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Impact of changes in barometric pressure on landfill methane emission

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Key Points:
- Landfill methane emissions strongly depend on changes in barometric pressure
- Current methods lead to uncertainty in estimation of annual landfill emission
- Continuous measurement is needed to quantify annual total landfill emission

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Abstract Landfill methane emissions were measured continuously using the eddy covariance method from June to December 2010. The study site was located at the Bluff Road Landfill in Lincoln, Nebraska, USA. Our results show that landfill methane emissions strongly depended on changes in barometric pressure; rising barometric pressure suppressed the emission, while falling barometric pressure enhanced the emission, a phenomenon called barometric pumping. There was up to a 35-fold variation in day-to-day methane emissions due to changes in barometric pressure. Wavelet coherence analysis revealed a strong spectral coherency between variations of barometric pressure and methane emission at periodicities ranging from 1 day to 8 days. Power spectrum and ogive analysis showed that at least 10 days of continuous measurements was needed in order to capture 90% of the total variance in the methane emission time series at our landfill site. From our results, it is clear that point-in-time measurements taken at monthly or longer time intervals using techniques such as the trace plume method, the mass balance method, or the closed-chamber method will be subject to large variations in measured emission rates because of the barometric pumping phenomenon. Estimates of long-term integrated methane emissions from landfills based on such measurements could yield uncertainties, ranging from 28.8% underestimation to 32.3% overestimation. Our results demonstrate a need for continuous measurements to quantify annual total landfill emissions. This conclusion may apply to the study of methane emissions from wetlands, peatlands, lakes, and other environmental contexts where emissions are from porous media or ebullition. Other implications from the present study for hazard gas monitoring programs are also discussed.

1. Introduction

Methane plays a critical role in the radiation balance and chemistry of the atmosphere. According to the most recently published Intergovernmental Panel on Climate Change Fourth Assessment Report [*Intergovernmental Panel on Climate Change* (*IPCC*), 2007] and other studies [e.g., *Rohde,* 1990; *Hansen and Sato*, 2001], methane is the second most important greenhouse gas after carbon dioxide and currently contributes 18% of the radiative forcing by long-lived greenhouse gases [*Dlugokencky et al.*, 2011]. The global average atmospheric methane concentration has increased steadily from a preindustrial level of about 715 ppb [*Etheridge et al.*, 1998] to about 1794 ppb in 2009 [*Dlugokencky et al.*, 2011]. In addition to climate forcing, the increase in atmospheric methane has contributed approximately half of the estimated increase in global tropospheric ozone [*Ehhalt and Prather*, 2001], which has a negative impact on the health of living organisms.

Atmospheric CH_4 concentration results from a balance between CH_4 sources and sinks. The major anthropogenic methane sources are anaerobic production from landfills, ruminant animals and their waste, release from mining operations, oil and natural gas production and distribution, and combustion of biomass [*Evans*, 2007]. A major portion of the methane from natural sources is thought to originate from biological processes in anoxic conditions, e.g., from peatlands, wetlands, and rice paddies. Globally, landfill methane emission contributes about 10–19% of the anthropogenic methane burden into the atmosphere [*Doorn and Barlaz*, 1995; *Bogner and Matthews*, 2003; *IPCC*, 2007]. In the United States, as much as 37% of annual anthropogenic CH_4 emissions come from landfills [*IPCC*, 2001; *Czepiel et al.*, 2003], which represent the largest source of anthropogenic methane emissions [*Bogner and Matthews*, 2003]. To have a good estimation of total landfill emission would require a strategy of sampling and statistics based on field measurement data [*Bogner et al.*, 1997; *Bogner and Matthews*, 2003]. Published field measurements of landfill methane emissions show several orders of magnitude in temporal and spatial variations [*Bogner et al.*, 1997; *Schroth et al.,* 2012], which makes estimation of total methane emissions difficult.

Quantitative understanding of methane emissions from landfills and how environmental variables control the emissions is essential for (1) process-based modeling studies, (2) determining a mitigation strategy, and (3) formulating controls and regulations by governments. Four methods that are commonly used to quantify landfill methane emissions are inventory, chamber-based, trace plume, and mass balance. The inventory method is based on the estimated production rate, the estimated rate of methane oxidation, and the amount of methane recovered [*Bingemer and Crutzen*, 1987]. The production rate normally is based on the rate of decay of the various biodegradable components of solid wastes, such as paper, food scraps, yard trimmings, and old furniture. This approach often leads to large uncertainties because of inaccuracies in the input data and the uncertainties in rate estimations [*Bogner and Matthews*, 2003]. The chamber-based technique estimates methane emission based on the rate of change in CH_4 concentration inside a chamber, the chamber volume, and the soil surface area covered by the chamber. This technique is based on a relatively simple theory, is easy to perform in the field, and has few constraints of topography at the measurement site. It is, however, labor intensive, and the results may be a poor representation of average surface emission due to the high spatial variability of methane emissions at a landfill [*Czepiel et al.*, 1996b; *Bogner et al.*, 1997; *Giani et al.*, 2002].

To address deficiencies of the chamber-based method, some large-scale methods have been used to measure landfill gas emissions, including the trace plume method [*Czepiel et al.*, 1996b; *Galle et al.*, 2001; *Oonk*, 2010] and the mass balance method [*Hashmonay and Yost*, 1999; *Thoma et al.*, 2005; *Oonk*, 2010]. For the trace plume method, a tracer is released from the landfill to the atmosphere at a known rate (F_t). Further downwind, the concentrations of the tracer and methane are measured, and the methane emission from the landfill (F_{CH4}) can be estimated from

$$F_{CH4} = F_t \frac{[CH_4]}{[\text{Tracer}]} \tag{1}$$

where $[CH_4]$ is the methane concentration above the atmospheric background, [Tracer] is the tracer concentration above the atmospheric background, and F_t is the rate at which the tracer is released at the landfill. Sulfur hexafluoride (SF_6) has often been used as a tracer for this method [*Czepiel et al.*, 1996b, 2003] because of its low atmospheric concentration and the availability of suitable gas analyzers. The mass balance method estimates landfill gas emission using methane concentration and wind speed for two vertical planes perpendicular to the wind direction, one located upwind and the other located downwind of the landfill. The plane-integrated concentration can be obtained with either Tunable Diode Laser Absorption Spectroscopy [*Goldsmith et al.,* 2012] or Fourier transform infrared spectroscopy [*Hashmonay and Yost*, 1999; *Thoma et al.*, 2005; *Oonk*, 2010]. The main advantage of this method is that it provides a large measurement footprint, so it can be used to estimate methane emission from area sources, such as feedlots, lagoons, or landfills. The disadvantages of these two methods include the need for specialized and expensive instrument, infrequent measurements due to the intensive labor required to set up instruments, atmospheric stability and wind direction dependence, and the challenge of obtaining gas concentration distributions along the two planes. It is important to note that all these methods provide methane emission snapshots with poor temporal resolution due to high labor requirements and the lack of the capability for automated measurement.

Another method, called the eddy covariance method, has recently been used in landfill gas emission studies [*Laurila et al.*, 2005; *Eugster and Plüss*, 2010; *Schroth et al.*, 2012]. Eddy covariance is a micrometeorological method using the theory of turbulent transport in the surface layer of the atmosphere [*Verma et al.*, 1986; *Baldocchi et al.*, 1988]. It estimates a particular gas flux from the covariance between vertical wind speed and the gas concentration measured at some carefully chosen height above a surface. Typically, vertical wind speed and gas concentration are both measured at 10 Hz and their covariance is calculated over a suitable interval such as 30 min or 60 min (see equations (3a) and (3b) below). The main advantages of this method over others are the following: (1) it provides an in situ and direct measurement of spatially weighted average gas flux over a large area, the so-called footprint [*Kljun et al.*, 2004], in the upwind direction (normally to a distance of 100 times the instrument height), (2) it has the capability of automation for continuous measurements, (3) it leaves the surface undisturbed, and (4) limited labor is required. Furthermore, because of its capability for continuous measurement, the eddy covariance technique [*Baldocchi et al.*, 1988; *Baldocchi*, 2003] is an excellent tool to investigate underlying processes, such as how each environmental variable affects landfill gas flux. Thus, it is an ideal method for long-term and continuous landfill gas emission measurements and modeling studies. One of the major challenges of the eddy covariance method is that the gas analyzer must have high precision and high speed to measure small-concentration variations in the

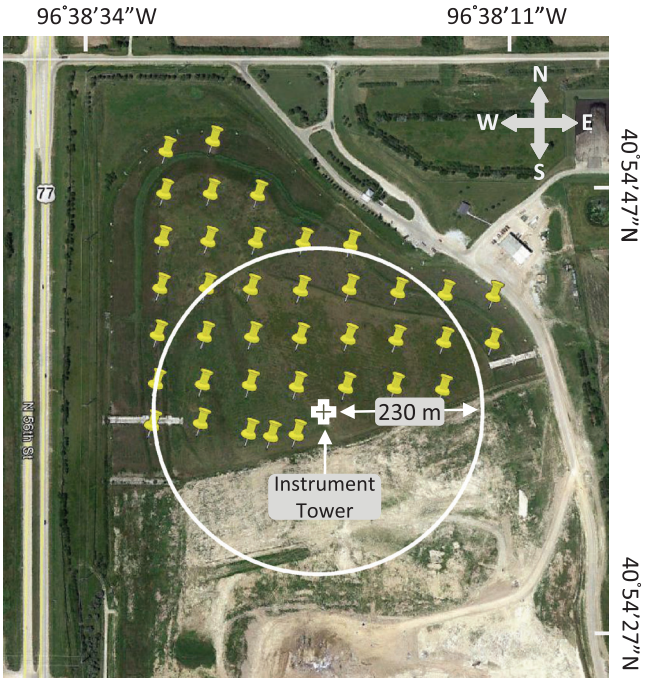

Figure 1. Layout of the experiment site at Lincoln landfill, showing the locations of the eddy covariance instrument tower (white cross) and passive vents (yellow pins). Footprint analysis [*Kljun et al.*, 2004] shows that under near-neutral or unstable conditions, 90% of measured methane emission was from the range of 0 to 230 m (white circle) in the upwind direction. The map was taken on 28 June 2010, courtesy of Google Earth.

turbulent atmosphere. In the late 1980s, fast response gas analyzers and data acquisition data loggers or computers became available. This allowed the eddy covariance method to be widely used by micrometeorologists and ecologists to measure exchanges of CO_2, H_2O vapor, other trace gases, and energy between various ecosystems and the atmosphere [*Baldocchi et al.*, 2001a; *Baldocchi*, 2003]. Despite its gain in popularity for flux measurements, however, it has disadvantages of needing high-performance equipment and a certain level of expertise.

To the best of our knowledge, there is only one study in the literature that tried to quantify landfill methane emissions on a continuous and long-term basis (about 6 months) with the eddy covariance method [*Lohila et al.*, 2007], although a few studies did use the eddy covariance method at landfills but only on a short-term field campaign basis ranging from a few hours to 2 weeks [*Hovde et al.*, 1995; *Tregoures et al.*, 1999; *Fowler and Muller*, 2005; *Eugster and Plüss*, 2010; *Schroth et al.*, 2012].

In this present paper, we report a half year of continuous methane emission measurements over a landfill near the city of Lincoln in Nebraska, USA, using the eddy covariance method. One of the main objectives of this study is to understand the impact of changes in barometric pressure on landfill methane emissions. The implications from the present study for monitoring and understanding landfill gas emissions and for formulating regulations will be discussed.

2. Materials and Methods

2.1. Site Information

The study was done at the Bluff Road Landfill, which is located north of Lincoln, NE, (40.908°N, 96.638°W, 367 m above sea level) and is just outside the city limit. The landfill opened in October 1988 and is due for closure around the year 2035. It covers approximately 69 ha, with a capped area of 16 ha. The total capacity of the landfill is 23.6 million metric tons. By the end of October 2010, the estimated total amount of waste already in place was 6.1 million metric tons. Decomposition begins immediately after waste is deposited in the landfill. During the early stage, carbon dioxide is the main gas produced while oxygen is still available [*Bogner and Matthews,* 2003; *Oonk,* 2010]. After all the oxygen is consumed, methane and carbon dioxide are the two major components. By volume, the landfill gas typically contains 40% to 60% carbon dioxide, 45% to 60% methane, and a few percent of other gases, including nitrogen, oxygen, ammonia, sulfides, hydrogen, and carbon monoxide [*Bogner and Matthews,* 2003; *Oonk*, 2010]. The landfill gas we collected at our site on 4 May 2011 contained 41.3% CO_2 and 54.4% CH_4 (K. Kruszynski, Civil & Environmental Consultants Inc., Lombard, Illinois, personal communication, 2011). The northern portion of the landfill was capped with a geomembrane liner and a 0.5 m layer of clay soil. The landfill gas passively diffused out to the atmosphere through evenly distributed 12.5 cm diameter vent pipes that penetrated the membrane to a depth of 2 m below the surface (Figure 1). On average, there was one vent per 3000 m^2. The southern portion of the landfill was capped only with a 0.5 m

layer of clay soil; no geomembrane liner was installed at that time, so the landfill gas diffused out through the layer of clay soil (Figure 1). The waste at the southern portion is relatively newer than that at northern portion of the landfill. Based on its design drawing, the waste depth at the capped area ranged from 18 to 40 m.

2.2. Eddy Covariance Flux System

The emissions of CH_4 and CO_2 over the landfill have been measured continuously with an eddy covariance system from June 2010 to present (October 2013). The data reported in this paper are from the period of June 2010 to December 2010 when no gas recovery (capture) was in operation. The instruments, which were mounted 3 m above the ground on a tripod, consisted of a 3-D sonic anemometer (Windmaster Pro, Model 1352, Gill Instruments Ltd., Lymington, England), an open-path CH_4 analyzer (Model LI-7700, LI-COR Biosciences, Lincoln, NE, USA), and an open-path CO_2/H_2O gas analyzer (Model LI-7500A, LI-COR Biosciences, Lincoln, NE, USA). The sonic anemometer was located between the CH_4 analyzer and the CO_2/H_2O analyzer. The outputs from the sonic anemometer and from the two gas analyzers provided turbulent fluctuations in three wind components (w, u, v, m s^{-1}), sonic temperature (T_s, °C), CH_4 density (ρ_{CH4}, mmol m^{-3}), water vapor density (ρ_v, mmol m^{-3}), and CO_2 density (ρ_{CO2}, mmol m^{-3}). The raw data from each 30 min period were recorded at a 10 Hz rate on a USB drive inside the CO_2/H_2O analyzer control box (Model LI-7550, LI-COR Biosciences, Lincoln, NE, USA). The raw data were transferred weekly to a computer at our office via the internet. Power spectra of CH_4, CO_2, and H_2O density and cospectra computations between gas density and vertical wind speed (w) showed that these sampling rates were adequate for measuring fluxes with this configuration. The instrument station was located near the middle of the relatively flat plateau of the landfill (Figure 1). The fetch from all directions was more than 250 m. Calculations with a footprint model [*Kljun et al.*, 2004] indicated that, under neutral or unstable conditions, 90% of the flux is from a distance within 230 m of the instrument tower in the upwind direction (Figure 1).

The barometric pressure was measured with a pressure transducer (model 1800-03A-L3N-B, Honeywell, Freeport, Illinois, USA) included in the open-path CH_4 analyzer. According to the manufacturer's specifications, the transducer has an accuracy of ±12.9 Pa and a response time of 1 ms.

The entire eddy covariance flux station was powered by a solar power system. The system included three solar panels (Model KC130TM, 130 W, Kyocera Solar, Scottsdale, Arizona, USA), one battery charge controller (Model TS-MPPT-45, MorningStar Corporation, Newton, Pennsylvania, USA), and four 6 V 190 Ah sealed AGM batteries wired in parallel to produce a 12 V power system. The solar system was designed for 5 days of autonomous (i.e., no Sun) operation at Lincoln, Nebraska's latitude (Figure 1) and average weather conditions. On average, the total power consumption of the entire flux station was approximately 24 W. Internet access was achieved with a line of sight microwave link using Wi-Fi radios and antennas commonly found in the commercial rural Wi-Fi delivery industry (Model Rocket M Radio, Ubiquiti Networks, San Jose, California, USA). The microwave link connected the instrument tower by radio to a maintenance building at the landfill that has public IP internet connectivity.

2.3. CH_4 and CO_2 Emission Rate Calculation

We define the emission rate of methane (F_{CH4}) or carbon dioxide (F_{CO2}) as the amount of the gas escaping from the landfill into the atmosphere per unit area per unit time. This differs from the concept of production rate of the gas (P_{CH4} or P_{CO2}) at the landfill, which is the amount of CH_4 or CO_2 produced inside the landfill per unit area per unit time. As an example, in steady state, the relationship between the CH_4 emission rate and production rate can be expressed as

$$F_{CH4} = P_{CH4} - O_{CH4} - \mathrm{Rec} \quad (2)$$

where O_{CH4} is the oxidation loss as methane diffuses out of the landfill and Rec is the amount of gas recovered, which was zero at the time of our field measurement.

The production rate depends on (1) the total amount of waste at the landfill, (2) the composition of the waste, (3) the age of the waste, and (4) the climate conditions, mainly temperature and precipitation [*Goldsmith et al.*, 2012]. The emission rate depends on (1) the amount of methane produced, (2) the amount of CH_4 oxidized in the top cover soil, (3) the amount of methane recovered, either flared or used for generating electricity, and (4) the changes in methane storage inside the landfill due to changes in barometric pressure,

as we will show in this present study. Therefore, the emission rate and production rate are two different concepts and are equal only under steady state conditions with no oxidation loss or gas recovery.

With the eddy covariance method, F_{CH4} and F_{CO2} can be calculated using the following equations [*Baldocchi et al.*, 1988]:

$$F_{CH4} = \overline{w\rho_{CH4}} \tag{3a}$$

$$F_{CO2} = \overline{w\rho_{CO2}} \tag{3b}$$

where w is the vertical wind velocity (m s^{-1}), ρ_{CH4} is methane number density (mmol m^{-3}), and ρ_{CO2} is carbon dioxide number density (mmol m^{-3}). The overbars indicate 30 min averages. In our calculations, the standard procedure for computing eddy covariance fluxes using a trace gas analyzer and a sonic anemometer is to decompose the time series of the vertical velocity and the trace gas scalar into a mean (e.g., $\overline{w}$) and a perturbation (e.g., w'). Other routine procedures include despiking, angle-of-attack correction [*van der Molen et al.,* 2004], coordinate rotation [*Baldocchi et al.*, 1988], sensor separation and spectral correction [*Moncrieff et al.*, 1997], and density correction [*Webb et al.,* 1980]. The open-path methane analyzer is a laser-based trace gas analyzer measuring near-infrared light absorption at the 1.651 μm wavelength. It employs a Herriott cell configuration [*Herriott and Schulte*, 1965] with a mirror spacing of 0.47 m and a total optical path length of 28.2 m [*McDermitt et al.*, 2011]. Methane density is measured using wavelength modulation spectroscopy [*Silver*, 1992]. As a result, the measured CH_4 density is affected by sensible heat and latent heat flux, and by spectroscopic effects (e.g., line broadening) due to changes in temperature, pressure, and water vapor. These spectroscopic effects occur in a predictable and consistent manner and can be accounted for with multipliers in the density correction equation [*Webb et al.*, 1980]. See equation (3) of *McDermitt et al.* [2011] for more details.

Normally, the eddy covariance method is used over a relatively uniform field in terms of the source and sink strengths for the trace gases of interest. The concentrated point sources (vents) distributed to the north of our eddy covariance tower could affect the CH_4 and CO_2 eddy flux measurements. However, if we can assume that the gases from vents are mixed well with local turbulence in the surface layer, then the eddy covariance method is valid for measuring emissions from the footprint area. We did a follow-up footprint modeling study [*Lin et al.*, 2013] to investigate the spatial and temporal variations of CH_4 and CO_2 emission in the context of vent locations. Our preliminary results suggest that CH_4 and CO_2 from the vents did mix well in the surface layer when friction velocity was higher than 0.1 m s^{-1}, which was about 90% of the time.

2.4. Wavelet Coherence Analysis

Many environmental variables influence landfill gas emissions over a wide range of time scales (or frequencies). The landfill methane emission rate is the sum of all influences superimposed on top of each other. Wavelet coherence analysis is a powerful tool to investigate the cause and effect relationships between two time series variables at different time scales [*Katul et al.*, 2001], a feature that a simple linear regression method cannot accomplish. We used wavelet coherence analysis to examine the coherence and the phase angle between barometric pressure (P) and methane emission rate (F_{CH4}) for different time scales at different times of the season. The coherence is a quantity between 0 and 1, which measures the cross correlation between the barometric pressure time series and methane emission time series as a function of time frequency [*Torrence and Compo*, 1998]. In addition, wavelet coherence allows us to examine whether or not the highest coherence regions in the frequency domain have a consistent phase angle, which indicates a phase-locked relationship between the two time series. Such a phase-locked relationship in turn represents a causality, the dependence of one variable on another [*Grinsted et al.,* 2004; *Detto et al.,* 2012; *Hatala et al.,* 2012]. In the wavelet analysis, coherence (Coh) is defined as the square of the cross-wavelet spectrum normalized by the individual wavelet power spectra for P and F_{CH4} time series [*Torrence and Compo*, 1998] as

$$\mathrm{Coh} = \frac{S\left(C^{*}_{F_{CH4}}(a,b)C_P(a,b)\right)}{\sqrt{S\left(\left|C_{F_{CH4}}(a,b)\right|^2\right)}\sqrt{S\left(\left|C_P(a,b)\right|^2\right)}} \tag{4}$$

where $C_{FCH4}(a,b)$ and $C_P(a,b)$ are the continuous wavelet transforms of F_{CH4} and P at scale a and position b. The superscript * represents the complex conjugate and S is a smoothing operator in time and scale.

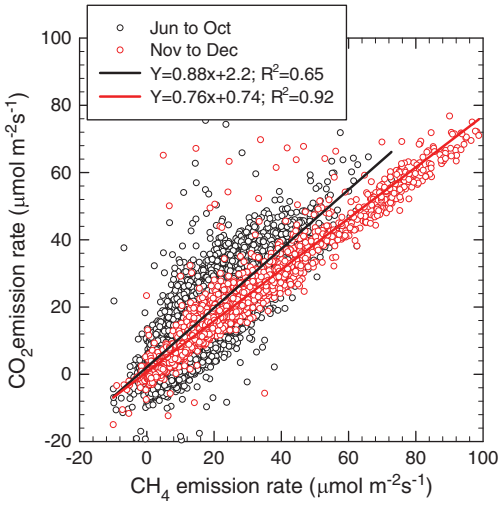

Figure 2. The relationship between CH_4 emission and CO_2 emission rates at Bluff Road Landfill, Lincoln, Nebraska, for the warm season (from June to October, black symbols) and for the cold season (from November to December, red symbols). The two straight lines are linear regressions fitted to data sets over the two seasons. The two slopes are significantly different at the confidence interval of 95%.

The coherence phase angle (φ) is defined as the arctan of the ratio between the imaginary part (quadrature-wavelet spectra) and the real part (cowavelet spectra) of the cross-wavelet transform:

$$\varphi = \tan^{-1}\frac{\mathrm{Imag}\left(C^*_{F_{CH4}}(a,b)C_P(a,b)\right)}{\mathrm{Real}\left(C^*_{F_{CH4}}(a,b)C_P(a,b)\right)} \quad (5)$$

Phase angle of 0° means that the two time series are in phase, while phase angle of 180° means out of phase.

3. Results and Discussion

3.1. Relationship Between CO_2 and CH_4 Emission Rate

With the eddy covariance method, we were able to continuously monitor methane and carbon dioxide emission rates (fluxes) at the landfill. From the data obtained during the cold season (November to December), we saw that the CO_2 and CH_4 emission rates were highly linearly correlated with a slope of 0.76 and R^2 of 0.92 (Figure 2). This slope was consistent with gas samples extracted from the landfill on 4 May 2011 that contained 41.3% CO_2 and 54.4% CH_4, which exactly yielded a $\frac{[CO_2]}{[CH_4]}$ ratio of 0.76. During the November to December time period, the air temperature at the Bluff Road Landfill fluctuated from −15°C to +15°C. At these temperatures, we would expect that soil CO_2 respiration and CH_4 oxidation in the 0.5 m deep soil cover over the southern portion of the landfill where a membrane and vents did not exist would be substantially reduced especially when the air temperature was below 0°C [*Czepiel et al.*, 1996a; *Börjesson et al.*, 2000]. In the northern portion of the landfill where a membrane and vents were present, there would be little opportunity for the emission gas to encounter oxidizing conditions before leaving the vents. Consequently, the gas composition emitted at the landfill surface would not be significantly modified as compared with that produced inside the landfill. If gas composition is not altered, then the slope in Figure 2 should be close to the ratio of CO_2 and CH_4 concentration in the landfill gas composition. This was indeed the case. *Lohila et al.* [2007], who also used the eddy covariance method at a landfill in Finland, observed a similar value between the ratio of CO_2 to CH_4 emission and the ratio of raw landfill gas composition when the wind came from the direction where CH_4 oxidation was limited.

As mentioned in section 2, CO_2 and CH_4 concentrations were measured with two independent gas analyzers. Emission rates were also computed independently. Figure 2 shows that the slope of the linear regression for CO_2 and CH_4 emission rates obtained during wintertime has the same value as the ratio of CO_2 to CH_4 concentration in the raw landfill gas composition. This gave us confidence that gas emission measurements with the eddy covariance technique at our landfill site were dominated by emissions from the landfill and were not substantially affected by fluxes from the surrounding landscape. Moreover, our result is consistent with other published reports, which show that landfill gas typically contains 50 to 60% methane, 45% carbon dioxide, and a few percent of other gases [*Czepiel et al.*, 1996a; *Bogner and Matthews,* 2003; *Lohila et al.*, 2007].

In the warm season (from June to October) the relationship between the CO_2 and CH_4 emission rates had a slope of 0.88 ($R^2 = 0.65$), significantly (at the confidence interval of 95%) higher than that from the cold season (Figure 2). This could be because of higher soil CO_2 respiration and higher oxidation rate of CH_4 from the top cover soil. As a result, the CO_2 emission rate probably was higher than the CO_2 production rate associated with CH_4 production, while the CH_4 emission rate was lower than the production rate. As mentioned earlier, methane oxidation might only happen in the southern portion of the landfill, because only there would methane pass through the cover soil before it diffused out into the atmosphere. In the northern portion of

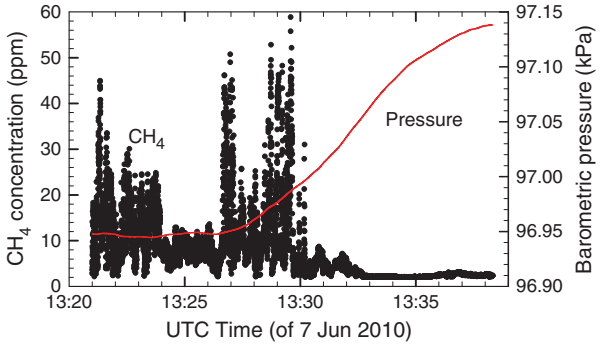

Figure 3. The response of atmospheric methane concentration (dark symbols) to an increase in barometric pressure (red line). This data set was obtained on 7 June 2010 during the passage of a cold front. The independent variable on the *x* axis is UTC time (universal time coordinated).

the landfill, we expect that the methane oxidation rate might be insignificant for both the cold season and the warm season, since methane diffuses out through vent pipes where the methanotrophic activity would be very limited. Note that the relationship between CH_4 and CO_2 emissions in the warm season was more scattered compared to the data set obtained in the cold season. This may arise for two reasons. First, both soil CO_2 respiration and CH_4 oxidation are strongly temperature dependent [*Czepiel et al.*, 1996a; *Börjesson et al.*, 2000], which may lead to large diurnal variations in CO_2 and CH_4 emission rates. For example, according to *Czepiel et al.* [1996a], the oxidation rate in the cover soil was 30% of the methane generated inside a landfill near Nashua, New Hampshire, in the summer (June to August) and almost 0% in the winter (December to February). Second, diurnal variation in photosynthetic CO_2 uptake by leaves from vegetation grown on the cover soil might affect warm season CO_2 fluxes, since leaf CO_2 uptake is driven by photosynthetically active radiation. This could also modify the ratio of CH_4 and CO_2 emission rates.

3.2. Response of Landfill Methane Emission to Change in Barometric Pressure

Figure 3 illustrates how quickly the atmospheric methane concentration at the landfill site responded to changes in the barometric pressure. Both the methane concentration and barometric pressure were measured with the open-path CH_4 analyzer at the height of 3 m. The data set was obtained on 7 June 2010 during the passage of a cold front. The barometric pressure started increasing at 13:27 (UTC, universal time coordinated). Three minutes later, the methane concentration dropped from 50 ppm to 7 ppm (vol/vol, parts per million). As the pressure continued to increase, the methane concentration further decreased to the atmospheric background level of about 2.0 ppm by 13:33. Note that for this, event the barometric pressure increased from 96.95 kPa at 13:27 to 97.13 kPa at 13:37, a 0.18 kPa increase in 10 min, and a change rate (dP/dt) of 1.08 kPa h^{-1}. This was a large dP/dt, as the rates we normally saw during the half-year experiment were in the range of ±0.1 kPa h^{-1}. We also note that the large variations in methane concentration before 13:30 (Figure 3) were because of changes in the sign of the vertical wind directions. The open-path methane analyzer most likely would see higher concentrations of methane when an air parcel was moving upward (positive w) and lower concentrations when an air parcel was moving downward (negative w). The relatively lower methane concentration (~13 ppm) observed around 13:25 was likely due to the wind direction shift and nonuniform distribution of methane source strength relative to the instrument tower.

The response of methane emission rates to changes in barometric pressure is even clearer on longer time scales, such as a month. Figure 4 shows a time series of methane emission rates and barometric pressures obtained from 16 November to 18 December 2010. Emission rates were strongly suppressed when atmospheric pressure increased ($dP/dt > 0$ Pa h^{-1}, light yellow shaded periods), and emission rates were strongly enhanced when barometric pressure decreased ($dP/dt < 0$ Pa h^{-1}, light blue shaded periods). For example, between 16 November at 23:00 and 18 November at 05:30, barometric pressure steadily rose from 95.647 kPa to 97.860 kPa, and the average F_{CH4} during this period was 2.3 ± 3.3 (mean ± standard deviation) μmol m^{-2} s^{-1}. By contrast, from 18 November at 05:30 to 19 November at 10:00, the pressure dropped from 97.860 kPa to 96.306 kPa, and the average emission rate increased to 24.3 ± 15.3 μmol m^{-2} s^{-1}, 10 times higher than that from the pressure-rising phase just a couple of days earlier.

Another example is during the period of 11 December at 08:30 to 13 December at 10:00, when pressure rose from 95.510 kPa to 97.985 kPa, and the average emission rate was 7.1 ± 9.5 μmol m^{-2} s^{-1}. This is followed by a pressure fall from 16:30 on 13 December until 18:30 on 15 December, during which the average F_{CH4} was 73.8 ± 14.3 μmol m^{-2} s^{-1}, also 10 times higher than the emission rate 2 days earlier. This phenomenon occurred repeatedly.

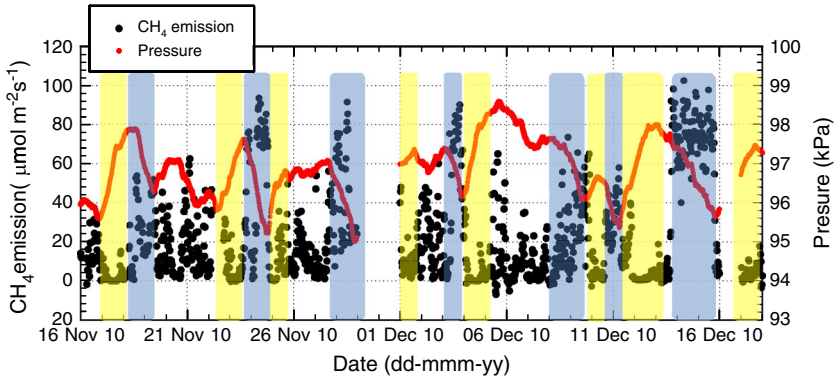

Figure 4. Examples of methane emission rate time series (dark circles) from 16 November to 18 December 2010 and barometric atmospheric pressure (red line) at the landfill, showing the impact of changes in barometric pressure on methane emission rate. Each data point represents half-hour mean methane emission rate and barometric pressure. Light yellow shaded periods represent periods when pressure was rising, and light blue shaded periods represent periods when pressure was falling. Data gaps were due to rain drops or snowflakes blocking the laser beam of the open-path methane analyzer.

The mean emission rate from all the periods when barometric pressure increased ($dP/dt > 0$ Pa h^{-1}, $n = 4487$) was 12.3 μmol m^{-2} s^{-1}, as compared to the mean rate of 22.9 μmol m^{-2} s^{-1} from all the periods when $dP/dt < 0$ Pa h^{-1} ($n = 4563$).

Hourly emission data for the duration of the experiment are shown in Figure 5. Hourly emission rates varied widely throughout the duration. In the warm season, they varied from almost zero to over 50 μmol m^{-2} s^{-1}; however, in the cold season, even larger variations from zero to around 100 μmol m^{-2} s^{-1} were observed (Figure 5). Note that the maximum hourly emission rate in the cold season nearly doubled as compared to that in the warm season. This might be attributed to the following reasons. First, weather systems common to the Great Plains in the winter are much stronger, in terms of duration and magnitude of barometric pressure change, as compared with those in the warm season. From the half-year experiment, we saw that longer and stronger high-pressure weather systems were correlated with higher emission rates (Figure 4). Second, data analysis revealed that almost all the maximum emission rates we observed in the cold season were from the time when the wind was coming from south (Figure 8). As described in section 2, the waste at the southern portion was relatively newer as compared with that at the northern portion of the landfill and was capped only with a 0.5 m layer of clay soil; no geomembrane liner was installed during the time of our field experiment. Thus, the moisture content inside the southern portion probably was higher than in the northern portion, because the moisture from precipitation could penetrate through the cover soil and into waste material. Relatively newer waste and higher moisture content would likely yield higher methane and carbon dioxide production rates. Third, since the landfill gas passively diffused out into the atmosphere via the 0.5 m

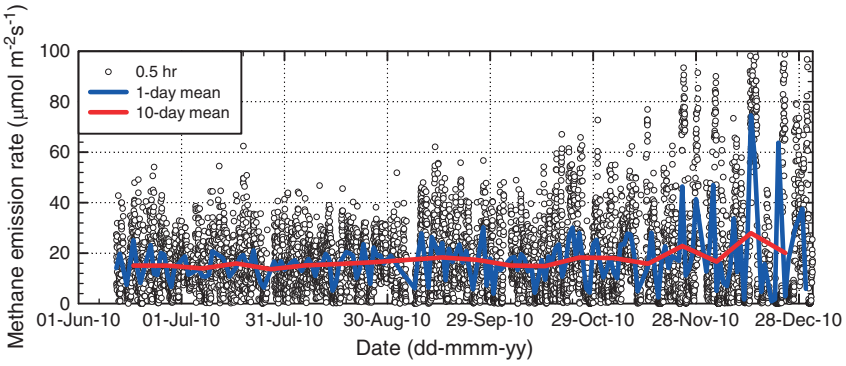

Figure 5. Seasonal variations in hourly methane emission rate (open black circles), 1 day mean (solid blue line), and 10 day mean (solid red line) methane emission rate over the experiment period. The mean methane emission over the entire experiment was 17.7 μmol m^{-2} s^{-1}.

layer of clay at the southern portion of the landfill, a certain amount of methane could be lost via the oxidation process in the warm season. In the cold season, the oxidation rate would likely be reduced, so a lower oxidation rate may play a role for the higher maximum hourly emission rate. So the higher hourly maximum methane emission rates observed in the cold season (Figure 5) can probably be due to combinations of influences from relatively newer waste, better moisture conditions, lower oxidation rates due to cooler temperatures, and the stronger weather systems.

When we averaged hourly methane emission rates into a daily mean, a large day-to-day variation could be easily seen (Figure 5). For example, the daily mean on 11 September was 6.4 μmol m^{-2} s^{-1}. The next day on 12 September, the mean increased to 26.4 μmol m^{-2} s^{-1}, a 4.1-fold increase. Like the hourly emission rate, the variation of daily mean emission was larger in the cold season than that in the warm season. For an example, the daily mean emission rate on 21 December was only 1.8 μmol m^{-2} s^{-1}. On the next day (22 December) the daily mean rate was 63.8 μmol m^{-2} s^{-1}, a 35.6-fold increase! When we averaged the hourly emission rate into 10 day means, little variation can be seen in the data in the warm season (Figure 5). This probably means that the impact of day-to-day changes in barometric pressure on the landfill methane emission was averaged out. In the cold season, even with an average time of 10 days, some variations in the 10 day mean still existed (Figure 5); probably, a longer time was needed to average out the impact of changes in barometric pressure.

It should be noted that the eddy covariance technique gives us a spatially averaged emission over a large area (footprint area) in the upwind direction. For example, when the wind comes out of the east, the measured emission would represent the weighted average rate over the footprint area in the easterly direction (normally from the instrument tower to a distance of 100 times the instrument height) [*Kljun et al.,* 2004]. From our data set, when winds were out of the north, no significant difference in methane emission was observed over the course of the experiment (data not shown). This is probably because, for the northern portion of the landfill, methane diffuses out through vents, bypassing the cover soil and preventing oxidation loss. However, when the wind came from the south, where no geomembrane was installed and it was capped with a 0.5 m layer of clay at the time of our field measurement, we saw slightly higher methane emission rates in the cold season than in the warm season (data not shown). This is consistent with the hypothesis that lower oxidation rates resulted from below freezing temperatures in the cover soil where oxidation would have occurred [*Chanton and Liptay*, 2000; *Lohila et al.*, 2007]. The oxidation rate could drop to nearly zero in the winter when the temperature goes below freezing. Higher CH_4 emission rates from landfills because of lower oxidation rate during cold seasons have also been reported in a few other studies [*Boeckx et al.,* 1996; *Bergamaschi et al.*, 1998; *Lohila et al.*, 2007].

The mean methane emission over this half year was 17.7 μmol m^{-2} s^{-1}. This mean value is well within the range of methane emission rates reported from earlier studies [*Lohila et al.,* 2007; *Goldsmith et al.,* 2012]. The mean methane emission was 15.8 μmol m^{-2} s^{-1} for the warm season (June to October) and 20.24 μmol m^{-2} s^{-1} for the cold season (November to December). The slightly higher emission rate observed in the cold season likely was due to the lower oxidation rate, as we just discussed above. From Figure 5 we can see that after the impact of changes in barometric pressure was averaged out, a fairly stable emission rate can be seen over the course of our experiment. The phenomenon of stable emission rate has been reported a few times in the literature [*Chanton and Liptay*, 2000; *Galle et al.*, 2001; *Lohila et al.*, 2007]. This has been attributed to the relatively stable temperature inside the landfill over a year. Thus, changes in seasonal ambient temperature would have a limited impact on methane production.

Earlier studies show that the temporal variation in methane emission rates can vary by several orders of magnitudes at a given landfill site [*Bogner et al.,* 1997; *Schroth et al.*, 2012]. This has been attributed mostly to different rates of CH_4 generation and different oxidation rates due to different cover soil temperature and soil moisture content [*Czepiel et al.,* 1996a; *Bogner et al.,* 1997; *Chanton and Liptay,* 2000; *Schroth et al.,* 2012]. From the results presented in our study, we suggest that such large temporal variations in landfill methane emissions reported in those earlier studies may have been due to barometric pressure fluctuations, instead of, or in addition to, changes in methane production or changes in oxidation rate. Changes in barometric pressure might play a larger role in regulating landfill methane emission than previously thought. The impact of changes in methane generation rate and oxidation rate on rapid and large temporal variations in methane emission might be very limited, as we will show in the power spectral analysis below.

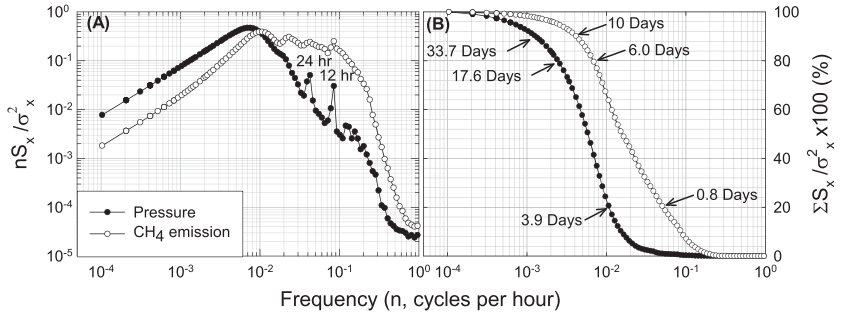

Figure 6. Power spectra (A, S_x) and ogive curves (B, ΣS_x) for the time series of barometric pressure and landfill methane emission rate. Both power spectral densities are normalized by their total variances (σ_x^2) and also weighted by the natural frequency (n). Ogive curves are integrated from high to low frequency and plotted as a percentage of total variance. Arrows on two ogive curves indicate various length of measurement needed to capture 20%, 80%, or 90% of total variance.

3.3. Power Spectrum Distribution and Wavelet Coherence Analysis

We applied Fourier analysis to time series of F_{CH4} and P. The power spectra and ogive curves are shown in Figure 6. Both power spectra and ogives were normalized by their total variances (σ_x^2). Ogive curves are integrated from high to low frequency and plotted as a percentage of total variance. The spectral peak in barometric pressure time series occurred from 0.0106 cycle h^{-1} to 0.00237 cycle h^{-1}, which corresponded to periodicities (the inverse of frequency) of 3.9 day per cycle to 17.6 day per cycle, respectively. This frequency range contributed about 60% of total spectrum power as we can see from the ogive curve (Figure 6b), and presumably, it was associated with the synoptic weather pattern. The pressure power spectrum (Figure 6a) also had two distinguishable sharp peaks (12 and 24 h) at frequencies corresponding to diurnal and semidiurnal barometric pressure variations, which harmonics were probably due to the diurnal cycle [*Mass et al.*, 1991; *Baldocchi et al.*, 2001b]. For the methane emission time series, the spectral peak occurred in the frequency range from 0.0504 h^{-1} to 0.00691 h^{-1}. This corresponded to periodicities of 0.8 days to 6.0 days, which contributed about 60% of variance. Note that the spectral peak for methane emission shifted toward higher frequencies as compared to barometric pressure. This probably occurred because of frequent wind direction shifts and the heterogeneity of methane emission sources over the landfill surface (Figures 1 and 8).

From the power spectrum and ogive analysis, we can see that even if the measurement duration for methane emission at our landfill site lasted for 12 h (0.0848 cycle per hour), one would only capture about 10% of the total variance. In order to capture 90% of total variance, at least 10 days of continuous measurements are needed (Figure 6b). Variation in F_{CH4} with frequencies longer than 10 days per cycle contributed about 10% of total variance. This means that to capture the total variance, one would need to continuously measure the landfill emission.

The power spectral analysis (Figure 6) illustrates the variance distribution among different frequencies for the time series of F_{CH4} and P. However, it does not provide any information about the cause and effect relationship between these two time series. As described in section 1, wavelet analysis can allow us to look into the cause and effect relationship between two time series at different time scales. We used wavelet coherence analysis to examine the coherence and phase angle between these two time series (Figure 7). First, the two time series were detrended to remove seasonal changes (3, 6, and 12 month cycles). Then they were standardized by subtracting their mean and dividing by their standard deviation [*Schruben,* 1985; *von Storch and Zwiers,* 2003]. This procedure gave dimensionless standardized variables (P_{std}). High coherence with a constant phase angle between two time series often indicates a cause and effect relationship [*Grinsted et al.*, 2004; *Hatala et al.*, 2012]. A circular mean [*Zar,* 2010] of the coherence phase was used in this study for a set of phase angles within the region between 1 day and 8 days (two pink dotted horizontal lines in Figure 7) when the coherence between the two time series was strong. The frequency space for significant cross correlations in the period ranges from 1 to 8 days through the entire set of observations, indicating a strong dependence of the methane emission on changes in barometric pressure. This indicates a simple causality between the barometric pressure and methane emission whose oscillations are phase locked. The phase angle within regions of 95% confidence interval (inside thick black contours) has a circular mean phase of 110.7 ± 27.6°

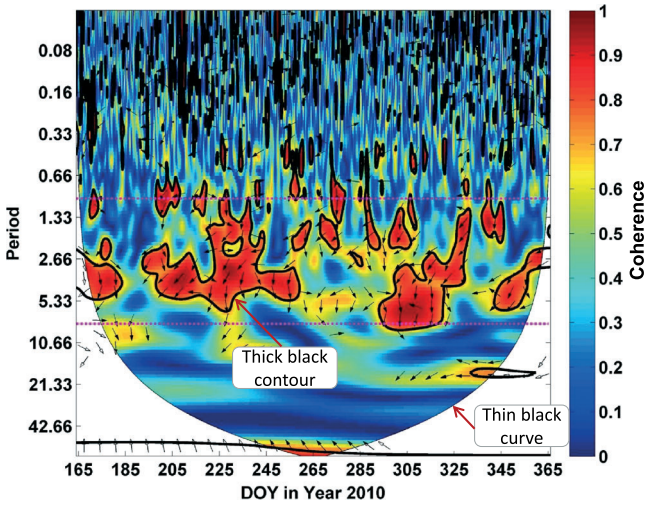

Figure 7. Analysis of squared wavelet coherence between barometric pressure and methane emission time series. Annual, semiannual, and 3 month cycles were removed, and both series were standardized [*Schruben,* 1985; *von Storch and Zwiers,* 2003]. Different colors represent the values of coherence between pressure and the methane emission. The thick black contours represent a 95% of confidence interval against red noise [*Torrence and Compo*, 1998]. The phase angles between the two time series are represented by directions of arrows with increasing phase angle in the clockwise direction. Arrows pointed to the right indicate that the phase angle is 0°, which means that the two time series are in phase; arrows pointed downward mean that the phase of pressure variation leads the methane emission by 90° and so on. The thin black curve is the cone of influence, which will occur at the beginning and end of the wavelet power spectrum [*Torrence and Compo*, 1998].

(mean ± standard deviation) [*Zar*, 2010]. This result shows that the barometric pressure and methane emission are neither in phase nor out of phase but that the barometric pressure leads the methane emission by 110.7° on average for the frequency range of 1 day to 8 days per cycle. This interpretation is consistent with the result we presented in Figure 4, which showed higher methane emission rates during the pressure-rising phases and lower methane emission rates during the pressure-falling phases. When studying the effect of barometric pumping on contaminant transport in the porous earth material, *Auer et al.* [1996] showed that in theory, the velocity of gas movement through the porous earth medium is ~90° out of phase from the barometric pressure changes. So our phase angle of 110.7 ± 27.6° was fairly close to the theoretical value.

From Figure 7, for periods shorter than 1 day per cycle, there was no significant relationship between methane emission rates and changes in barometric pressure. This is because barometric pressure variations faster than this contributed less than 1.5% of total variance, yet more that 22% of variance in methane emission rates were associated with frequencies faster than 1 cycle per day (Figure 6b). Emission variations occurring at this frequency range are likely due to hourly variations in wind direction and spatial heterogeneity of methane emission sources. About 62% of the total variance in methane emission rate came from periods between 1 day and 8 days per cycle, which can be attributed mainly to fluctuations in barometric pressure, since they were coherently correlated in this frequency range. The rest of 16% of total variance was coming from periods longer than 8 days per cycle in the landfill methane emission. Those longer periods may have been associated with seasonal changes in methane production rate due to small changes in temperature inside the landfill and with seasonal changes in oxidation rates due to changes in cover soil moisture and temperature.

The time series of F_{CH4} and P we presented in Figure 4, the power spectrum analysis in Figure 6, and the wavelet coherence analysis in Figure 7 all suggest that changes in barometric pressure played a dominant role in regulating landfill methane emissions for periods ranging from 1 to 8 days. Could this be due to some artifact? We know that changes in barometric pressure are often associated with a shift in wind direction. We also know that CH_4 emissions over a landfill have a high spatial variability [*Czepiel et al.*, 1996b; *Lohila et al.,* 2007], since the methane generated inside the landfill most likely will diffuse out to the atmosphere through the least resistant pathways. Also, the distribution of vents relative to the location of our eddy covariance flux tower is not uniform among different wind directions (Figure 1). So it is likely that wind direction affects measured CH_4 concentrations and emission rates in addition to changes in barometric pressure (Figures 3 and 4). To test this hypothesis, we plot methane emission rate versus wind direction in Figure 8. The data set was color coded with the rate of change in standardized barometric pressure (dP_{std}/dt). Using the slopes of linear regressions of consecutive barometric pressure half-hour means over periods of 9 h, dP_{std}/dt values were obtained. Each mean was a running average advanced every half hour. The red color represents the time with the barometric pressure rising ($dP_{std}/dt > 0$), and the blue color represents the time with the barometric pressure falling ($dP_{std}/dt < 0$). The time series of methane emission rates was detrended to

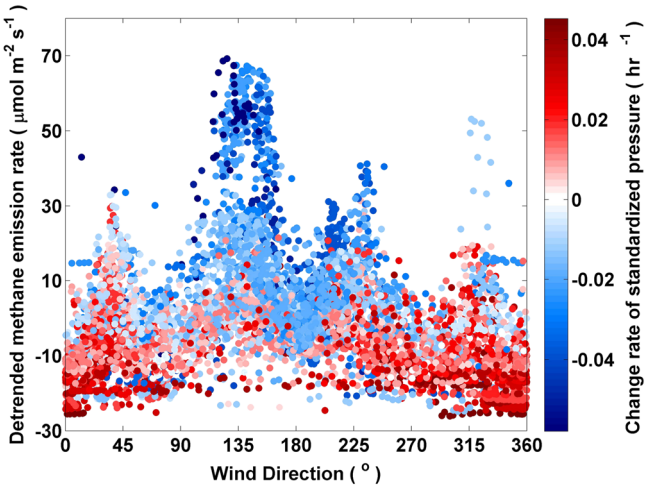

Figure 8. The relationship between methane emissions and wind direction. Different colors represent the rate of standardized barometric pressure change (dP_{std}/dt) with the red color representing rising barometric pressure ($dP_{std}/dt > 0$) and the blue color representing falling barometric pressure ($dP_{std}/dt < 0$). The time series of methane emission rate was detrended to remove seasonal variations (3, 6, and 12 month cycles). Negative methane emission rates were resulted from the detrending procedure. See the text for description of the detrending and standardization procedures.

remove seasonal variations (3, 6, and 12 month cycles). As expected, Figure 8 shows strong spatial variations in CH_4 emission rates from different wind directions, most likely due to locations of the vents in relation to the tower, and possible heterogeneity of methane source strength; however, for all wind directions, higher rates are associated with dropping P_{std} (blue symbols), and lower rates are associated with increasing P_{std} (red symbols). To demonstrate this, we binned the methane emission data set based on the wind direction from zero to 360° with a bin size of 10°. Paired *t* test analysis showed that for every bin, the mean value of methane emission was significantly higher when $dP_{std}/dt < 0$ than when $dP_{std}/dt > 0$ ($P < 0.01$). From this exercise, we can rule out the possibility that the close relationships presented in Figures 3 and 4 between changes in barometric pressure and methane emissions were artifacts due to changes in wind direction associated with barometric pressure changes. Note that when winds are out of the south, dP_{std}/dt more likely tends to be negative and fluxes are enhanced, and when winds are out of the north, dP_{std}/dt more likely tends to be positive and emission rates are suppressed.

Previous studies of landfill methane emissions have shown some correlation between landfill methane emission and barometric pressure [*Young*, 1990, 1992; *Czepiel et al.*, 1996b, 2003; *Poulsen et al.*, 2003; *Gebert and Groengroeft*, 2006]. For example, using the atmospheric tracer method, *Czepiel et al.* [2003] showed that measured emission rates from the Nashua, New Hampshire municipal landfill were negatively and significantly correlated with barometric pressure. A similar negative correlation between methane emission and the barometric pressure over wetlands or peatlands has been reported in a few studies [*Mattson and Likens*, 1990; *Shurpali et al.*, 1993; *Tokida et al.*, 2005; *Tokida et al.*, 2007]. *Shurpali et al.* [1993] found that strong episodic methane emissions at a wetland in Northern Minnesota were associated with drops in barometric pressure. In a study done by *Tokida et al.* [2007], it was found that within a matter of 10 min, the methane emission rate from a peatland in Japan was increased by more than 2 orders of magnitude because of a fast drop in barometric pressure.

Moreover, this phenomenon has been known to mining ventilation engineers in the United Kingdom for more than 250 years [*McQuaid and Mercer,* 1991]. In fact, some mining explosions in the United Kingdom have been linked to low barometric pressure weather systems. According to *McQuaid and Mercer* [1991], if barometric pressure falls too quickly, mining operations need to be suspended regardless of whether or not a modern ventilation system is in use and not resumed until pressure starts increasing.

From the data presented here and those published in the literature [e.g., *Shurpali et al.*, 1993; *Young,* 1992; *Tokida et al.*, 2007], we conclude that dynamic variations of CH_4 emission observed from landfills with periods of 1 day to 8 days per cycle were largely due to dP/dt. Using a deterministic mechanistic physical model, *Young* [1992] was able to illustrate that the emission rate depended linearly on dP/dt, not on the absolute value of barometric pressure.

Since the production of methane inside the landfill may change only slowly over the course of a year, the short-term sensitivity of methane emission to fluctuations in barometric pressure are likely due to advective flow into or out of the landfill and resulting changes in the diffusion gradient between landfill and atmosphere. During the rising pressure phase, a layer of fresh ambient air would be continuously injected

into the landfill. This would greatly reduce the diffusion gradient across the soil surface, making it very difficult for CH_4 and CO_2 to diffuse out (Figure 3). While during the falling pressure phase, the landfill gas would be drawn out (advective flow) and transported away from the surface by turbulent eddy movements, greatly increasing methane emission (Figure 4). This phenomenon, so-called barometric pumping, has been well studied and documented in the literature from studies of the transport of soil gas [*Wyatt et al.*, 1995] and contaminants [*Auer et al.*, 1996] in the soil profile or in fractured permeable media [*Nilson et al.*, 1991], from soil moisture transport through unsaturated fractured rock [*Martinez and Nilson*, 1999], from radon transport across the soil-atmosphere interface [*Clements and Wilkening,* 1974; *Riley et al.,* 1999], and from gas transport inside a well [*You et al.*, 2010]. Those studies have all shown that variations in barometric pressure greatly enhance soil gas or contaminant transport inside the porous medium, because advective flow is orders of magnitude more efficient in transporting gases than molecular diffusion alone [*Nilson et al.*, 1991; *Auer et al.*, 1996]. This barometric pumping mechanism probably is the main reason for the fast and strong response of methane emission to the changes in barometric pressure we observed from our landfill site (Figures 3 and 4). Moreover, vents at the northern portion of our landfill and possible cracks at the southern portion could serve as low resistance air pathways, allowing air to move into and out of the landfill more freely in response to fluctuations of barometric pressure. Vents and cracks may also allow air to exchange more quickly with deeper landfill layers than at a site with a homogeneous cover soil [*Nilson et al.,* 1991].

On the temporal scale of one barometric cycle (3.9 day to 17.6 day), we probably can assume a steady state for gas transport into or out of the landfill in response to the change in barometric pressure. Then the volume of air (ΔV) injected into or drawn out of the landfill during a time interval Δt would be positively correlated to the pressure change rate ($\Delta P/\Delta t$) and the total air space inside the landfill (V_t). The latter depends upon the average air-filled porosity (f_a) and the depth of the landfill (d). At our site, the waste depth ranged from 18 to 40 m. After many years of decomposition, we would expect that f_a would be much higher as compared with that from wetland or peatland. The high V_t because of the deep and highly porous waste material at the landfill probably also plays a role for the strong response of F_{CH4} to changes in P we observed in this present study. This explanation is consistent with the conclusion from a study done by *Wyatt et al.* [1995] who show that the response of landfill gas transport to barometric pumping is stronger at a location with a large soil gas volume in permeable cover soil than with less permeable cover soil.

During our field experiment, the peak-to-peak amplitudes of barometric pressure changes were in the range of 3 kPa in the summer to 5 kPa in the winter, and the standard deviation was 0.677 kPa across the half year. To determine whether the barometric pressure variation we observed during our experiment was unusual or unique, we examined 2008 barometric pressure data from three Ameriflux Research Stations: (1) Howland Forest in Maine (45.207°N, 68.725°W, 61 m above sea level (asl), courtesy of Dr. David Hollinger at USDA Forest Service, Northeastern Research Station), (2) Mead in Nebraska (41.180°N, 96.440°W, 363 m asl, courtesy of Dr. Shashi Verma at University of Nebraska in Lincoln), and (3) Ione in California (38.407°N, 120.951°W, 129 m asl, courtesy of Dr. Dennis Baldocchi at University of California Berkeley). They represent the east coast, midcontinent, and west coast, respectively. The peak-to-peak amplitudes of the barometric pressure fluctuations were in the range of 3 to 5 kPa for all three locations. The standard deviations, calculated based on the hourly barometric pressure data from the whole year, were 0.896 kPa, 0.778 kPa, and 0.670 kPa for Howland, Mead, and Ione, respectively. Power spectral analyses for those three sites reveal a similar spectral power distribution as shown in Figure 6 (data not shown). We also examined hourly barometric pressure time series from Mead, Nebraska, located 50 km north of our landfill site, for the years 2001 to 2010 (courtesy of Dr. Shashi Verma at University of Nebraska in Lincoln). Time series of P from all 10 years have amplitudes and power spectral distributions similar to the data we observed during our experiment. Thus, the barometric pressure variation we saw at the Bluff Road Landfill in the second half of 2010 was normal and not unique to our site.

3.4. Applications

We have demonstrated that methane emissions at the Bluff Road landfill near Lincoln, NE, responded dynamically to changes in barometric pressure (Figures 4 and 5). We have also shown that the seasonal P variation in Lincoln, NE, is similar to that observed across the United States. It is reasonable to expect that responses of landfill methane emissions to barometric pressure variations in other parts of the United States would be similar to what we observed in this present study. Therefore, we recommend that whenever landfill

gas emission is measured, P should be continuously monitored over the measurement duration. Otherwise, it may be difficult to explain the large variations in measured landfill F_{CH4} that are likely to occur [*Young,* 1992].

As we pointed out above, some of the commonly used methods for quantifying landfill methane emission rates, including chamber-based methods, the trace plume method, and the mass balance method, are snapshots in time. Our results demonstrate that emission rates measured with these methods can be subject to large variations in measured emission rates because of the strong dependence of methane emissions on barometric pressure changes. Estimates of long-term integrated total methane emissions based on such measurements will inevitably yield some uncertainties. We used the emission data from two periods (12 to 21 June 2010 and 1 to 10 November 2010) as an example to illustrate the magnitude of this uncertainty. We first calculated the mean emission for each period when $dP/dt > 0$ and $dP/dt < 0$, respectively, then compared them with the annual mean value (17.7 μmol m^{-2} s^{-1}). For the first period (12 to 21 June 2010), the mean emission rate was 12.6 μmol m^{-2} s^{-1} when $dP/dt > 0$ and 19.1 μmol m^{-2} s^{-1} when $dP/dt < 0$. Based on this, one could have 28.8% underestimation to 7.8% overestimation from those snapshot field measurement. For the second period (1 to 10 November 2010), the mean emission rate was 13.5 μmol m^{-2} s^{-1} when $dP/dt > 0$ and 23.4 μmol m^{-2} s^{-1} when $dP/dt < 0$. So one could have 23.8% underestimation to 32.3% overestimation. This simple analysis demonstrates that using the trace plume method, the mass balance method, or the closed-chamber method could yield uncertainties ranging from 28.8% underestimation to 32.3% overestimation in annual total landfill emission. This illustrates the value of making continuous emission measurements. Point-in-time measurements may be subject to large errors.

As we discussed in section 1, there are two major sources that contribute to methane loads to the atmosphere. One is anthropogenic, including landfills, ruminant animals and their waste, mining operations, oil and natural gas production and distribution, and combustion of biomass [*Evans*, 2007]. The other is natural sources, which include peatlands, wetlands, and rice paddies [*Dlugokencky et al.,* 2011]. Reliable estimates of total methane emissions from these major sources are essential to understand the global methane budget and to predict their impacts on climate change, global warming, ozone depletion, and many other environmental issues [*Healy et al.*, 1996; *Bogner and Matthews*, 2003; *IPCC,* 2007; *Dlugokencky et al.,* 2011]. The so-called bottom-up method is a commonly used approach to upscale field measurements of methane emission rates to larger scales [*Tian et al.*, 2010; *Xu et al.*, 2010]. Field measurements often have a low temporal resolution. From our results and many published studies, changes in barometric pressure can play a key role in regulating methane emissions from a landfills [*Young,* 1990, 1992; *Czepiel et al.*, 2003; *Poulsen et al.,* 2003], wetland [*Shurpali et al.*, 1993], peatland [*Tokida et al.*, 2005, 2007], lakes [*Mattson and Likens*, 1990], and mining operations [*McQuaid and Mercer*, 1991]. For this reason, we suggest that field flux measurements over landfill, wetland, peatland, or lakes should be made over longer time scales to average out the impact of barometric pumping. Otherwise, measured emission rates may be subject to order of magnitude variations as we saw in the present study. Also, when using published field emission data sets to develop process-based emission models, it is important to note the duration and the temporal resolution of the field measurements in those data sets.

Although our data set was obtained from a landfill site, the same phenomenon would also occur for gas transport through porous media in other environmental contexts. As example, studies have shown that changes in barometric pressure influence soil gas movement, soil contaminant transport, and radon transport across the soil surface in a similar way as we observed in this paper [*Clements and Wilkening*, 1974; *Young*, 1992; *Wyatt et al.,* 1995; *Auer et al.,* 1996; *Riley et al.,* 1999]. Therefore, programs aimed at monitoring gas concentration over porous media such as soil to detect trace gas emissions may not be effective unless the monitoring is continuous. Leaks may be missed if measurements are made during the pressure-rising phase because emission would be suppressed. If continuous monitoring is not feasible due to resource constraints, then it is recommended that surface concentration measurements be made during a phase of falling pressure, "especially if there is a requirement to issue hazard warning" [*Young*, 1992, p. 611].

The eddy covariance technique is a powerful tool widely used in research fields of agricultural and forest meteorology, soil science, ecology, and carbon cycle science [*Baldocchi,* 2003], in part because it is capable of automation allowing for continuous flux measurements over various surfaces without causing disturbance [*Baldocchi et al.,* 2001a]. High temporal resolution of gas and energy fluxes can be measured on a long-term basis. The eddy covariance technique has not been as widely used in landfill gas emission studies as in other research fields because of its requirement for a relatively flat site and a large fetch. However, there have been

a few studies documented in the literature in which it has been used successfully to quantify landfill methane emissions on a short-term basis [*Hovde et al.*, 1995; *Tregoures et al.,* 1999; *Fowler and Muller*, 2005; *Lohila et al.*, 2007; *Eugster and Plüss*, 2010; *Schroth et al.*, 2012]. The eddy covariance method provides a spatially weighted average emission rate over its footprint area in the upwind direction [*Kljun et al.*, 2004], but it does not provide total methane emission from the whole landfill, which is the information landfill owners and government agencies need to know. Heterogeneity due to nonuniformity in waste depth, age of waste, vent distribution, and other factors at a particular landfill make it difficult to upscale from measured methane emission rates to the whole landfill. For this reason, additional research is needed to understand how to upscale measured landfill emission rates, perhaps using a combination of surface modeling and footprint analysis.

Our conclusions are based on results obtained from a landfill site that did not use an active gas collection system at the time the data were taken. According to the EPA website (http://www.epa.gov/lmop/faq/lfg.html), as high as 58% of landfills in the United States do not have active gas collection. This percentage could be even higher for the rest of world. We acknowledge that the results presented in this paper are not applicable to landfills that use active gas collection systems. Gas transport at those landfills will heavily depend on the negative pressure applied by the gas collecting system, causing surface emissions to be much smaller and less sensitive to changes in barometric pressure. Subsequent to the work reported here, the Bluff Road Landfill has begun active gas collection, and as expected, observed emissions are dramatically lower (data not shown). But that is another story.

4. Conclusions

By using the eddy covariance method, we were able to measure landfill methane emissions continuously over half a year from June to December 2010 at the Bluff Road Landfill near Lincoln, Nebraska. We found that the methane emission rate was strongly dependent on changes in barometric pressure, i.e., rising barometric pressure suppressed the emission, while falling barometric pressure enhanced the emission. With power spectral analysis, we were able to show that 60% of methane emission variance was associated with the periods of 0.8 days to 6.0 days (Figure 6b). The time series of methane emission and barometric pressure had strong coherence for the periods ranging from 1 to 8 days, indicating a cause and effect relationship. Our results have two important implications. First, point-in-time methane emission measurements made at monthly or even longer time intervals with techniques such as the trace plume method, mass balance method, or closed-chamber method may be subject to large variations in measured emission rates because of the strong dependence of methane emissions on changes in barometric pressure. Estimates of long-term integrated total landfill methane emissions based on such measurements may have uncertainties (Figure 5), ranging from 28.8% underestimation to 32.3% overestimation. These results demonstrate that continuous measurements greatly improve the likelihood of accurately estimating annual total landfill methane emissions. Second, for programs aimed at detecting trace gas leaks by monitoring gas concentrations over soil surfaces, field measurements should be done during periods when barometric pressure is falling if the monitoring cannot be done on a continuous basis. Otherwise, leaks may be missed because lower concentrations obtained from periods when barometric pressure is rising do not necessarily mean no leakage is occurring at the site.

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