Mobile monitoring of fugitive methane emissions from natural gas consumer industries

Xiaochi Zhou[1], John D. Albertson[2], Adam Gaylord[3], Joe von Fisher[3], Joe Rudek[4], Eben D. Thoma[5]

[1] Department of Civil and Environmental Engineering, Duke University, Durham, NC. [2]School of Civil and Environmental Engineering, Cornell University, Ithaca, NY. [3]Department of Biology, Colorado State University, Fort Collins, CO. [4]Environmental Defense Fund, Raleigh, NC. [5]U.S. EPA, Office of Research and Development, RTP, NC

Abstract

Natural gas is used as a feedstock for major industrial processes, such as ammonia and fertilizer production. However, fugitive methane emissions from many major end-use sectors of the natural gas supply chain have not been quantified yet. This research introduces new tools to estimate meth emission rates, and examines results from recent field measurements conducted downwind of several industrial plants using a specialized vehicle equipped with fast response methane sensor that circles around the targeted facility. Using these measurements along with the local meteorological data, a Bayesian approach is applied to probabilistically infer methane emission rates. Data from controlled tracer release experiments are presented and used to validate the approach. With access via public roads, this mobile monitoring method is able to quickly assess the emission rate of facilities. This work is developing the capacity for efficient regional coverage of potential methane emission rates in support of leak detection and mitigation efforts.

Introduction

Natural gas is considered as a bridge fuel towards clean energy due to its potential lower greenhouse gas (GHG) emissions comparing with other fossil fuels [Alvarez et al., 2012]. Natural gas is the largest source of anthropogenic emission of methane (CH_4), which is a more potent GHG than CO_2. Natural gas leaking could happen at any point along the path from production to end use, thus reducing the potential GHG advantage over competing fossil fuels such as coal.

Current literature focuses on characterizing fugitive methane emission budgets of industrial sectors such as the top-down approaches to estimate regional fluxes [Caulton et al., 2014] and the intensive experimental investigations of individual sites [Allen et al., 2013] to support an estimate of emissions from representative types of facilities. However, these efforts do not address the practical need to identify the location and strength of individual leaks in order to guide direct mitigation efforts.

This work examines the feasibility of quantifying fugitive methane emission in suburban and rural environments using a mobile sensor platform. A recently developed plume integration method [Albertson et al. 2015] is used to probabilistically infer leak rate based on Bayesian inference. Data collected from controlled release experiments is used to validate this method. Then, it is applied to estimate fugitive methane emissions from several ammonia fertilizer plants based on field data.

Field experiments

Sampling locations: Enid, OK; Woodward, OK; Borger, TX.

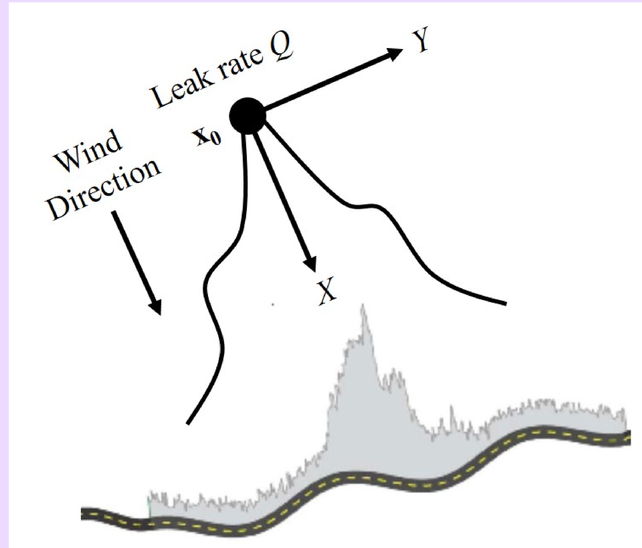
Time: Jun. 16th to 19th, 2015

Sampling platform: Google Street View car equipped with a methane analyzer based on the cavity ring-down spectrometer (Picarro Inc., Santa Clara, CA, USA)

Sampling strategy:

1. Set-up a 3-D sonic wind sensor in a nearby, relative open place
2. Driving around the targeted facility to identify possible leaks
3. Over-sampling identified plume transects by multiple traversals

Mobile sensing approach with plume integration



Consider a steady leak located at $\mathbf{x_0}$ with a leak rate of Q^T. Using a local coordinate system $F = (X, Y, Z)$ with origin at the leak source and its X axis directed along the mean wind direction. The above-ambient ensemble average plume concentration, $C(\mathbf{x})$, can be modeled using a Gaussian dispersion model [Pasquill, et al. 1983; Horst et al., 1992]

$$C(\mathbf{x}) = \frac{Q^T}{U(\bar{z})} D_y(x,y) D_z(x,z)$$

where $U(\bar{z})$ is the effective speed of plume advection. $D_y(x,y)$ and $D_z(x,z)$ are the cross-wind and vertical dispersion factors, respectively [Horst et al., 1992].

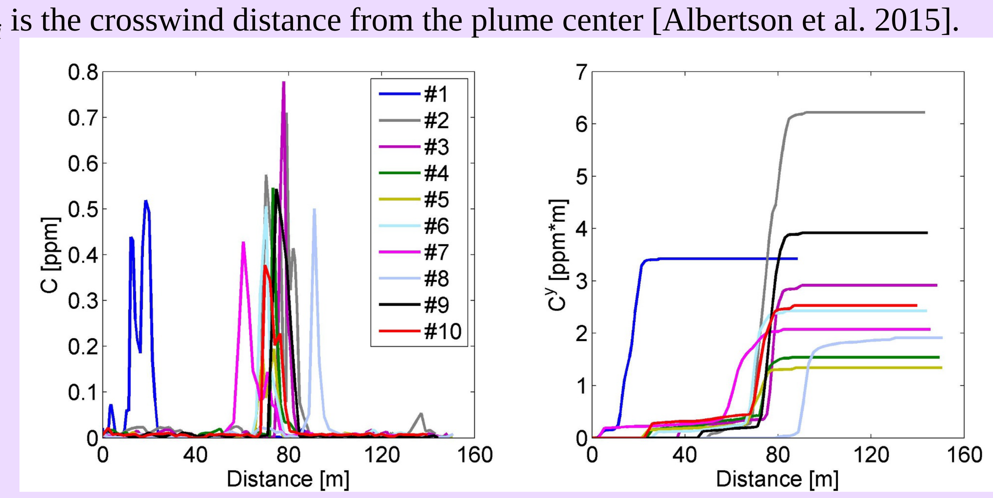
In the cases of a mobile sensor traversing a plume perpendicular to the main wind direction the integral of $D_y(x,y)$ is unity ($\int_{-\infty}^{\infty} D_y(x,y) = 1$) when $D_y(x,y)$ is formulated in both instantaneous or ensemble-average manner. This removes most of randomness caused by the lateral dispersion when dealing with instantaneous plume and reduces the previous equation as:

$$C^y = \int_{-\infty}^{\infty} C(\mathbf{x}) dy = \frac{Q^T}{U(\bar{z})} D_z(x,z)$$

where C^y is the cross plume integrated concentration. $D_z(x,z)$ can be estimated as: $D_z(x,z) = \frac{A}{\bar{z}} \exp\left(-\left(\frac{Bz}{\bar{z}}\right)^s\right)$, where s, A, and B are functions of atmospheric stability and downwind distance [Gryning et al. 1987, Foster et al. 2015]. Note that this equation is applicable only in the case when the path mobile sensor is *perpendicular* to the wind direction. In field applications, the sensor paths are typically limited by road access that may not perpendicular to the wind direction. In these cases, we apply a numerical integration of C^y [Albertson et al. 2015]:

$$C^y = \sum_{i=0}^{\infty} C(x_i)\Delta t V = Q^T \sum_{i=0}^{\infty} \frac{\Delta t V}{U(\bar{z}_i)} \left(\frac{A}{\bar{z}_i} \exp\left(-\left(\frac{Bz}{\bar{z}_i}\right)^s\right)\right) \left(\frac{\exp\left(-\frac{1}{2}\left(\frac{y_i}{\sigma_y}\right)^2\right)}{\sqrt{2\pi}\sigma_y}\right)$$

where i is a index for the sensor position, Δt is the sensor time step, V is the vehicle speed, $D_y(x,y)$ is approximated by the traditional Gaussian shape function, σ_y is the horizontal length scale of the plume, and y_i is the crosswind distance from the plume center [Albertson et al. 2015].



Following Bayes theorem, we can estimate the posterior probability density function (PDF) of the source rate Q given the cross-plume integrated concentration C^y, and the local meteorological and landscape conditions I, such as surface roughness length z_0 and atmospheric stability conditions [Yee 2010]:

$$P(Q|C^y, I) = \frac{P(Q)P(C^y|Q,I)}{P(C^y|I)}$$

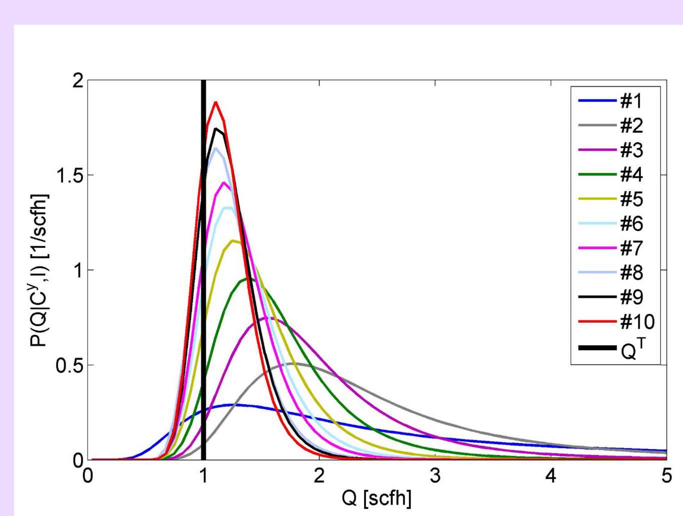
where $P(Q|C^y, I)$ and $P(Q)$ represent posterior and prior distribution of Q, respectively. $P(C^y|Q,I)$ is the likelihood function, which represents the probability of observing C^y given Q. $P(C^y|I)$ is the "evidence" which normalize $P(Q|C^y, I)$ [Albertson et al. 2015]. For a general case, we adapt a uniform distribution of Q assume that the upper (Q_{max}) and lower (Q_{min}) bounds of leak rate are known. However, after the j^{th} traversal, we update the prior with the posterior $P(Q|C^y, I)$ of the previous pass.

$$P(Q) = \begin{cases} 1/(Q_{\max} - Q_{min}) & j = 1 \\ P(Q|C^y, I)_{j-1} & j > 1 \end{cases}$$

The likelihood function can be described by a traditional Gaussian likelihood function [Yee 2010]:

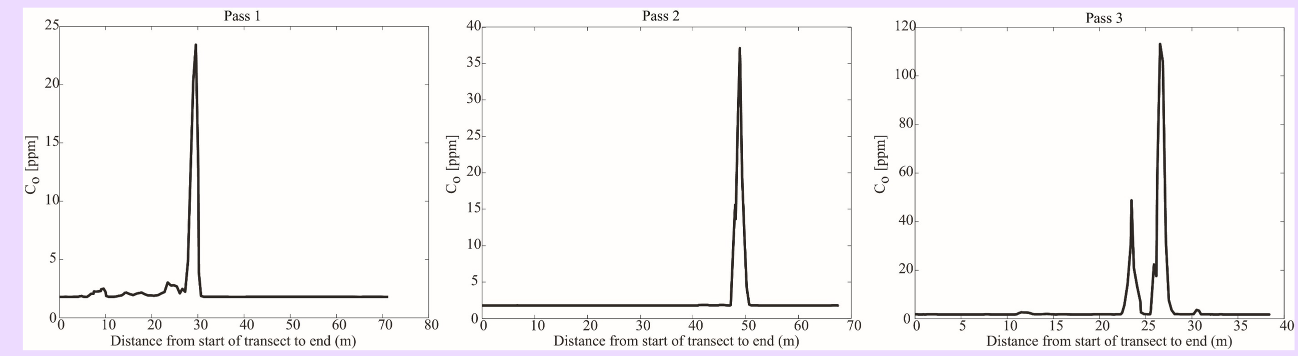
$$P(C^y|Q,I) = \frac{1}{\sqrt{2\pi}\sigma_e} exp\left(-\frac{1}{2}\left(\frac{C^y - C_M^y(Q)}{\sigma_e}\right)^2\right)$$

where $C_M^y(Q)$ is the integral of modeled cross-plume concentration for a given Q, and σ_e is combined model and measurement errors [Yee 2010].

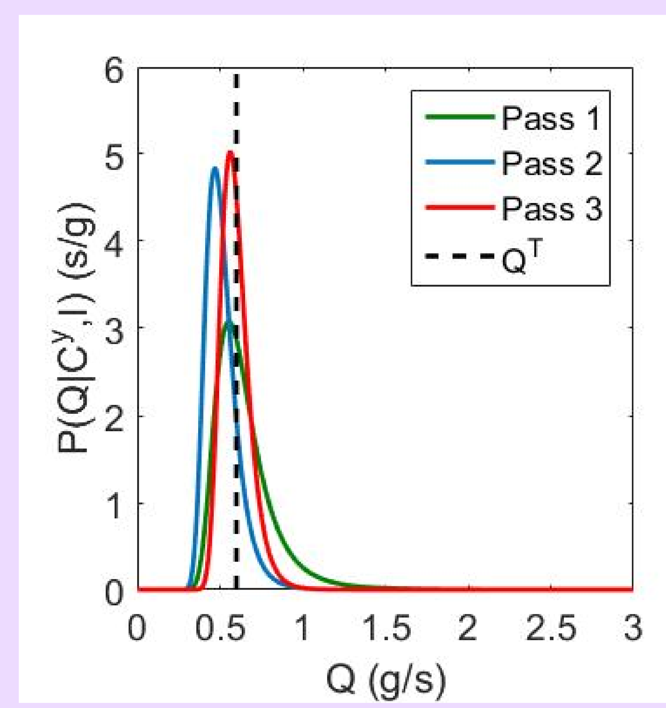


Method validation with controlled release

Using a mobile OTM 33 collection approach, data were collected for a controlled release (CR) experiment conducted on May 15, 2010 in Durham, North Carolina where three passes were made. The point source release rate was controlled at 0.6 gram/s. The passes do not always start and end in the same location, which explains why the plumes in the figure below do not always appear at the same distance along the transects.



The sensor vehicle was first parked in a nearby open area to collect meteorological data. Then, the sensor vehicle traversed the plume of elevated CH_4 concentrations multiple times. Finally, the vehicle was parked to obtained additional meteorological data. The meteorological data collected before and after the passes are used to estimate the mean wind speed, friction velocity, and stability parameter for use in the dispersion model along with the methane concentration measurements. Based on the mobile method, the PDF of leak rate can be estimated after each pass. It is encouraging that the posterior PDF peaks around the true release rates of 0.6 g/s, and the recursive updating leads to more accurate leak rate estimation and reduced uncertainty (a sharpening of the estimate) with increased sensor passes.



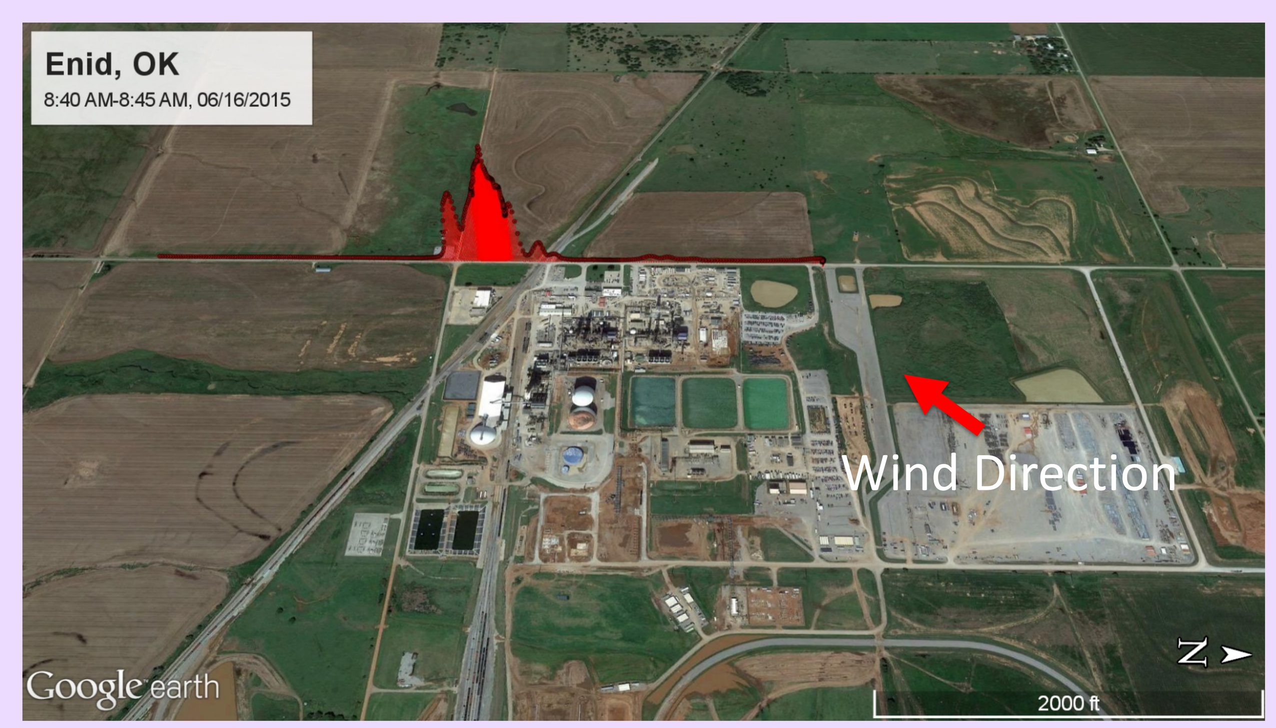
The emission rate is computed as the expected values of posterior PDF:

$$Q^E = \int_{-Q_{min}}^{Q_{max}} p(Q|C^y, I) Q dQ$$

Pass	Q^T (g/s)	Q^E (g/s)
1	0.60	0.65
2	0.60	0.51
3	0.60	0.60

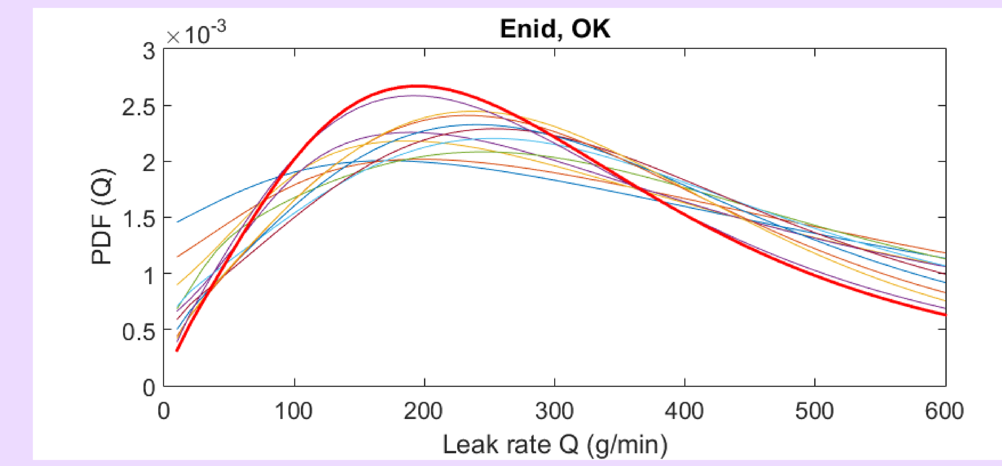
An example of plume transect in the field

Here an example plume transect is plotted in Google Earth with the red bar indicates above-ambient CH_4 concentration. At that time, the wind was blowing from northeast, which also suggests fugitive emission from the facility. We found a total of 12 plumes transects from this facility during a span of 30 minutes, during which the wind direction is almost steady. These data are then utilized to analyze the emission rate from the facility using the mobile approach.



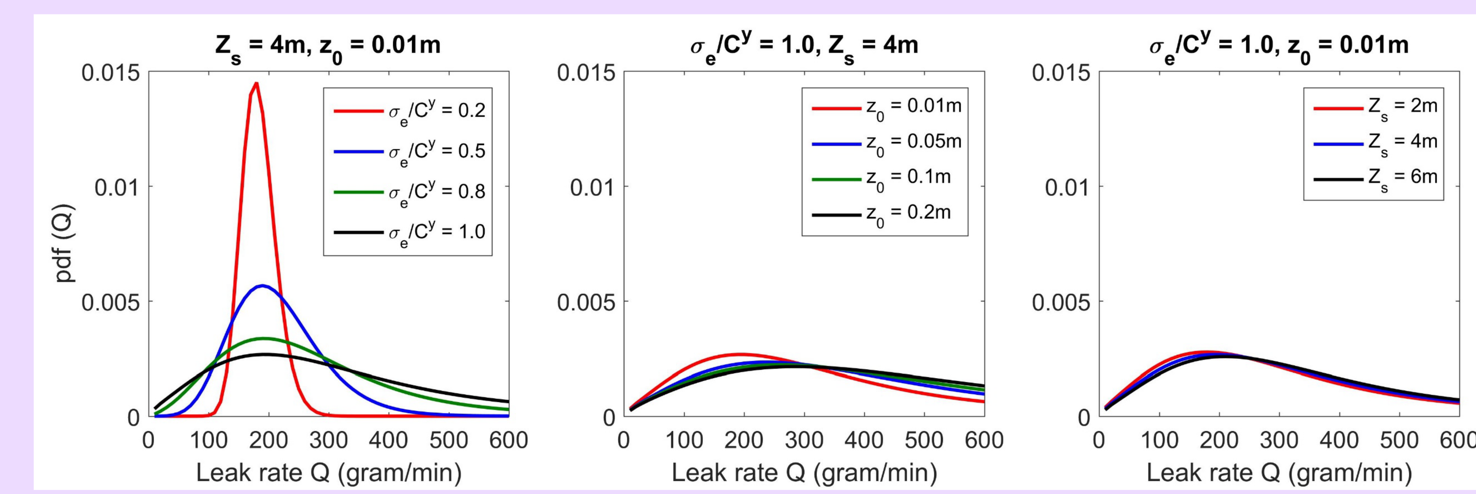
Preliminary results

We consider a case where surface roughness (z_0) is 0.01 meters (suggested by 3-D sonic data), source height (Z_s) is 4 meters (typical height of a tank), and model noise-to-signal ratio (σ_e/C^y) is 1 as a first estimate. The posterior pdf of Q was updated after each pass and the final posterior is plotted as the thick red line. Around 200 gram/min, which is 0.29 metric ton/day, of fugitive methane emission is identified from this ammonia fertilizer plant.



Sensitivity analysis

A sensitivity analysis of z_0, Z_s, and σ_e/C^y is performed here and only the posterior pdf of the final pass is plotted. It is clear that σ_e/C^y is most effective in controlling the shape of the posterior pdf comparing with z_0 and Z_s. A small σ_e/C^y indicates higher confidence thus help the inference converges faster with narrow pdf. Despite the different simulation results, the estimated emission rate is still around 200 gram/s.



Conclusion

1. Considerable success has been achieved using the mobile sensing approach for detecting fugitive methane emission in suburban and rural environments.
2. More analytical/experiment work needs to be done to quantify σ_e under varying meteorological and obstacle conditions

References

Allen, D. T., Torres, V. M., Thomas, J., Sullivan, D. W., Harrison, M., Hendler, A., Herndon, S. C., Kolb, C. E., Fraser, M. P., Hill, A. D., Lamb, B. K., Miskimins, J., Sawyer, R. F., and Seinfeld, J. H. (2013), "Measurements of methane emissions at natural gas production sites in the United States," *PNAS*, 110, 17768–17773.

Alvarez, R., Pacala, S., Winebrake, J., Chameides, W., and Hamburg, S. (2012), "Greater focus needed on methane leakage from natural gas infrastructure," *PNAS*, 109, 6435–6440.

Albertson, J. D., Harvey, T., Foderaro, G., Zhu, P., Zhou, X., Ferrari, S., Amin, M. S., Modrak, M., Brantley, H., and Thoma, E. D. (2015), "A Mobile Sensing Approach for Regional Surveillance of Fugitive Methane Emissions in Oil and Gas Production," *ES&T.* (under review).

Caulton, D. R., Shepson, P. B., Santoro, R. L., Sparks, J. P., Howarth, R. W., Ingraffea, A. R., Cambaliza, M. O., Sweeney, C., Karion, A., Davis, K. J., et al. (2014), "Toward a better understanding and quantification of methane emissions from shale gas development," *PNAS*, 111, 6237–6242.

Brantley, H., Thoma, E., Squier, W., Guven, B., and Lyon, D. (2014), "Assessment of Methane Emissions from Oil and Gas Production using Mobile Measurements," *ES&T*, 48, 14508–14515.

Foster-Wittig, T. A., Thoma, E. D., and Albertson, J. D. (2015), "Estimation of point source fugitive emission rates from a single sensor time series: a conditionallysampled Gaussian plume reconstruction," *Atm. Environ.*

Pasquill, F. and Smith, F. B. (1983), Atmospheric diffusion: study of the dispersion of windborne material from industrial and other sources, JOHN WILEY & SONS, 605 THIRD AVE., NEW YORK, NY 10016, USA.

Yee, E. and Flesch, T. (2010), "Inference of emission rates from multiple sources using Bayesian probability theory," *J. of Environ. Monit.*, 12, 622–634.

Acknowledgement

This research is supported by EDF and the Atkinson Center for a sustainable future at Cornell University. The authors thank the support from the Google Earth Outreach for help with collecting measurements from public road next to the ammonia plant. We also thank Denzil Smith and David Allen From UT Austin for their help with the sampling planning, M. Shahrooz and Mark Modrak from Arcadis who conducted the controlled release experiments, and Tierney Harvey from UCO who did the analysis of the controlled release data.