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Development of a mobile tracer correlation method for assessment of air emissions from landfills and other area sources

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HIGHLIGHTS

- Advancing a standardized mobile tracer correlation method for large area source measurements.
- Executed measurements on 15 landfills in 56 studies over 5 years.
- Proposed data acceptance criteria: R^2, emission rate difference, and SN ratio.
- Best TC results for landfill emissions are under moderate wind speeds at 1–3 km.
- Expected increasing trend over a three year period shown for new landfill.

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ABSTRACT

A standardized version of a mobile tracer correlation measurement method was developed and used for assessment of methane emissions from 15 landfills in 56 field deployments from 2009 to 2013. Using cavity ring-down spectroscopy and acetylene tracer gas, this method has potential implementation and cost advantages over other mobile tracer correlation approaches. The field deployment, data acquisition and analysis procedures, and a range of use conditions are discussed. To test real-world method application, the field studies were conducted by engineering technician-level personnel under randomly-encountered daytime atmospheric conditions. A total of 1876 mobile tracer correlation measurement transects were attempted over 131 field sampling days. Of these, 1366 transect (73%) were successfully completed and passed basic data acceptance criteria as valid measurement attempts. Invalid data were caused primarily by equipment failures, transect execution errors, or poor plume transport conditions. Valid transects were further analyzed using signal-to-noise ratio, plume correlation, and emission rate difference method quality indicators described here. Encountered scenarios that can result in high emission measurement uncertainty or bias are discussed in term of these indicators. Reasonable values for the acceptance levels of the method quality indicators that help protect against method errors and reduce measurement noise are discussed. The application of a default indicator set to the valid data yield 456 transects (33%) that pass data acceptance criteria. Transects that fail were associated with insufficient advected plume transport, poor correlation between the tracer and source plumes, and potential emissions pooling conditions.

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1. Introduction

Development of measurement techniques to quantify air pollutant and greenhouse gas (GHG) emissions from area sources such as landfills, waste water ponds, open-source processing, and

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Nomenclature

TC	Tracer Correlation
MQI	Method Quality Indicator
ERD	Emission Rate Difference
ROI	Region of Interest
CRDS	Cavity Ring-Down Spectrometer
CMI	Concentration Measurement Instrument
OTM	Other Test Method
SNR	Signal-to-noise ratio

agricultural operations is an important emerging environmental topic. In many cases, the spatial extent and heterogeneity of area source emissions make direct (on-site) measurements difficult to execute and interpret. Methane (CH_4) emissions from landfills, for example, can be particularly complex from a spatiotemporal standpoint making stand-off remote sensing approaches of the advected plume appealing (Babilotte et al., 2010; Thoma et al., 2009; Goldsmith et al., 2012).

Tracer correlation (TC) is a measurement approach used to characterize area source emissions through simultaneous downwind measurement of a released tracer gas and the target source compound, CH_4, in the case of landfills (Czepiel et al., 1996). With proper plume mixing conditions, the known-rate tracer release provides an indirect measure of atmospheric dispersion at the downwind observing location, allowing the area source emission rate to be determined by a ratio calculation (Borjesson et al., 2009). Although powerful in concept, the TC approach has been somewhat limited to research applications in part due to the cost and complexity of the instrumentation required to quantify the part-per-billion levels of tracer and target gas at kilometer-scale downwind observation distances.

The use of TC to assess air pollutant emission sources such as waste water impoundments, dung heaps, landfill leachate wells, and natural gas facilities has been documented (Howard et al., 1992; Fredenslund et al., 2010; Lamb et al., 1995; Shorter et al., 1996, 1997; Skiba et al., 2006). A number of studies (Table 1) have used TC to measure CH_4 emissions from landfills using a variety of tracer gases and concentration measurement instruments (CMIs). Common tracer gases include nitrous oxide (N_2O) or sulfur hexafluoride (SF_6), however, carbon monoxide (CO) has also been used. Some disadvantages to these gases include cost, impact as GHGs (N_2O, SF_6), background concentrations (N_2O, CO), or toxicity to humans and animals [e.g., CO Fredenslund et al. (2010)]. Primary CMIs used for TC measurements include Fourier transform infrared spectroscopy (FTIR), tunable diode laser absorption spectroscopy (TDLAS) based on mid-infrared quantum cascade laser technology, and electron capture detectors (ECD) for SF_6. Although generally high in detection sensitivity performance, the operational complexity, and data analysis requirements of many CMIs have generally limited their use to research applications conducted by a few expert groups.

The emerging availability of field-implementable, high-performance instrumentation is facilitating development of new fugitive and area source measurement approaches (Green et al., 2010; Thoma et al., 2009; Goldsmith et al., 2012). A growing number of companies use low cost near-infrared tunable diode lasers in conjunction with open-path or optical cell measurement approaches [e.g., cavity ring-down spectroscopy (CRDS) or some variant thereof] to produce robust instrument packages that are easy to use compared to previously described absorption spectroscopy methods. Leveraging these advances, the U.S. Environmental Protection Agency (EPA) is investigating in development of the mobile TC approach [designated EPA Category C "other test method" (OTM) 33B] as a standard method for whole-facility and area source emissions measurement. Since 2009, Waste Management (WM) Inc. and Environmental Research Education Foundation (EREF) have worked with EPA under a cooperative research and development agreement to investigate development of a large area source TC approach, with particular emphasis on landfill applications.

In this study, a mobile TC approach, based on an easy to use CRDS CMI and acetylene (C_2H_2) tracer gas, was deployed in 56 field trials (131 sampling days) to investigate practical aspects of the method and determine landfill emissions. Although C_2H_2 is flammable and must be handled with strict safety protocols, it is relatively low cost ($30 per m^3) and has generally low background interference and environmental impact compared to some other tracer gases. Field trials were conducted at 15 landfill sites in eight U.S. States consisting of a variety of landfill configurations,

Table 1
Summary of previous landfill (LF) tracer correlation studies.

Reference	CMI	Tracer	LF	Sampling time
Babilotte et al. (2010)	TDLAS	N_2O	1	10 days
Borjesson et al. (2009)	FTIR	N_2O	7	21 studies
Czepiel et al. (1996)	TDLAS/ECD	SF_6	1	5 days
Czepiel et al. (2003)	TDLAS/ECD	SF_6	1	4 studies
Galle et al. (2001)	FTIR	N_2O	1	≈6 studies, 12 days
Green et al. (2010) [a]	CRDS	C_2H_2	2	2 studies, 6 days
Jacobs et al. (2007)	TDLAS/FTIR	N_2O	2	3 studies, 3 days
Mønster et al. (2014)	CRDS/FTIR	C_2H_2	3	5 studies
Mosher et al. (1999)	TDLAS/ECD	SF_6	5	≈10 studies
Scheutz et al. (2011)	FTIR	CO/N_2O	1	3 studies, 6 days
Scheutz et al. (2013)	FTIR/CRDS	N_2O/C_2H_2	13	6 years
Other Test Method 33B[a]	CRDS	C_2H_2	15	56 studies, 131 days

[a] Current study.

Table 2
Landfill study site locations, year measurements were taken, number of studies (test days), number of transects, number of release locations (R_L) used, and tracer release points with average separation distance (D) between release points. The landfill sites with emission rate less than 6000 g/min are labeled as low emitting landfills while those greater are high.

Site code	Location	Year	Studies (days)	Transects	R_L	Avg D (m)	Low/ High
CA1	CA	2009	1 (1)	26	3	359	High
CA2	CA	2009	1 (1)	21	2	329	High
GA1	GA	2012	1 (2)	9	2	100	High
GA2	GA	2011 −2013	10 (35)	443	2	276	Low
IL1	IL	2013	3 (10)	163	3	269	Low
IN1	IN	2010 −2011	5 (9)	100	3	242	High
IN2	IN	2010 −2011	6 (8)	149	2 −3	202	Low
IN3	IN	2009 −2012	11 (24)	331	3	251	High
KS1	KS	2012 −2013	6 (18)	321	3	174	High
KY1	KY	2010	3 (5)	58	3	440	High
NM1	NM	2011	1 (2)	7	2	177	Low
OH1	OH	2010	2 (4)	41	2 −3	376	Low
OH2	OH	2010	1 (1)	20	2 −3	29	Low
OH3	OH	2010	2 (4)	48	3	144	High
OH4	OH	2010	3 (7)	139	3	97	High
15 sites	8 states	5 years	56 (131)	1876	–	–	–