Methane Emissions Measured at Two California Landfills by OTM-10 and an Acetylene Tracer Method

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ABSTRACT: Methane emissions were measured at two municipal solid waste landfills in California using static flux chambers, an optical remote sensing approach known as vertical radial plume mapping (VRPM) using a tunable diode laser (TDL) and a novel acetylene tracer method. The tracer method uses an ultra-sensitive, dual species gas analyzer based on wavelength scanned cavity ring-down spectroscopy that has been developed to measure the concentrations of methane and acetylene with the required sensitivity. The static chamber and VRPM measurements were made on sections of the landfills with active landfill gas collection systems and intermediate or long-term soil covers. Mobile and stationary tracer plume measurements were made at distances of 0.5 to 3 km from the landfills.

Field measurements were performed over a period of several weeks during the fall of 2009 with each landfill being measured twice using the static chambers and VRPM approach. A single set of tracer plume measurements were made at each of the landfills. Mean methane emission rates determined from the VRPM and chambers will be presented and compared to methane flux results obtained using the acetylene tracer method.

INTRODUCTION

Understanding the accuracy of modeling approaches to estimate the GHG emissions of landfills is of increasing importance as the focus of emissions inventorying and reporting shifts from the international or national levels to the facility level. Studies evaluating the methods of measuring or modeling landfill emissions to develop site specific methane budgets have recently been reported (Abichou et al, 2010; Babilotte et al., 2008, 2009; Spokas et al., 2006; Borjesson et al., 2009; Bogner et al., 2009).

In 2006 Waste Management (WM) undertook a study to evaluate methane emissions at landfills located in various climates across the U.S. using measurement and modeling techniques. Emissions measurements were made with static chambers and ground based optical remote sensing (ORS) using a tunable diode laser-based VRPM approach. Information on the VRPM approach is contained in a method termed Other Test Method 10 (OTM-10) (USEPA, 2006). In 2009, WM began a collaboration with

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USEPA and Picarro, to evaluate an acetylene tracer method of determining emissions that utilizes cavity ring-down spectroscopy (CRDS). The methane flux measurements reported in this paper were made with the VRPM approach using tunable diode lasers, static flux chambers and tracer plume techniques using CRDS.

METHODS

Landfill Study Sites

Methane emission measurements were made at the Redwood and Altamont landfills in October of 2009. Each facility is an active large-scale MSW landfill with an active landfill gas collection system (LFGCS). Information on each landfill is listed in Table 1. Aerial photographs of the facilities are shown in Figures 1 and 2.

Table 1. Landfill Study Sites Detail

Site	City, State	Coordinates	Field Campaign Dates
Redwood Landfill	Novato, CA	38°9'57"N 122°37'53"W	10/06/2009–10/08/2009
Altamont Landfill	Livermore, CA	37°45'13"N 121°39'7"W	10/20/2009–10/22/2009

Fig. 1 Aerial Photograph of Redwood Landfill

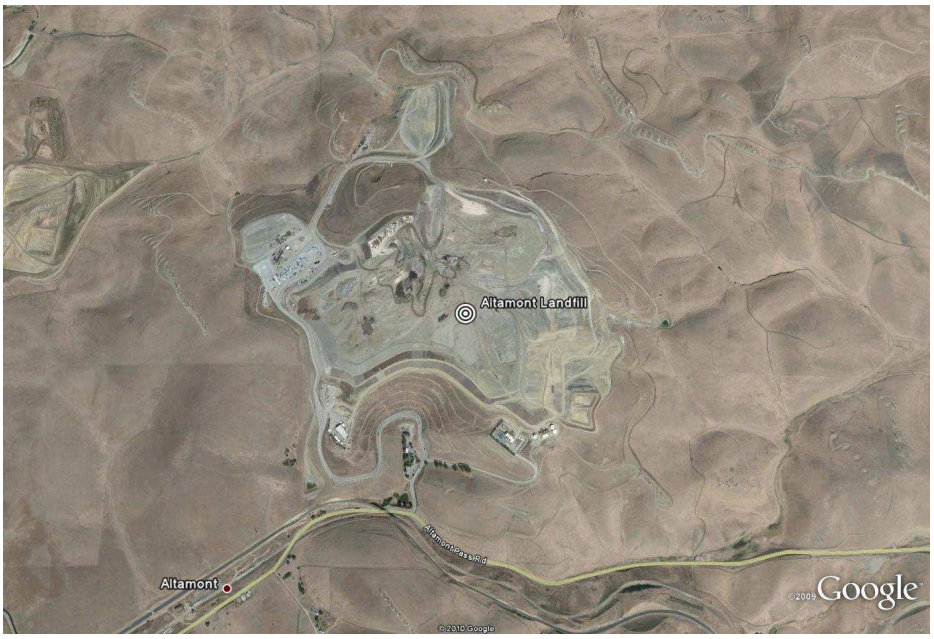

Fig. 2 Aerial Photograph of Altamont Landfill

Static Chamber Measurements

The principle of the technique is to seal a volume of air above a gas-emitting or consuming surface so that the emitted (or consumed) gas cannot escape and its accumulation in the volume can be monitored. The chambers used in this study were constructed of aluminum sheeting with dimensions of 0.63 x 0.63 x 0.2 m covering a surface area of 0.4 m^2. Each chamber consisted of a lid, collar and a small fan to circulate air within the enclosure. Measurements consisted of sealing the chamber lid to the ground on previously installed collars. Gas samples were collected from the chambers immediately after sealing (time = 0) and every 5 minutes over the next 25 minutes using a 60 mL plastic syringe fitted with plastic valves. Samples were analyzed for methane on a gas chromatograph equipped with a flame ionization detector. Methane flux was determined by plotting methane concentration (C) versus elapsed time (t). The slope of the fitted line (dC/dt) was determined by linear regression and a non-zero flux was reported only if there was a 90% confidence ($p<0.1$) in the correlation between methane concentration and time, otherwise a zero flux was reported (Barlaz et al., 2004). The summary flux results and statistics presented are based on the arithmetic mean of all measurements. Chambers were located in a systematic grid established in the area measured by the VRPM method to facilitate comparison.

Vertical Radial Plume Mapping

VRPM configurations were used to measure methane emissions at the landfills. Each VRPM measurement plane consisted of five retro-reflecting mirrors. Two

retroreflectors were placed along the surface at 1/3 and 2/3 of the full optical path, while the remaining three were arranged vertically at the end of the optical path with one at the ground surface and the others approximately 6 and 12 m above the ground surface. Methane-specific TDLs (GasFinder 2.0, Boreal Laser) scan the optical path to each of the five retroreflectors dwelling at each for 15 to 30 seconds during each measurement cycle. Wind speed and vector data were acquired with calibrated meteorological heads (R.M. Young, Model 05103) located approximately 2 m and 14 m above the ground.

VRPM Flux Calculations and Area Contributing to Flux: The average methane mass flux (g/s) was calculated for three cycle groupings of VRPM measurements using the VRPM algorithm provided in the Flux Calculator (v. 1.09 beta) software provided by ARCADIS Inc. The landfill surface that contributes to the mass flux calculated by the VRPM algorithm varies as function of wind direction, wind speed, atmospheric stability and the surface emissions rate.

The landfill surface area contributing to flux (ACF) was determined by the multiple linear regression model (MLRM) described by Thoma et al; (2009) and the approach outlined by Abichou et al; 2010 based on the ISC3 and Pasquil Stability Class Model (PSCM). For both models, data were rejected in determining unit flux rates when the wind speed was < 1 m/s or the wind angle from a vector perpendicular to the observing VRPM plane was > 30º.

Mobile and Stationary Plume Measurement with CRDS

A Picarro Model G1203 analyzer was used to measure concentrations of acetylene (tracer gas) and methane at ppb and ppt levels. The unit was mounted in an SUV fitted with an external snorkel for gas sample collection. The analyzer was integrated with a GPS (Hemisphere R100) and a compact weather station including self-aligning sonic anemometer (Climatronics AIO Compact Weather Station). Concentration, position, and meteorological data are recorded in a time-synchronized data file.

Mobile transect measurements were made by driving the analyzer along roads located around the landfill. Continuous measurements of acetylene and methane concentration are recorded as the analyzer makes transects through the plumes. The emission rate of methane is determined as the product of the release rate of the tracer and ratio of line integrals of the concentrations of acetylene and methane in the plume transects as shown in Equation 1.

$$Q_m = Q_t \Delta C_m / \Delta C_t \quad (1)$$

Where: Q_m = CH_4 emission rate

Q_t = Tracer gas release rate

ΔC_m = CH_4 observed in the plume, relative to background

ΔC_t = Tracer concentration in the plume, relative to the background

Stationary measurements were performed by positioning the analyzer downwind from the landfill and performing an extended time series of methane and acetylene concentration measurements. An assumption of this approach is that the tracer and methane plumes are of essentially the same shape at the point of measurement. The emission rate of methane is calculated by plotting the methane concentrations versus the acetylene concentrations and determining the best-fit line. The slope of this line represents the ratio of the total methane emissions to the total acetylene emissions over the period of measurement.

RESULTS AND DISCUSSION

Twenty-six chamber measurements were made on one day at Redwood landfill, while 52 chamber measurements were completed over the course of two days at Altamont landfill. The mean methane flux at Redwood was 0.018 $g/m^2/d \pm 0.019$ (standard error), while the mean fluxes were 0.125 $g/m^2/d \pm 0.013$ and 0.079 $g/m^2/d \pm 0.066$ for Altamont. Static chamber results at Redwood and Altamont are presented in Tables 2 and 3.

VRPM measurements at Redwood were made using two VRPM planes that bisected the landfill along its long axis. At Altamont, four VRPM planes were established along the top surface of the active area of the landfill. Surface methane flux rates in units of $g/m^2/d$ were calculated using the PSCM and MLRM methods described earlier. The arithmetic mean flux rates for each day of measurement at Redwood and Altamont are presented in Tables 2 and 3. The overall average PSCM and MLRM derived flux rates for Redwood were 8.3 and 14.9 $g/m^2/d$ respectively. For Altamont, study average PSCM and MLRM flux rates were 6.8 and 7.5 $g/m^2/d$. The fact that the MLRM methodology provided a consistently higher value than the PSCM approach is consistent with the observations of Goldsmith et al, (2010).

The mobile and stationary plume measurements performed at Redwood were made at or along sections of US 101 and a dirt access road south of the landfill. There was limited access to the east of the landfill which necessitated waiting for winds from the north and east of the landfill. Meteorological conditions were favorable on October 7 and mobile and stationary plume measurements returned methane flux results of 5,355 and 4,987 g/min. In order to facilitate comparisons, these values were converted to unit flux rates by dividing the emission rate by the area of the Redwood landfill and are shown in Table 2. Additional stationary measurements made the next day yielded a methane flux rate estimate of 3,432 g/min.

During the Altamont study, meteorological conditions were relatively unfavorable with low wind speeds and unstable conditions. However, better conditions prevailed on October 22 allowing mobile and stationary plume measurements to be made. Methane flux results for the mobile and stationary measurements were 9,452 and

8,704 g/min, respectively. The corresponding unit flux rates are shown in Table 3. The mobile transects of acetylene and methane plumes are shown in Figure 3.

Table 2. Summary of Chamber, VRPM and CRDS Plume Flux Results for Redwood

Site and Study Date	Chamber Flux ($g\ m^{-2}\ d^{-1}$)	VRPM (PSCM) Flux ($g\ m^{-2}\ d^{-1}$)	VRPM (MLRM) Flux ($g\ m^{-2}\ d^{-1}$)	Mobile Plume Flux ($g\ m^{-2}\ d^{-1}$)	Stationary Plume Flux ($g\ m^{-2}\ d^{-1}$)
Redwood 10/6/09	—	9.2	17.0	—	—
Redwood 10/7/09	0.018	9.4	16.7	8.5	7.9
Redwood 10/8/09	—	6.2	11.1	—	5.4

Table 3. Summary of Chamber, VRPM and CRDS Plume Flux Results for Altamont

Site and Study Date	Chamber Flux ($g\ m^{-2}\ d^{-1}$)	VRPM (PSCM) Flux ($g\ m^{-2}\ d^{-1}$)	VRPM (MLRM) Flux ($g\ m^{-2}\ d^{-1}$)	Mobile Plume Flux ($g\ m^{-2}\ d^{-1}$)	Stationary Plume Flux ($g\ m^{-2}\ d^{-1}$)
Altamont 10/20/09	—	6.9	10.2	—	—
Altamont 10/21/09	0.125	8.1	10.5	14.3	13.1
Altamont 10/22/09	0.079	5.2	7.5	—	—

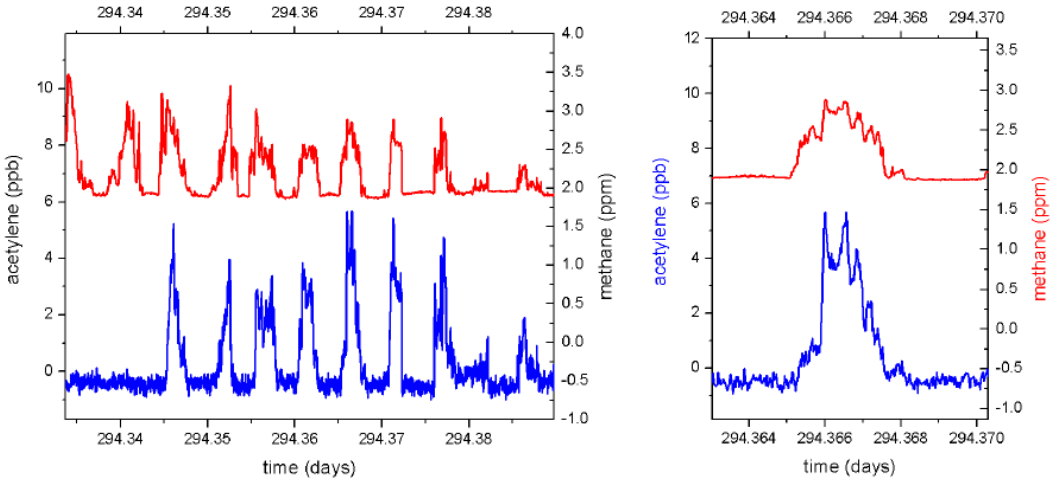

Fig. 3 Mobile transects of methane (red) and acetylene (blue) plumes at Altamont landfill on the left and a single transect on the right.

Comparison of the flux results listed in Tables 2 and 3 shows that the static chamber results were consistently and significantly lower than the VRPM or tracer plume derived values. This result is consistent with findings of other measurement method comparison studies that have included static chambers, ORS and tracer plume methods (Babilotte et al.; 2009, Goldsmith et al.; 2010). The differences between the VRPM and tracer results is generally less than a factor of two, with the tracer results indicating higher emissions at Altamont and the VRPM approach indicating higher emissions at Redwood.

CONCLUSIONS

An early stage application of CRDS to mobile and stationary plume measurements of whole landfill methane emissions using an acetylene tracer gas was successfully demonstrated. Mobile and stationary plume based measures of methane flux were approximately the same order of magnitude as VRPM derived flux rates and two orders of magnitude higher than static chamber rates. The differences in the observed rates of emission may be a function of the scale of the measurements. While understanding the factors that influence systematic and measurement errors of the plume methods requires further investigation, the method appears to offer potential advantages in this area as multiple measurements can be made and combined.

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