**Supplementary Information**

**Quantification and source apportionment of the methane**

**emission flux from the city of Indianapolis**

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*Determination of the relative contributions of Southside landfill (SSLF) and the Harding St. transmission regulating station using measured plumes from surface mobile measurements in Indianapolis: derivation of correction factor due to dispersion*

When winds are out of the west and the drive-route is nominally perpendicular to the wind direction (i.e. on Harding Street), the plumes from Southside landfill (SSLF) and the transmission regulating station (TRS) on Harding and Raymond intersection are distinctly separated such as the mobile surface survey data on January 21, 2013 shown in Figures 5A & 5B in the main manuscript.

To partition the combined flux from SSLF and TRS, we measure the areas under the curves of these two distinct plumes and determine their relative contributions to the total area. We assume that the relative contributions to the total area are the same as the relative contribution to the total (SSLF + TRS) flux determined from our aircraft measurements. W note, however, that the distances of the two sources from Harding Street were not the same, i.e., the TRS was ~40m while SSLF was ~1300m from Harding St. Further downwind from the source, the plume is expected to be increasingly diluted with distance both in the lateral (crosswind) and vertical directions. Thus, a correction to the relative contribution of TRS was determined by quantifying its effective plume area at 1300 m, the same distance as the landfill from Harding St. Here, we use the simple Gaussian plume model to determine the correction.

The simple Gaussian plume equation for a point source is given as:

 $χ\left(x,y,z\right)=\frac{Q}{2πUσ\_{y}σ\_{z}}exp\left[-\frac{y^{2}}{2σ\_{y}^{2}}\right]\left\{exp\left[-\frac{\left(z-H\right)^{2}}{2σ\_{z}^{2}}\right]+exp\left[-\frac{\left(z+H\right)^{2}}{2σ\_{z}^{2}}\right]\right\}$ (1)

In the relationship above,

Q: source emission rate, assumed to be constant

U: average wind speed at source height (m s-1); Note that the average wind speed is also assumed to be constant within the duration of measurement

σy, σz: are the dispersion coefficients of the concentration distribution in the crosswind and vertical directions, respectively, (m)

H: source height, (m)

x: distance downwind from the source, (m)

y: crosswind distance from the plume centerline, (m)

z: vertical distance from ground level, (m)

χ: Concentration at point (x, y, z)

We can simplify equation (1) by noting that the transmission regulating station is essentially located on the ground (so H ≈ 0). Since we are also interested in examining the centerline, ground level concentration, y = z = 0 (i.e. χ(x, y = 0,z = 0)), equation (1) becomes:

$χ\left(x,0,0\right)=\frac{Q}{2πUσ\_{y}σ\_{z}}$ (2)

We can rearrange equation (2) to show

 $χ\left(x,0,0\right)σ\_{y}σ\_{z}=\frac{Q}{2πU}$ (3)

Recall that *Q* and *U* are assumed to be constant, and as such, the right-hand-side of equation (3) is a constant. Thus, upon examination of the left-hand-side of equation (3), we note that the centerline concentration (χ(x, 0, 0)) is not constant with downwind distance, and neither are the dispersion coefficients of the concentrations. The dispersion coefficients increase with diffusion time, or distance from the source, and the rates of growth depend on the meteorological conditions. Thus, the areas under the curves, that is, the integrated concentrations are also not constant with downwind distance. In particular, the plume is expected to be further diluted with distance from the source. Based on the logic presented in equation (3), the correction applied to determine the effective area under the curve for the TRS at 1300 m (the approximate downwind distance of SSLF from Harding Street) is given as:

$A\_{2}=A\_{1}\frac{\left(x\_{1}\right)σ\_{y}\left(x\_{1}\right)σ\_{z}\left(x\_{1}\right)}{\left(x\_{2}\right)σ\_{y}\left(x\_{2}\right)σ\_{z}\left(x\_{2}\right)}$ (4)

In Equation (4), A1 is the integrated concentration at downwind distance x1 (that is, the area under the methane enhancement versus latitude curve in Figure 5B, ppb-deg), and σy(x1), σz(x1) are the lateral and vertical dispersion coefficients at x1; the same definitions hold for A2, σy(x2), σz(x2) but for downwind distance x2.

There are many formulations for the dispersion coefficients (e.g. Passquill-Gifford (P-G) dispersion diagrams). We used here the formulation by Briggs (1973), since Briggs modified the P-G curves and fitted them with numerical expressions that are useful for computations. Briggs also presented different formulations for open country conditions and urban conditions.

During the 21 January 2013, 15 October, and 12 November 2012 surface mobile measurements, the wind direction was westerly and the wind speeds were > 6 m s-1. The stability conditions for those three days were neutral (stability class D). The expressions for the standard deviations in the lateral and vertical directions under urban conditions are (Briggs 1973):

$σ\_{y}=0.16x\left(1+0.0004x\right)^{-1/2}$ (5)

$σ\_{z}=0.14x\left(1+0.0003x\right)^{-1/2}$ (6)

We find that the average citywide CH4 emission was 135 ± 58 mol s-1 from five flight experiments in 2011, from which we determined the combined flux from SSLF and TRS to be 45 ± 14 mol s-1. To further partition the contributions from the two sources, we applied equation (4) to TRS to determine the effective area at 1300 m. We then calculated the relative contributions of the two sources to the total area, to determine their relative source strengths. We find that the contribution from TRS was negligible relative to the landfill, with an average contribution of only 0.05%. Thus, the measured emission flux of 45 mol s-1 was essentially entirely attributable to the landfill.

*Correction for the contribution of propane in vehicle exhaust to the observed propane enhancement in flask to obtain the natural gas propane component*

We assume that the observed enhancement of propane in flask samples ([ΔC3H8]*obs*) is due to the combined contribution of propane from vehicle exhaust ([ΔC3H8]*exhaust*) and from the natural gas distribution system ([ΔC3H8]*NG*). Thus, the propane in natural gas is given by

$\left[ΔC\_{3}H\_{8}\right]\_{NG}=\left[ΔC\_{3}H\_{8}\right]\_{obs}-\left[ΔC\_{3}H\_{8}\right]\_{exhaust}$ (7)

To obtain the propane contribution from vehicle exhaust ([ΔC3H8]*exhaust*), we used the observed enhancement of acetylene ([ΔC2H2]*obs*, a tracer of vehicle emission) and the emission factor (C3H8/C2H2 ratio) derived from chassis dynamometer emissions measurements of light duty vehicles by Pang et al. (2014) as shown in equation (8)

 $\left[ΔC\_{3}H\_{8}\right]\_{exhaust}=\left[ΔC\_{2}H\_{2}\right]\_{obs}\left(\frac{C\_{3}H\_{8}}{C\_{2}H\_{2}}\right)\_{exhaust}$ (8)

where the following were the vehicle emissions data (Pang et al., 2014)

C3H­8: 0.5 ± 0.5 mg mile-1

C2H2: 6.6 ± 4.2 mg mile-1

These figures are equivalent to the following emissions in moles/mile:

C3H8: $\frac{0.0005 \frac{g}{mile}}{44\frac{g}{mole}}=1.136 × 10^{-5} \frac{mole}{mile}$

C2H2: $\frac{0.006 \frac{g}{mile}}{26\frac{g}{mole}}=2.3 × 10^{-4} \frac{mole}{mile}$

yielding a (C3H8/C2H2) emission ratio of 0.049 moles C3H8 per mole of C2H2 from vehicle exhaust.

*Estimation of the energy equivalent of Southside landfill methane emission flux*

The mean CH4 emission flux from Southside landfill was estimated to be 45 mol s-1. Using the ideal gas law and assuming T = 298 K, and P = 1 atm,

$V=\frac{nRT}{P}=\frac{\left(45 \frac{moles}{s}\right)\left(0.0821 \frac{L atm}{mol K}\right)\left(298 K\right)}{1 atm}=1100\frac{L}{s}=39\frac{ft^{3}}{s}$

For natural gas, the energy generated per unit of fuel used is 127 kWh per 1000 cubic feet (US Energy Information Administration, http://www.eia.gov/tools/faqs/faq.cfm?id=667&t=6, accessed 18 Oct 2013). Thus, the 45 mols s-1 of CH4 has an energy equivalent of

$E\_{eq}=39\frac{ft^{3}}{s}∙\frac{127 kWh}{1000 ft^{3}}=4.95\frac{kWh}{s}=1.6×10^{8}\frac{kWh}{yr}$

In the state of Indiana, the average 2011 electricity consumption for a residential utility customer was 11,960 kWh, which is equivalent to an average of 997 kWh per month (U.S. Energy Information Administration, www.eia.gov/electricity/sales\_revenue\_price/xls/table5\_a.xls, accessed 01 September 2014). Thus,

$\frac{1.6×10^{8}\frac{kWh}{yr}}{11960\frac{kWh}{yr-household}}=13000 households $

**Table S1. Summary of CALMIM 5.4 inputs. To model emissions from the daytime open face, we assumed a final cover profile (as underlying waste was fully methanogenic) overlain by 6” sand. Nighttime daily cover emissions as shown in table.**

1. CLF, Shelby County (Footprint: 80 acres)

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cover Type | % Area | % Gas Recovery | % Vegetation Present | Organic Matter Content | Material & Thickness |
| Daily | 0.2 | 0 | 0 | Low | 6” clay |
| Intermediate | 48.8 | 0 | 80 | Medium  | 36’ clay |
| Final | 50 | 0 | 95 | Medium | 6” top soil over 48” clay |

1. NCLF, Newton County (Footprint: 196.4 acres)

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cover Type | % Area | % Gas Recovery | % Vegetation Present | Organic Matter Content | Material & Thickness |
| Daily | 15 | 25 | 0 | Low | 6” clay |
| Intermediate | 60 | 90 | 50 | Medium | 36” clay |
| Final | 25 | 100 | 100 | Medium | Multi-layered\* |

\* 6” top soil + 2.5’ clay + 8 oz double-sided geocomposite + 40 mil LLDPE geomembrane + 12” compacted clay liner + 12” clay structural fill

1. RFLF, Randolph County (Footprint: 112.4 acres)

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cover Type | % Area | % Gas Recovery | % Vegetation Present | Organic Matter Content | Material & Thickness |
| Daily | 1 | 0 | 0 | Low | 6” clay |
| Intermediate | 66 | 75 | 80 | Medium | 36” clay |
| Final | 33 | 100 | 100 | Medium | 6” top soil over 48” clay  |

1. SSLF, Marion County (Footprint: 228 acres)

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cover Type | % Area | % Gas Recovery | % Vegetation Present | Organic Matter Content | Material & Thickness |
| Daily | 2 | 0 | 0 | Low | 6” small tire shreds |
| Intermediate | 85 | 90 | 80 | Medium | 36” clay |
| Final | 13 | 100 | 100 | Medium | 6” top soil (loam) over 48” clay  |

1. TBLF, Hendricks County (Footprint: 173 acres)

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cover Type | % Area | % Gas Recovery | % Vegetation Present | Organic Matter Content | Material & Thickness |
| Daily | 4 | 0 | 0 | Low | See note below\* |
| Intermediate | 39 | 100 | 0 | Medium | 36” clay |
| Final | 57 | 100 | 100 | Medium | See note below+ |

\* Two Daily covers: 6” clay and “extended daily cover” of 31” loamy sand; each 2% of area

+ Two Final covers: 48” silty clay loam (34% of area) and geomembrane composite cover (23% of area) of 12” loam over HDPE geomembrane over 24” silty clay loam.

**Table S2. List of towns surrounding Indianapolis that are also served by SSLF.**

|  |  |  |
| --- | --- | --- |
| Town | County | 2010 Population (US Census Bureau, http://www.census.gov/) |
| Avon | Hendricks | 12446 |
| Brownsburg | Hendricks | 21285 |
| Greenwood | Johnson | 49791 |
| Moorseville | Morgan | 9326 |
| Carmel | Hamilton | 79191 |
| Noblesville | Hamilton | 51969 |
| Zionsville | Boone | 14127 |

Supplemental Figure Captions

**Figure S1. Downwind CH4 horizontal transect distribution as a function of altitude for 30 June 2011.** The black broken lines represent the boundary of the project city width on the downwind horizontal transects. The –x to +x city limits are -38 km to +21 km.

**Figure S2. Measurement flight path on 01 March 2011 color coded by the observed methane concentration.** The plot in (A) shows the entire flight path including the downwind horizontal transpects while the image in (B) shows the close up view of a section of the flight path as the airplane flew upwind to interrogate the source of observed enhancement in the downwind horizontal transect.

**Figure S3. CALMIM modeled monthly CH4 emissions with standard deviations for the five Indiana landfills.** The seasonal trends of the modeled CH4 emissions for both the oxidized and un-oxidized cases are shown for (A) CLF, (B) NCLF, (C) RFLF, (D) SSLF, and (E) TBLF.

**Figure S4. Observed propane versus acetylene enhancements obtained from flask measurements downwind of the city of Indianapolis.** Also shown are the C3H8:C2H2 emission ratios from Los Angeles city (Borbon et al., 2013), and from New York City and Boston (Warneke et al., 2007). These emission ratios are not statistically significantly different from the slope of the regression line obtained for Indianapolis at the 95% confidence interval. The solid lines represent the lower and upper 95% confidence limits.