (1)$$

$$\underbrace{\int_0^H \int_0^{L_y} c_0 v_0 \, dy \, dz}_{Inflow\ mass\ flux} + \underbrace{E_{LF} + E_{non-LF}}_{Sources} = \underbrace{\int_0^H \int_0^{L_y} c_1 v_1 \, dy \, dz}_{Outflow\ mass\ flux}$$

where c_0 and c_1 are the DMVPC at the inflow and outflow boundaries of the landfill cell, respectively. v_0 and v_1 are the wind vector components normal to inflow and outflow boundaries. E_{LF} and E_{non-LF} are the landfill and non-landfill methane emissions, respectively. L_y and H are the width and height of the landfill cell. By the CSTR assumption, c_1 and v_1 are representative of the cell concentration and wind speed. Further, they are horizontally uniform among variations in the y direction. Therefore, the mass fluxes in Eq. 1 simplify to Eq. 2.

$$\int_0^H \int_0^{L_y} c_0 v_0 \, dz \, dy = L_y \int_0^H c_0 v_0 \, dz; \qquad \int_0^H \int_0^{L_y} c_1 v_1 \, dy \, dz = L_y \int_0^H c_1 v_1 \, dz \quad (2)$$

The validity of these assumptions will be discussed later with atmospheric modeling. Further, using the weighted average velocity, the integral of wind speed in elevation is equal to the vertically average wind speed $\bar{v}$ multiplied by integral of elevations.

$$L_y \int_0^H c_i v_i \ dz = L_y \bar{v}_i \int_0^H c_i \ dz; \qquad \bar{v}_i \stackrel{\text{def}}{=} \frac{\int_0^H c_i v_i \ dz}{\int_0^H c_i \ dz}, \ \ i = 0,1 \qquad (3)$$

Substituting Eq. 2 and 3 into Eq. 1 yields Eq. 4.

$$E_{LF} = L_y \bar{v} \, [\int_0^H (c_1 - c_0) dz] - E_{non-LF} \qquad (4)$$

From Eq. 4, the vertically integrated methane concentration difference between the landfill cell and the adjacent upwind cell has a linear relation with the landfill emission under the abovementioned assumptions.

2-2 Field methane Emission Measurements

Turkey Run landfill, located in Georgia, USA was selected for this study. This landfill contains common household and residential waste. The TDM was utilized to quantify LME during several days. In this method, the tracer gas is released at a certain emission rate from tracer releasing points located on the landfill. Under fully mixed conditions, the same transport patterns for the tracer gas and methane in the atmosphere are assumed. At the same time, the tracer gas and methane concentrations are measured far enough downwind of the landfill, where both the landfill and the tracer releasing points are taken as point sources. It assumes that both the released tracer gas and the emitted methane are sufficiently well-mixed downwind of the landfill. The mass flux of methane can be calculated by multiplying the known tracer flux to both concentrations and molecular weights ratios.

$$E_M = E_T \frac{M_M}{M_T} \frac{C_M}{C_T} \qquad (5)$$

where E_M and E_T are the methane and tracer gas emission rates expressed as mass flowrate $[MT^{-1}1]$, M_M and M_T are the molecular weights of the methane and tracer gas ($\frac{g}{mole}$), and C_M and C_T are the concentrations of the methane and tracer [*ppm*]. The success of the TDM is strongly dependent on the consistency of the ratio $\frac{C_M}{C_T}$ during an experiment.

The Cavity Ring-Down Spectroscopy instrument was employed to measure methane and acetylene (C_2H_2) as a tracer gas, to implement the TDM. A model 81000 3- D sonic anemometer (R.M. Young, Traverse City, MI, USA) and high-resolution global positioning instruments were mounted on a mobile mast to determine measuring locations and wind conditions (speed and direction) at 2m above the ground level. Wind direction varies from southerly, southeasterly and southwesterly with a 20º standard deviation in most of the field measurement days. In addition to this, the average wind speed was about 3.6 m/s and varied between 1.7 and 6.5 m/s. The average wind direction was first calculated at every 5min, and then for each day. The daily-averaged wind direction was used to determine the background cell on the upwind side of the landfill cell for daily emission estimations. LME measurements were usually carried out between 9 a.m. and 5 p.m. local time, and most of the observations occurred at noon and in the afternoon. Table 1 displays the distribution of field measurements during the daytime. It reveals that about 87% of the measurements were carried out between 10 a.m. and 5 p.m., which is closer to the daytime passage of AIRS than its nighttime passage.

Table 1- Frequency percentage of the diurnal time that field measurements were carried out during a two-year campaign in Turkey Run landfill (%)

Diurnal time (hour)	<10	10-11	11-12	12-13	13-14	14-15	15-16	16-17	>17
Frequency (%)	8	13	15	8	9	14	14	15	5

Visual inspections of field concentration measurements reveal that all measured values are not reliable. An acceptable regression should exist between C_M and C_T to represent the average of $\frac{C_M}{C_T}$ more accurately. The $\frac{C_M}{C_T}$ could vary a lot during a field campaign, which weakens the certainty about methane emission quantifications. Lack of strong correlation in various data can be due to various factors. The TDM works well under atmospheric conditions when the wind is strong and unidirectional. The actual conditions can be far from ideal during calm measurement days when wind speed is mild, and wind direction fluctuations were strong. Further, appreciable differences must exist between measured concentrations and background concentrations for the TDM to apply. Thus, data quality control is needed to filter the raw data in order to meet the abovementioned criteria. Mønster et al. (2014) reported that some errors in the TDM emission quantification might be related to the post-processing of data. Foster-Wittig et al. (2015) developed some filtering criteria and examined performances of these criteria in the reduction of emission estimation uncertainties. They introduced a set of regression criteria including R^2, emission rate differences, and the signal to noise ratio. The regression coefficient is known as an important quality indicator that presents the level of mixing for plumes. The emission rate difference is useful in case probable emission overestimations must be removed due to low wind speeds overnight. Under this criterion, two different quantification approaches (weighted average and regression) should provide similar emission estimations. The signal to noise ratio filtering criterion helps to differentiate noticeable background variations or weak signals. Further explanations about these filtering criteria and their performances on the TDM accuracy can be found in this reference (Foster-Wittig et al. 2015).

2-3 Remote Sensing Technique

The basic principle of remote sensing (RS) of methane is to measure radiance at particular wavelengths that are sensitive to methane. The relevant products retrieved from satellite data are total column concentrations. The AIRS, which is a high resolution spectrometer with 2378 bands in the thermal infrared spectral region (3.74–15.4 mm) and 4 bands in the visible spectral region (0.4–1.0 mm), measures methane concentrations, temperature, humidity, water vapor and other geophysical parameters with high retrieval accuracy. The AIRS instrument is an infrared spectrometer with a nadir cross-track scanner (Pagano et al. 2003), and was launched into a 705 km altitude polar orbit on the EOS Aqua spacecraft on May 4th 2002. Every day, the satellite crosses the equator at approximately 1:30 A.M. and 1:30 P.M. local time, giving near global coverage twice a day. The AIRS retrieval algorithm is discussed in detail by Aumann et al. (2003). The AIRS measures approximately 200 channels in the 7.66 µm absorption band of methane, of which 71 channels are used to retrieve methane. Xiong et al. (2008) discussed the retrieval of methane, uncertainties, and validation in detail. Level 3 of AIRS data, available in 1^0x1^0 resolution was downloaded from (http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgiinstance_id=AIRS_Level3Daily) over 5 cells as shown in Fig.2. These datasets were collected for day and night times for these 5 cells. The landfill is located in cell 5 and the rest of the cells (1, 2, 3 and 4) are taken as adjacent cells for estimating methane emission.

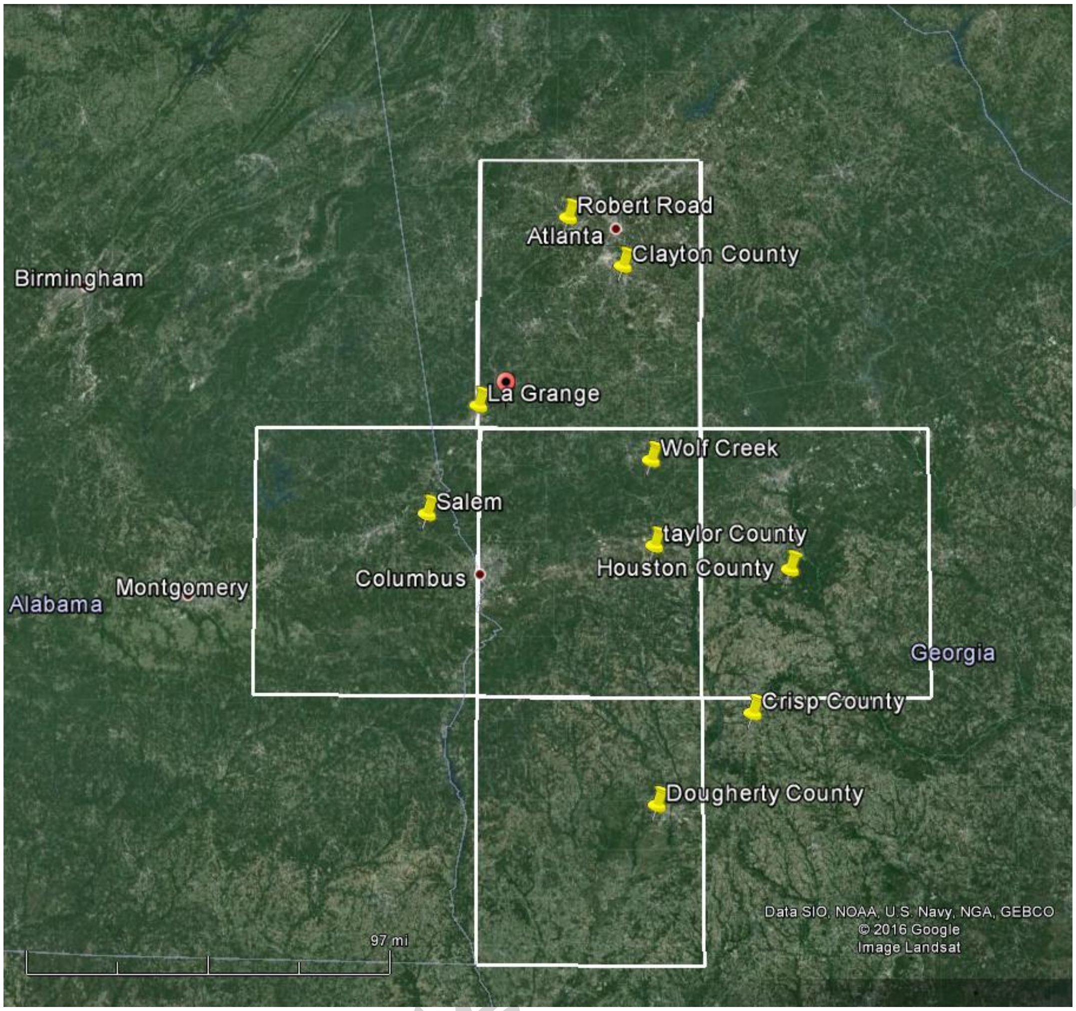


Figure 2- AIRS datasets are considered for 5 cells. Turkey Run landfill is shown by red color, other surrounding landfills are marked by yellow color. Big cities are indicated by red points.

2-4 Methane in Georgia State

Beside solid waste systems, different methane emission sources, such as petroleum and paper industries, have been recognized in urban areas. In order to assess how much landfill methane emissions are discernible compared to other sources, methane emission inventories are required. Methane emission inventories are tabulated in Table 2 for the state of Georgia between

2012 and 2013 according to the USEPA (http://ghgdata.epa.gov/ghgp/main.do). Solid waste is the dominant contributor to Georgia's methane emission. Livestock is another important methane emission source that is not listed in Table 2. Assuming that annual methane emissions from livestock are about 15 kilotons, this source emits about 375 kiloton CO2e. Although this source has a noticeable contribution to total methane emissions, waste systems emit one order of magnitude higher than livestock, which renders methane emissions from waste systems are significant signals.

Table 2- Georgia state Annual methane emission (Metric kilotons CO2 e)

Emission source		Power Plants	Petroleum	Refineries	Chemicals	Other	Waste	Metals	Minerals	Paper
Year	2011	179.8	117	0.8	0.4	2.2	4703	0.0	1.5	134
	2012	129.2	141	0.0	0.5	1.9	5382	0.0	1.7	133
	2013	130.5	127	0.4	0.4	1.8	4760	0.0	1.4	25

Measuring methane emissions from a target landfill is the aim of this methodology. However, multiple landfills might be collocated in the same AIRS cell, which is the case for the Turkey Run landfill. These landfills are pinned in Fig. 2. Methane emissions from these landfills are listed in Table 3 based on the information from the USEPA website (http://ghgdata.epa.gov/ghgp/main.do). Further the days with northerly wind were ignored as explained later. Thus, all the landfills that are located at the top of cell 5 play negligible roles in estimating methane emissions from Turkey Run landfill.

Besides, since daily methane emissions from landfills vary due to barometric pressure variations (Xu et al., 2014), it is assumed that daily emission variations from landfills that are located close to each other have similar patterns. This assumption originates from the fact that barometric pressure is usually uniform over a mesoscale area, such as the length of an AIRS cell. Pressure gradients tend to exist on the synoptic-scale over hundreds of kilometers. Therefore, it is assumed that barometric pressure variations bring about equal percentages in methane emissions from landfills. In other words, any variation in concentration difference between the centers of two AIRS cells can still be associated with Turkey Run landfill emissions.

Table 3- Annual methane emissions of landfills around Turkey Run landfill (metric kilotons CO2e)

Landfill	State	CH4 emission
Salem	AL	82.5
Robert Roads	GA	5.4
City of La Grange	GA	48
Taylor County	GA	73
Houston county	GA	49
Clayton County	GA	44
Crisp county	GA	40
Dougherty county	GA	67
Wolf creek	GA	168

One concern about the proposed methodology is temporal variations of other emission sources (E_{non-LF} in equation 1) regardless of the amount of methane emitted by these sources. It is assumed that other sources, including different plants, livestock and industries, have almost invariant temporal emissions. For instance, methane emissions from different groups of livestock consisting of 70 sheep had less than 5% emission variations during a three day period (Lassey et al. 2011). For annual emission variations, methane emissions from non-dairy livestock varied about 3% between 1995 and 2003 (Zhou et al., 2007). However, methane emissions from these sources depend on season due to strong temperature variations. In order to minimize the effects of these sources, the data is categorized seasonally. These investigations reveal that livestock can be considered a steady methane emission source in seasons. Moreover, the most of livestock operations nearby the landfill are poultry, which are likely to manage manure dry instead of anaerobic lagoons. Thus, the livestock operations should not be a significant concern in this methodology.

We assume that the only other source that might have noticeable temporal variations is megacities. Noticeable variations of these emission sources compared to solid waste systems (E_{LF}) can lead to violations in the current methodology. On the other hand, seasonal variations for urban areas have been acknowledged in the literature (Zhang et al., 2016). In order to diminish the effect of these temporal variations, seasonal data is grouped together to find correlated linear equations. It means that separating summer data from winter data removes the majority of methane emission variations from urban areas. Another way to keep the dataset away from impacts of megacities is to neglect the days when wind mixes the methane emitted from the landfill with that from other megacities. For Turkey Run landfill, the adjacent megacity, Atlanta, is located north of landfill. To remove the roles of Atlanta from this methodology, the days when northerly winds blew were discarded. The northerly wind can carry methane from this city, since the AIRS data indicates the methane concentrations at the center of the cell and Atlanta is located above the cell. Otherwise, it can be assumed that the measured methane is not related to this city.

Based on the advective mass transfer, whenever there are emissions and noticeable flows (high Peclet number), methane plumes are advected along the wind directions from an adjacent upwind cell toward the landfill cell. On the other hand, due to different topography around the landfill, wind direction impacts on gas emission are anisotropic (Xu et al. 2014). Thus, the daily data should be categorized to study correlations between the AIRS data and field emission measurements separately for each wind direction. It is assumed that the area swept by centerline of each two cells (the landfill and the adjacent upwind cells chosen based on wind directions) with $\pm 45^o$ is related to that upwind cell. Therefore, the landfill cell and the upwind cell were taken to estimate methane emissions.

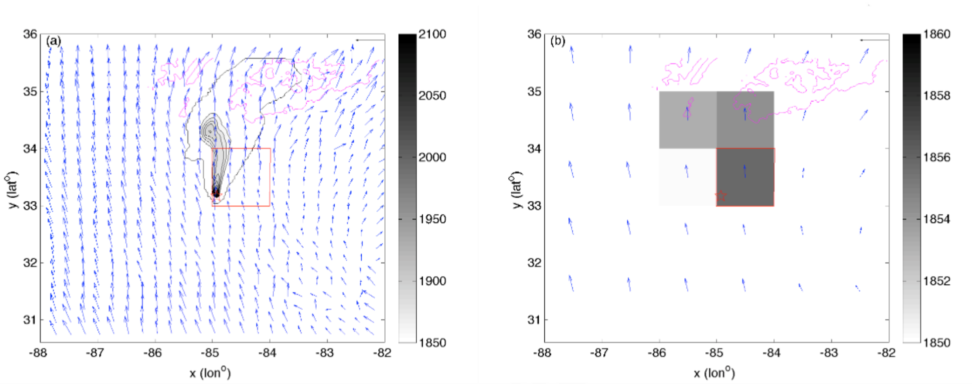
2-5 Atmospheric numerical simulation

The Advanced Regional Prediction System (ARPS) was used to simulate methane transport processes over Turkey Run landfill. The 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 (MPI). Descriptions of the model can be found in Xue et al. (2000) and Xue et al. (2001) with relevant details summarized below.

To resolve the local flow around the landfill, a multi-scale approach was adopted. Simulations were first performed on a 576 km by 576 km horizontal domain centered at Turkey Run landfill (-84.9146^o W, 33.1751^o N). Realistic initial and lateral boundary conditions were obtained from the meteorological analysis data that was produced by the North American Mesoscale Forecast System (Rogers et al. 2009). The lateral boundary conditions were updated every 6 hours, and linearly interpolated in-between. The horizontal resolution of the model grid was 2400 m, and the average vertical resolution was 320m. Hyperbolic grid stretching was adopted in the vertical direction to create 50 m spacing near the surface. This was done to improve the resolution of the atmospheric boundary layer. The number of grid points used were (243, 243, 53) in (x, y, z) directions. The ARPS model was run in mesoscale mode with a TKE-1.5 turbulence closure with the planetary boundary layer parameterizations of Sun and Chang (1986). Since 2400m spacing was generally considered a convection-resolving scale, cumulus parameterization was turned off. The Lin ice scheme (Lin, et al. 1983) was adopted to parameterize microphysical processes.

The simulation started from 1200 UTC (0700 LST), April 9, and ended at 0000 UTC (1900 LST), April 13, 2013. During the simulations, a conservative tracer was continuously emitted at a constant rate of 910 ($\frac{g_{CH_4}}{min}$) from a single surface grid point at the center of the model domain to mimic methane emissions from landfills. The emission rate is taken from the average annual methane emission estimate of the landfill from the USEPA. No other sources of methane were considered in this simulation. The first 6 hours of simulation results were excluded from the discussion to avoid any numerical artifacts from the model spin-up. The synoptic wind direction was southerly for the first 72 hours of the simulation. It shifted gradually to northerly at 84 hours, and remained so afterwards. Since northerly wind directions were not considered in this work to avoid interference due to emissions from Atlanta (see discussion in Section 2-5), simulations results after 84 hours were excluded in the following discussion. Results were presented from 1800 UTC (1300 LST), April 9 to 1200 UTC (0700 LST) April 12, 2013 for a total of 66 hours. Simulation results were output at every half hour for analysis. During this period, the atmospheric stability levels (1.5-6.5) and turbulence intensity indexes (0.08-10.16) were measured at 2 meter above the ground, which renders this period appropriate to assess the validity of the assumptions under different conditions. Since this period has some wide variations of atmospheric parameters, if the assumptions achieve validation here, they can be generalized for other periods.

A snapshot of the column integrated concentration $\int_{z=0}^{H} cdz$ (ppm m) along with surface wind vectors is presented in Fig. 3 to offer a direct visualization of the simulation that would be seen by AIRS. A background concentration of 1.85 *ppm* measured during the field campaign is imposed. The background methane is assumed to be uniform in the horizontal directions and the vertical direction up to the atmospheric boundary layer depth (Z_i). A characteristic Z_i of 1 km is chosen for the estimate (Stull 1988). Therefore, the background column integrated concentration is estimated to be 1850 ppm m. Note that the terrain is mostly flat in this area with some moderate topography to the northeast of the site.



(a) (b)

Figure 3. Contours of column integrated tracer concentrations $\int_{z=0}^{H} c dz$ (*ppm m*) at 1530 LST, April 9, 2013. H is 16 km at the top of the model domain. (a) represents the simulation results at the model grid spacing. (b) represents the same results as (a), but up-scaled to 1° x 1° resolution as would be seen by the AIRS, notice the difference in the color scale. The contour interval is set to 10 (*ppm m*) in (a). The surface wind vectors are represented by blue arrows. The top right horizontal arrow represents 10 m s^{-1}. Terrain height is represented by magenta colored contours. The contour interval is 500 m. The location of the Turkey Run landfill is marked by a red star in the center of the domain. The red box marks a 1° x 1° cell that represents cell 5 in Fig. 2.

3- Results and Discussion

3-1 Atmospheric simulation results

3-1-1 Assumption #1: Non- accumulative AIRS cells

When deriving the simple model (see Fig.1), it was assumed that methane is not accumulated in the AIRS landfill cell, i.e.

$$\frac{\partial}{\partial t}\int_x \int_y \int_z c_1(x,y,z,t)dxdydz \approx 0$$

The volume-integrated concentration $\int_x \int_y \int_z c_1(x,y,z,t)dxdydz$ is computed for the landfill cell (see Fig. 1 and Fig. 3), with its NE corner at (-84°W, 34°N), and its SW corner at (-85°W, 33°N). Fig. 4 presents the areal-mean column integrated methane concentration $\frac{1}{L_x L_y}\int_{L_y}\int_{L_x}\int_{z=0}^{H} cdzdxdy$ over cell 5 from 0000 UTC, April 10 to 0000 UTC, April 13, where L_x and L_y are the zonal and meridional dimensions of cell 5. The background column-integrated concentration is assumed to be 1850 *ppm m*, as explained in the previous section. Overall, the standard deviation of the areal-mean column integrated concentration is 1.39 *ppm m*, while the time-mean is 1853.80 *ppm m*. The resulting coefficient of variation is 7.48×10^{-4}, indicating small temporal variations due to the LME from Turkey Run upon the relatively large background concentration.

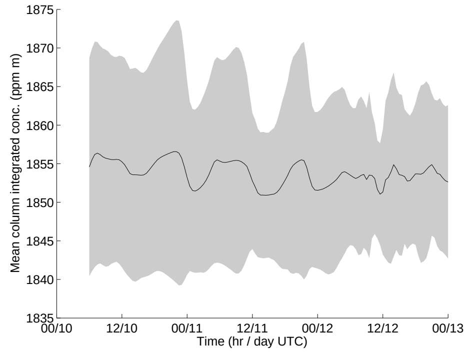


Fig. 4. Time series of areal-mean column integrated methane concentration over cell 5 in Fig. 2. Shading represents the standard deviations of column integrated methane concentration within cell 5. The background concentration is assumed to be 1.85 ppm, and vertically uniform over the atmospheric boundary layer (assumed to be 1km deep), and 0 above the boundary layer.

Accumulation of methane in this AIRS cell depends upon atmospheric stability and turbulence conditions. For instance, during April 11 and 12 (days 2 and 3 in Fig. 4), when the atmosphere was more unstable and turbulent, lower tracer gas concentration variations were found. Overall, the time series of normalized gas concentrations is almost stationary which implies that mass accumulation does not change more than one order of magnitude in an AIRS cell. As noted earlier, this methodology is supposed to provide an estimate of the LME on or within the same order of magnitude (e.g. estimate is roughly within 1/10 and 10x the actual value) for the days without field measurements in order to give a better overall annual LME estimation. Thus, variations of normalized gas concentration in a fraction less than <1% supports this assumption that the unsteadiness of methane concentration is not noticeable at the scale of the AIRS cells.

$$\frac{\partial}{\partial t}\int_x \int_y \int_z c_1(x,y,z,t)dxdydz \approx 0 \int_x \int_y \int_z c_1(x,y,z,t)dxdydz$$

3.1.2 Assumption #2: Uniform wind distribution in horizontal planes

Wind is also assumed to be fairly uniform over the entire cell, i.e. within the cell $u(x, y, z, t) \approx u(z, t)$. This assumption allows moving the wind vectors outside the horizontal integral in the Eq. 3. In the simulation, cell 5 where the landfill is located has 39x47 grids covering the 1° by 1° cell. This assumption considers a uniform distribution of horizontal wind speed and direction among these grids. The spatial average as well as the standard deviations of wind speed and direction in these grids near the surface (25 m AGL) is shown in Fig. 5. Due to variations in topography and roughness elements, surface winds are usually the most variable within the atmospheric boundary layer. Especially during the daytime, the strongest variations in the horizontal winds are mostly observed near the surface, where the vertical wind shear is strong and shear production of turbulence kinetic energy is the most vigorous (Kaimal and Finnigan 1994). Horizontal wind speed variations usually decrease with height within the boundary over a relatively homogeneous topography, in the absence of mesoscale weather events or synoptic fronts. Both these conditions are met for the Turkey Run landfill site during April 10 -13. Therefore, the relative variations of horizontal winds at higher elevations were smaller and not presented.

In general, the relative standard deviations (coefficient of variations) in both wind speed and direction are small for April 10 and 11, when the atmosphere condition was calm, and the daytime convective mixing was strong. On April 12, a synoptic front came by and changed wind direction from southerly to northerly over the course of a day. The wind speeds also increased significantly from April 12 to April 13. Even then, the highest relative standard deviation for both wind speed and direction were less than 20%, which means that the horizontal winds were relatively uniform across the scale of the AIRS cell during the passage of a front. Therefore, from the simulation results, it seems reasonable to postulate uniform horizontal wind speed and direction over the AIRS cell.

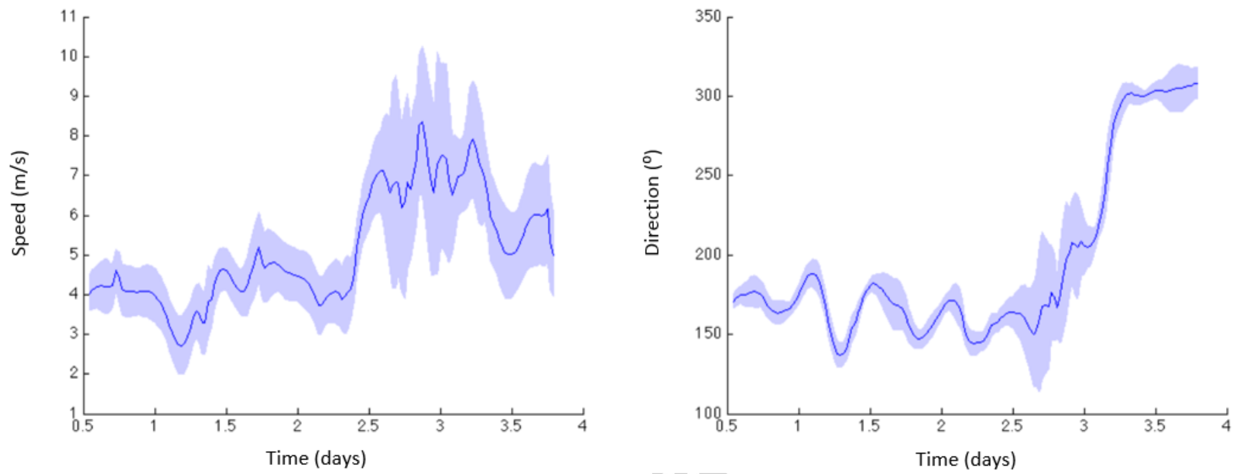


Figure 5. Time series of wind speed (left) and wind direction (right) at 25 m AGL. Solid lines represent spatial average over entire the cell, the shading areas represent one standard deviation of the spatial variations over the cell.

3.1.3 Assumption #3: Completely stirred cell

When deriving the mass balance equation in Section 2, it is assumed that methane concentrations are well mixed within the AIRS cell. This assumption allows the use of methane concentration at the center of the cell to prescribe the effluent concentration at the outflow boundary. To test the validity of this assumption, the standard deviation of the column integrated methane concentration within cell 5 is computed. The shading area in Fig. 4 represents one standard deviation from the cell-averaged column integrated methane concentration. For the cell to be considered well-mixed, the coefficient of variations must be much smaller than unity.

Given the relatively high background methane concentration, this ration is about 1%. This suggests a relatively narrow (i.e. concentrated) distribution of methane within cell 5, lending some support to this assumption.

3-2 Field methane emission measurements

Average of the daily LME measurements after applying filtering criteria were taken as daily field emission values. Turkey Run landfill is relatively young (opened in Dec. 2009) and is still accepting waste. Thus, the incoming waste stream leads to an increase in annual LME. Daily LME vary from (274-1820) g min^{-1} with an average of 910 g min^{-1}. Although an increase in LME is observed for this landfill, most of the old landfills behave in different manners. Aged landfills usually have seasonal emission fluctuations which are different compared to this landfill. This ascending trend in methane emissions for Turkey Run landfill is associated with its age and the rate of waste disposal (Foster-Wittig et al. 2015).

3-3 Correlations between field methane emission estimation and AIRS data

Overall, a dataset that consists of field measurements in 19 days is available. Some of the days were removed after applying filtering criteria. For some other days, the AIRS data was not available because of cloudy sky conditions. Finally, a dataset that contains 13 days was selected to study correlations between the satellite and the ground measured data. The dataset was divided into different groups according to wind directions. The DMVPC value of the landfill cell was subtracted from the DMVPC value of the adjacent upwind cell chosen based on wind directions. Then, this subtracted value was plugged into the methane mass balance (Eq. 4).

In order to develop a linear model, first, we aim to evaluate the model performance in methane emission predictions from the landfill was aimed. About 70% of dataset for each group was used to train a linear regression model and then the rest of dataset was used to examine the accuracy of the regression model. The relative error between the predicted emission and the measured emission varied between 0.06-0.3, which reveals the promising prospects of the obtained regression model.

The regressions between all the DMVPC data for each AIRS cell with measured methane emission are presented in Fig. 6. In all of these figures, x-axis is the difference between the DMVPC in two adjacent cells chosen by wind directions, and y-axis is the measured LME in fields after applying filtering criteria. High regression coefficients and correlations between measured LME and the DMVPC help predict emissions during unmeasured periods. The cell 3 has the best regression accuracy which might be related to the landfill location regarding this cell. This landfill is located at the line passing from the center line of cell 5 (where the landfill is located) and cell (3).

In addition to neighboring cells, one farther cell was considered to examine this methodology for a non-adjacent cell (Fig. 6d). Comparing the regression results for the neighbor cells (1, 2 and 3) and that off cell (4) demonstrates that: the farther cell to the landfill is selected, the inaccurate regression is found. This might be related to more dispersion, longer methane travel time to reach the cell 5 from the farther cells, and plume (wind) pathway variations on the way from the farther cells to get the landfill cell. In addition, the probability of the presence of unknown sources is increased in farther cells that affects the emission estimation procedure. This result proves that a meaningful regression between the landfill cell and the adjacent cells is supported by the mass transfer concept. If any good result was found by chance, it would have reoccurred for a farther cell, or one adjacent cell might have had much weaker results than farther cells. However, all neighboring cells that shows high correlation coefficients between the field LME and DMVPC, while this conclusion is weaker for a farther cell to the landfill.

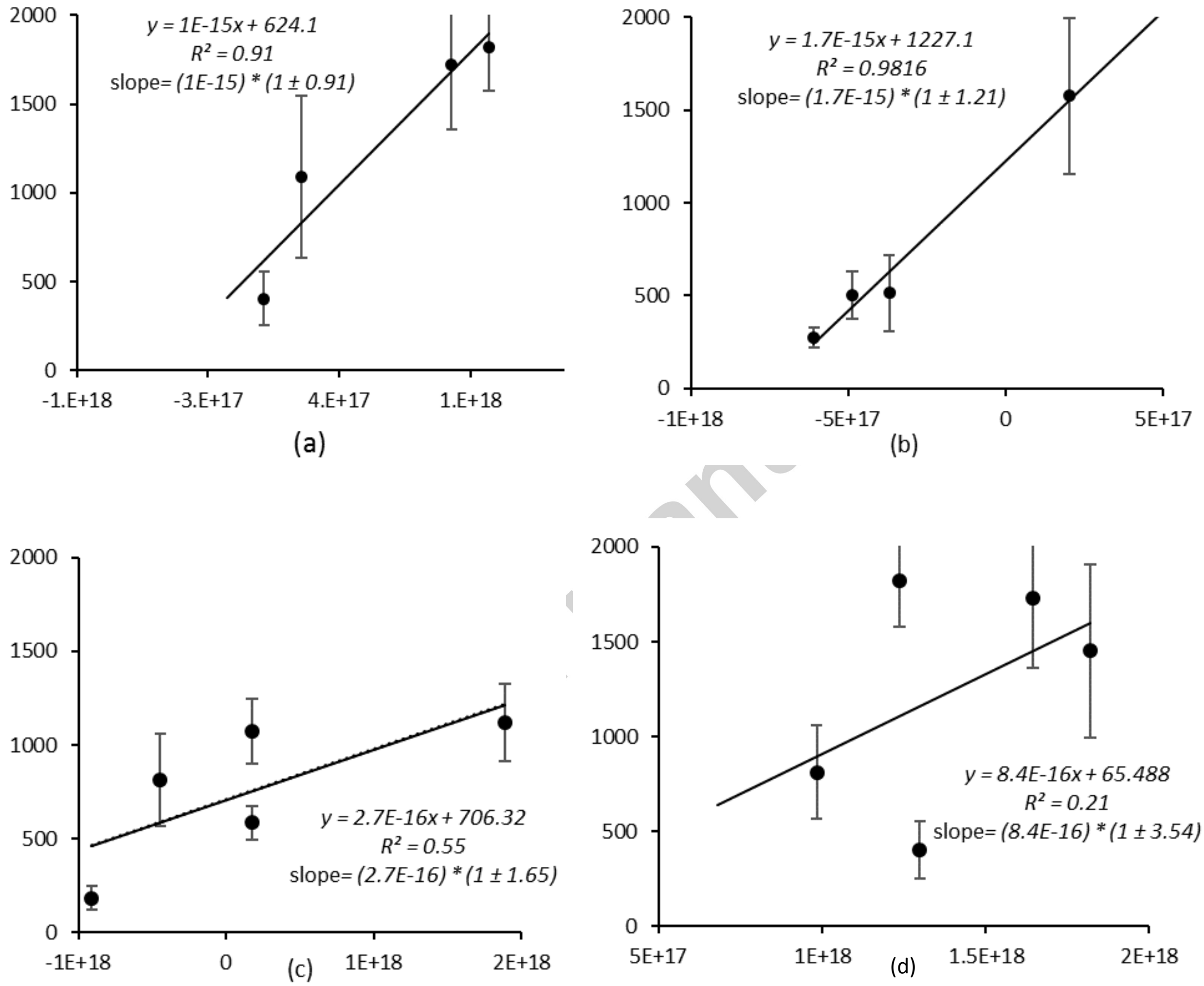


Figure 6- E_{LF} in Y-axis (g/min) vs. $\left(\int_0^H c_1\,dz - \int_0^H c_0\,dz\right)$ for DMVPC data in X-axis (mol/cm²), if wind comes from: a) cell 1, b) cell 2, c) cell 3, d) cell 4. Value of 95 % Confidence Interval range for the slopes are presented in the legends.

In order to assess the performance of this methodology for NMVPC, the same procedure was followed and the results are shown in Fig. 7. NMVPC was recorded at 1:30 a.m. which has about 12 hours time lag as opposed to the field measurements. Besides, it is well-known that there are significant differences between daily and nightly methane emissions from landfills (Gebert et al. 2011; Xu et al. 2014). Therefore, since LME were measured during the day, nightly regressions with field measurements are much weaker compared to the daily AIRS data for cells adjacent to the landfill cell. Although a weaker performance of NMVPC was obtained compared to the daily analysis, cell 2 shows an acceptable performance, which can be related to that leverage point in this figure.

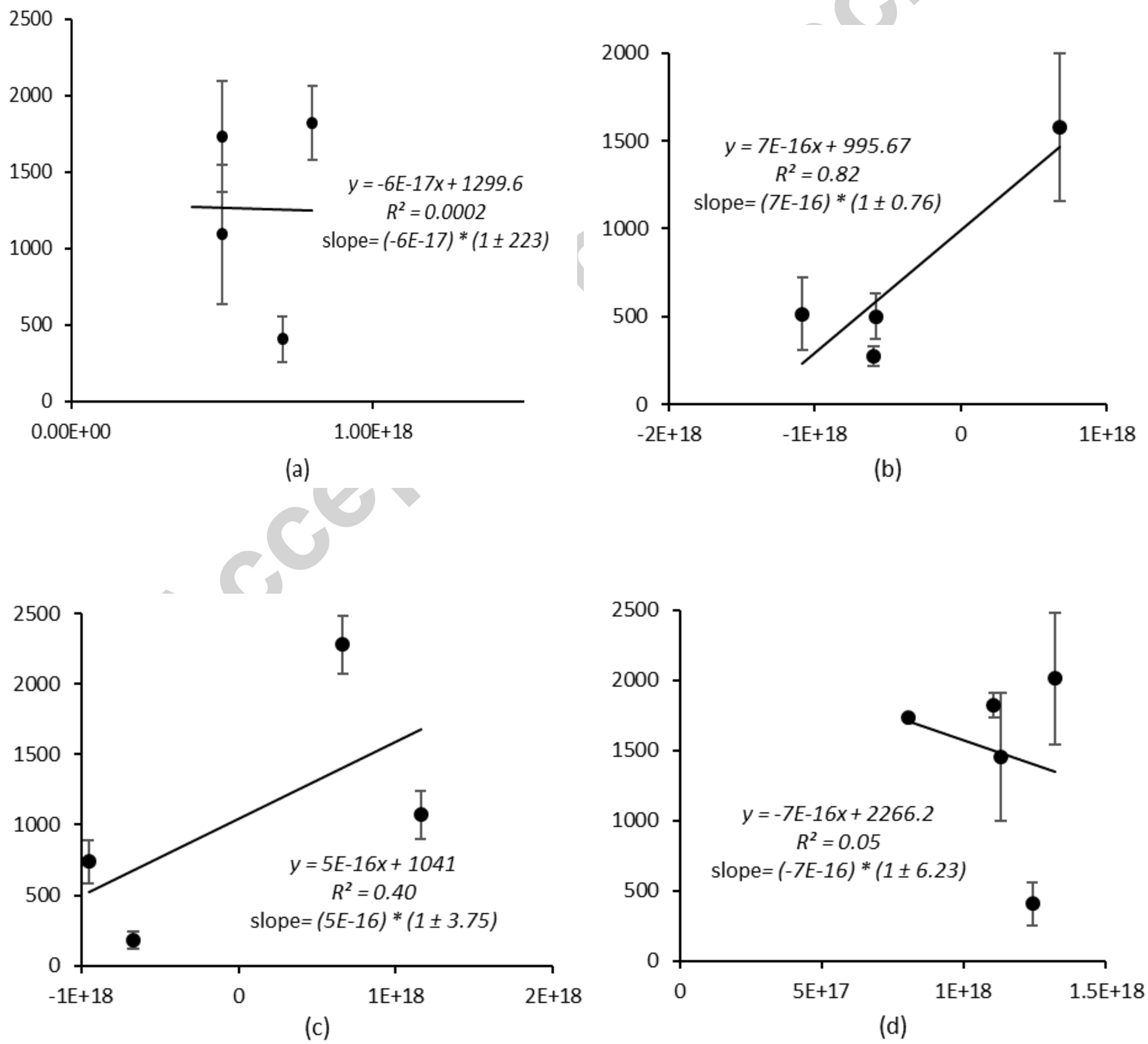


Figure 7- E_{LF} in Y-axis (g/min) vs. $\left(\int_0^H c_1 dz - \int_0^H c_0 dz\right)$ for NMVPC data in X-axis (mol/cm²), if wind blows from: a) cell 1, b) cell 2, c) cell 3, d) cell 4. Value of 95 % Confidence Interval range for the slopes are presented in the legends.

3-4 Comparing two approaches for annual estimations of LME

Here, two different approaches in estimating annual LME are compared. The common method assumes a linear relation between any two consecutive emission measurements for days without any field measurement (Mønster et al. 2015). This approach was used to estimate methane emission for this measurement period. It dismisses methane emission variations during the time gaps which can be in the order of month or shorter. Such dismissal can cause remarkable deviations in estimation of the actual LME. During the second approach, which is proposed in this paper, the AIRS satellite data and field measurements were employed first to train a set of simple regression equations. The AIRS data for unmeasured days were plugged into the trained regression models to estimate LME. This approach estimates methane emissions according to the AIRS data that captures emission fluctuations. It is expected to outperform the traditional stepwise linear emission interpolation approach.

3-4-1 Interpolating discrete tracer dilution method data

Measured LME after applying the filtering criteria were combined to give monthly-average emissions. The linear piecewise interpolation was applied between each two consecutive months to interpolate emissions between these two measurements. The trapezoidal integral calculation method was utilized to calculate the LME during measurement days. Estimated total landfill methane mass between April 2012 and May 2013 using this approach is about 556 (Mg). This approach might not be accurate for estimating annual LME, because it assumes that LME vary linearly during the period between two field campaigns (which is around one month in this case). Temporal methane emission variations have been discussed in detail (Czepiel et al. 2003; Gebert et al. 2011; Xu et al. 2014), ignoring these emission fluctuations may give significant errors.

3-4-2 Coupling AIRS and discrete TDM field data

The AIRS datasets are used to estimate LME during the unmeasured periods. Wind direction is needed to determine the adjacent upwind cell in order to apply AIRS data using the methods introduced in Section 2. Hourly wind direction was obtained from the national climate center website (http://www.ncdc.noaa.gov/isd/data-access) for stations located close to the landfill. Based on daily wind directions, the background upwind cells were determined. The appropriate linear regression models with respect to specific wind directions presented in Fig. 6 were used to estimate LME. The results are presented in Fig. 8. The AIRS measurements are instantaneous snapshots (around 1:30 p.m.) of vertically integrated concentrations. Since the nighttime linear models had poor results, those equations were not used to predict methane emissions. Such measurements might not be perfectly representative of the daily emissions. Averaging emission data from a day before and a day after the studied day makes the daily emission more realistic. Because the emission of a day before the studied day can indicate the emission before 1:30 p.m. of the studied day and the emission of a day after the studied day can represent the emission after 1:30 p.m. in that studied day. Further, this averaging smooths the emission curve and prevents abrupt instantaneous emission fluctuations.

The barometric pressure fluctuations were correlated against the predicted LME. LME have negative correlations with barometric pressure for a period of a couple of days (Czepiel et al. 2003; Gebert et al. 2011; Rachor et al. 2013; Xu et al. 2014). The emission fluctuations during this period would be associated with barometric pressure variations. Daily-averaged barometric pressure derived by a meteorological station around the landfill as well as the predicted LME are shown in Fig. 8. These negative correlations encircled in this figure are visible for several periods. It reveals that the LME are varying inversely as atmospheric pressure changes for periods over several days. While the LME estimated by the proposed methodology depict moderate negative correlations with barometric pressure (-0.54) for the period of August 1 - September 18, 2012, (-0.67) for the period of October 1 - November 8, and (-0.55) from December 21, 2012 until January 14, 2013, low or positive correlations between them are observed for a few periods as well. A plausible reason for these observations would be violating at least one of the assumptions made in this model. For instance, weakly negative or even positive correlations would be explained by weaker mixing during the colder days. Negative correlations between barometric pressure and LME give credence to this methodology.

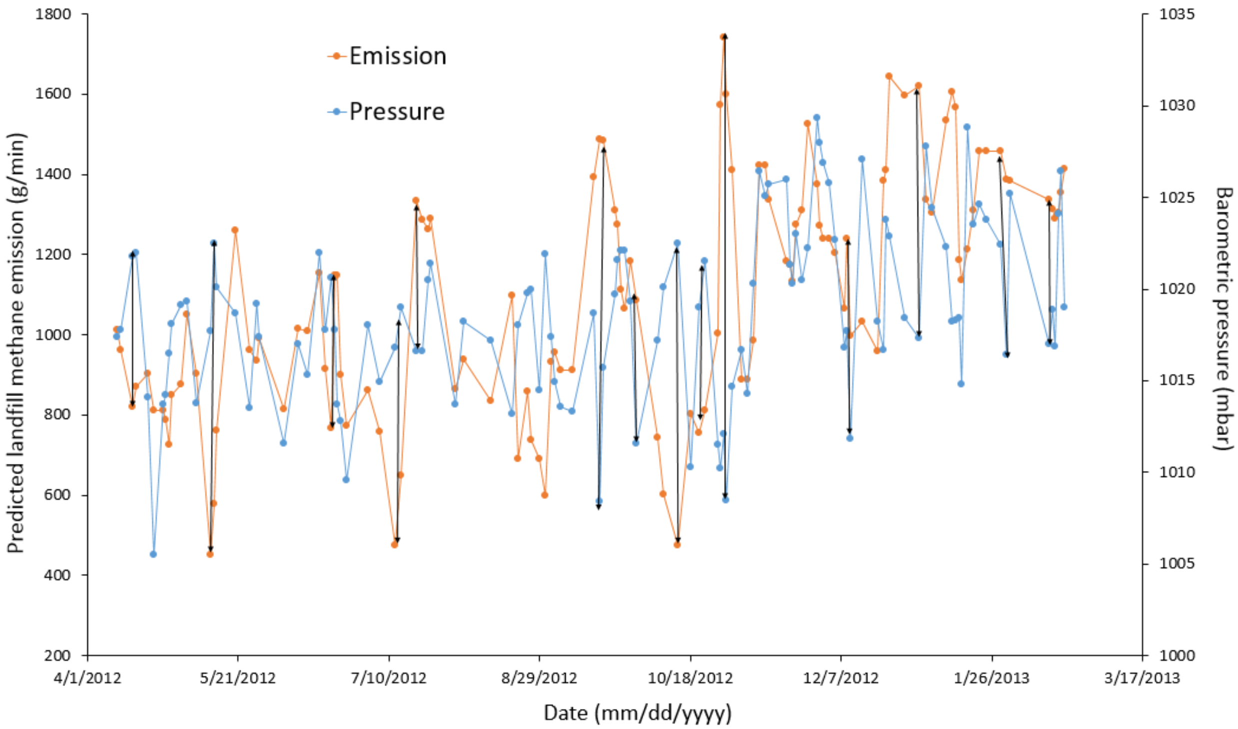


Figure 8- Estimated emission and barometric pressure variations. Encircled points representing corresponding emission and pressure variations.

Figure 9 shows the results of applying the standard and the proposed methodologies to estimate annual LME. Comparing plots in this figure demonstrates that the RS technique captures more methane emission fluctuations than the common TDM interpolation. Daily emission fluctuations have been well-documented (Czepiel et al. 2003; Xu et al. 2014); however, they are not observed using the TDM with gaps of several days (unmeasured days). Since the performance of the LME estimated by the AIRS was examined in section 3.3, it is concluded that the satellite derived methane emissions are in agreement in the order of magnitude with field measurements. The integrated mass of emission since April 2012 until May 2013 was estimated to be 627 (Mg) using the trapezoidal method. This result is about 13% higher than the normal TDM interpolation estimation.

The results discussed in this paper will provide an insight for environmental engineers/scientists and managers to plan for controlling landfill gas emissions with higher confidence. LME prediction using the AIRS data has some deviations compared to field results; however, the TDM's uncertainties have been reported as well (Mønster et al. 2014). Thus, there is no accurate data to meticulously examine the AIRS performance in the LME estimations. If it is presumed that the TDM results equal the actual LME, the AIRS approach has a 22% normalized root mean square error.

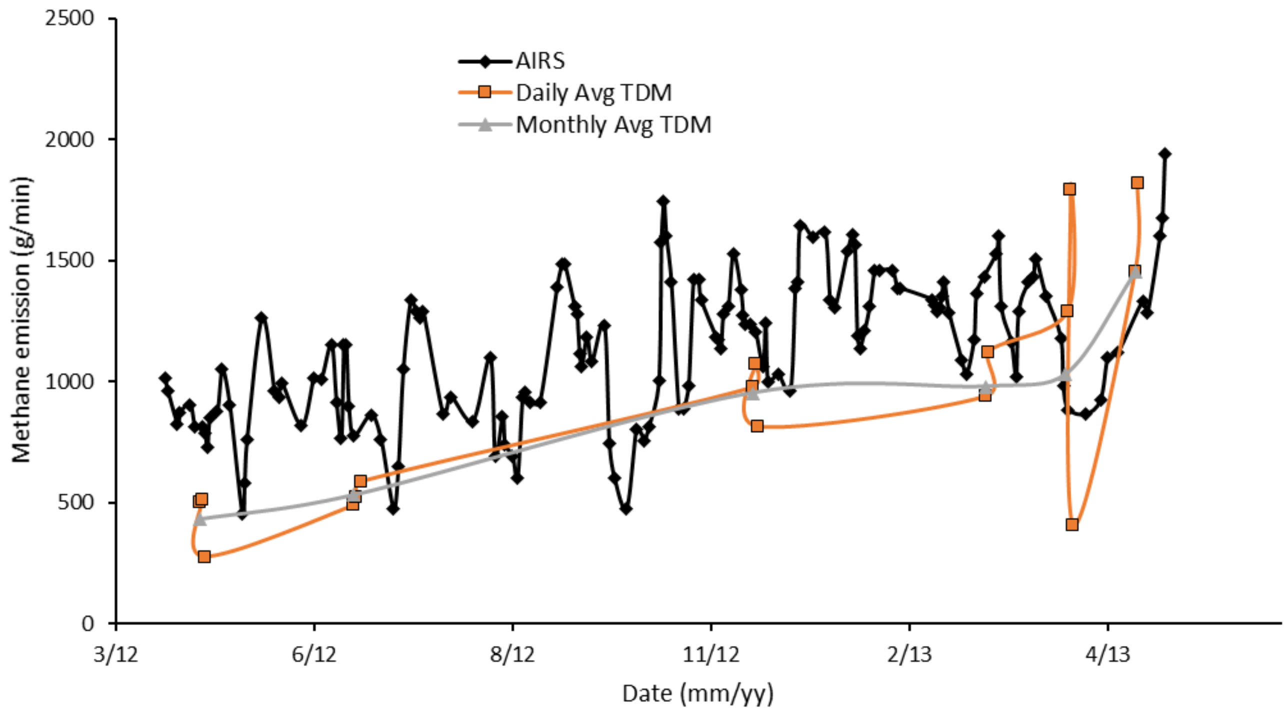


Figure 9. Comparing coupled AIRS-TDM methane emission estimations with that from TDM measurements alone.

4- Recommendations and limitations

Several methods have been developed to quantify LME; however, there is no single perfect method to continuously measure methane emissions from landfills yet. Each of the methods has its own limitations. The TDM, which is the most popular method, is applicable in measuring methane emissions from landfill areas with any size. Unfortunately, it can be used just for few hours and might underestimate/overestimate other unmeasured times (Delkash et al. 2016). On the other hand, one TDM campaign costs about US $30,000 (Oonk 2010), which restricts measurement durations. Further, this method needs trained operators to use the instruments. Therefore, a new inexpensive approach that estimates emissions more frequently is needed to control greenhouse gas emissions more efficiently.

Although satellite measurements may be limited due to cloudy conditions, satellite data that represents LME is easy to access and use for daily emission estimations. There are some satellites that are able to sense atmospheric methane columns such as AIRS (Pagano et al. 2003), CarbonSat (Bovensmann et al. 2010), GOSAT (Schepers et al. 2012) and SCIAMACHY (Bovensmann et al. 1999). These satellites have different spatial and temporal resolutions, which render them useful for certain applications. For instance, GOSAT, CarbonSat, and SCIAMACHY have 3, 5, and 6 days as revisiting times (orbiting times), respectively. It means that it takes three days for the GOSAT satellite to reach a certain point on the earth twice. It is the best to use field measurements with relatively long durations (3 to 5 days), because the corresponding satellite data, which have higher spatial accuracy, can be employed. In other words, satellites with greater orbiting time have a finer resolution (higher spatial accuracy). Under this higher spatial accuracy, other significant non-landfill emission sources will be taken out of the landfill cell. This leads to enhancing the accuracy of the proposed technique. Since there are consecutive daily measurements for this landfill, a satellite with less spatial accuracy has to be chosen (larger cell size) to have more satellite data corresponding with field measurements for regression analysis between the field and satellite data. Due to the available field data and using the AIRS data, some assumptions were made to interpret landfill methane emission using the AIRS larger cell size, which might bring about some uncertainties in LME estimations. Some assumptions considered in the present study are easy to adopt in the model. This model provides continuous daily methane emissions from landfills. These assumptions are made because of the large footprint of AIRS cells compared to the landfill size. An atmospheric model was employed to evaluate the accuracy of the considered assumptions. The proposed atmospheric model indicates these assumptions are likely valid most of the time; however, assuming fully mixed reactor might be challenging. This assumption is more viable under an unstable atmosphere. This might restrict the application of the proposed method during winter when the atmosphere is mostly stable.

An issue to consider, which stems from large RS images, is the presence of some other significant methane sources in the landfill AIRS cell. Landfills are usually located far from urban areas; however, Atlanta is located in the same cell with the landfill. Even though some measures were taken to overcome this problem such as discarding the data with southerly wind directions, and considering the measured data with similar weather conditions in a group to minimize emission variations of other sources, still some uncertainties might affect the LME estimations. The authors believe that there are many landfills located far enough from urban areas or other sources that make the proposed methodology more applicable.

Since the assumptions that were made are general and common, this technique is applicable for other landfills. LME can be predicted by this method under most conditions that make this approach promising. This paper is a pioneer in developing a more inexpensive and easier technique to estimate annual LME. It only requires a few ground field measurements to establish the regression models for this purpose.

The proposed method would be a good start for thinking about a new approach. Training the RS data with field measurements would predict LME reasonably well. This would lead to saving significant amounts of money and better landfill managements in order to control greenhouse gas emissions into the atmosphere, which is one of the sources for the climatic change and global warming.

5- Broad Impacts

Emissions of methane are of particular concern. Methane is the major component of natural gas and a powerful greenhouse gas. A recent modeling study indicates that methane has an even greater global warming potential than previously believed, when the indirect effects of methane on atmospheric aerosols are considered (Shindell et al. 2009). Because large of amounts of methane emissions are linked to society's fundamental needs for food and energy, they will continue to increase and further warm the climate unless substantial efforts are undertaken to reduce them worldwide (Denman et al., 2007). Increasing methane emission to the atmosphere endangers water and food security (Wheeler and von Braun 2013), and can lead to intensified drought and flooding (Dai, 2011).

As global warming continues, the limited capabilities in developing countries will become an increasingly important issue in global efforts to mitigate the negative impact of climate change. Thus urgent actions are needed to control methane emissions that causes climate change. Based on USEPA, landfills are known as an important anthropogenic methane source

(18%), therefore, controlling the gas emissions would help in mitigating greenhouse gas emission (http://www3.epa.gov/climatechange/ ghgemissions/gases/ch4.html).

On the other hand, population growth and developing industries increase energy demand. Methane produced during landfilling processes is known as promising energy source. Perfect landfill management not only can decrease greenhouse gas emission, but it also enhances energy recovery efficiency and supplies some energy portion needed (Themelis and Ulloa 2007). Thus, the recovery of landfill gases for use as an energy resource has become the center of interest since it solves both environmental pollution and energy shortage. Accurate emission understating assists landfill managers to evaluate gas collection system efficiency to retrieve methane more efficiently. On the other hand, more precise knowledge about surface emission illuminates topsoil methane oxidation performance and paves the way for promoting methane oxidation capacity.

Applying any new action on landfill operation system needs reliable data about annual landfill emission. On the other hand, methane emission measurements from the landfill is costly and laborious, therefore short period campaign measurements usually are carried out and the observations are extrapolated to estimate annual emission. This short-term emission fluctuation can lead to significant errors in annual landfill emission inventories. Here it was aimed that how emission variations could be predicted using wind and satellite data to produce continuous emission data. This would be a significant step in climate change studies. The authors believe that applying this regression model can improve the accuracy of the annual emissions and mitigate greenhouse gas emission efficiently.

6- Conclusion

Application of a satellite (AIRS) remote sensing technique in monitoring and estimating methane emissions from a landfill is presented. Three assumptions were made to develop this method and numerical simulations were utilized to examine the validity of these assumptions. Several methane emission measurements of 13 days were carried out for Turkey Run landfill, USA, whose area is about 20 acres. Two total DMVPC and NMVPC were obtained using the AIRS data to correlate with field measurements. Since field measurements were carried out during the daytime (9a.m. -5 p.m.), the DMVPC shows a better correlation with field measurements compared to the NMVPC. The results depicted higher errors in LME estimations for a farther cell than adjacent cells to the landfill. Annual LME was estimated using both the linear interpolation of the TDM measurements as well as training a linear regression model using the TDM coupled with the satellite data to predict LME for unmeasured days. The results clearly showed that AIRS data predict daily variations that are neglected during the TDM interpolation. In view of the negative correlation between the LME predicted by the proposed method and the barometric pressure, we can conclude that this methodology estimates LME reasonably well.

7- Acknowledgements

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HIGHLIGHTS

- This paper examines performances of remote sensing in landfill methane emission
- AIRS remote sensing data was taken for estimating landfill methane emission
- Remote sensing technique was validated with field measurements
- ARPS model was employed to evaluate the assumptions that were made to develop the model
- The predicted emission is in order of magnitude with field measurements