Methane Emissions from the Baltimore-Washington Area: Airborne Observations and Comparison with Inventories

Russ Dickerson
Xinrong Ren, Hao He, Jonathan Hansford, Doyeon Ahn, Ross Salawitch,
The University of Maryland
Olivia Salmon and Paul Shepson (Purdue Univ.)

Outline

• Aircraft measurements and mass balance approach for CH$_4$ emissions
• Comparison with CH$_4$ Emission inventories
• CH$_4$ emissions based on CO & CO$_2$ inventories and observed CH$_4$/CO & CH$_4$/CO$_2$ ratios
Close collaboration with NIST & MDE.

- CO$_2$ is the big climate forcing factor.
- Initial results indicate emissions inventories for CO$_2$ are good to within 10-15%.
- It is not possible to do better with current technology, but we’re working on improvements.
- Methane (CH$_4$) is harder to quantify.
- There are many sources of methane natural gas delivery systems, waste water treatment, agriculture, and landfills.
- UMD is working with MDE to improve CH$_4$ emissions estimates.
UMD Cessna & Purdue Duchess Research Aircraft

**UMD Cessna**
- Gas Inlet
- Aerosol Inlet
- Met Sensors

**Purdue Duchess**
- CRDS pump (behind)
- Calibration box
- CRDS
- Ozone monitor
- Wind computer

**GPS Position** (Lat, Long, Altitude)
- **Met** (T, RH, P, wind speed/direction)

**Trace gases:**
- \( \text{O}_3 \): UV Absorption, modified TECO
- \( \text{SO}_2 \): Pulsed Fluorescence, modified TECO
- \( \text{CH}_4/\text{CO}/\text{H}_2/\text{O} \): Cavity Ringdown, Picarro
- \( \text{NO}_2 \): Cavity Ring Down, Los Gatos
- NO: Chemiluminescence, modified TECO
- VOCs: grab canisters/GC-FID

**Aerosol Optical Properties:**
- Scattering: \( b_{\text{scat}} @ 450, 550, 700 \text{ nm} \), Nephelometer
- Absorption: \( b_{\text{ap}} (565 \text{ nm}) \), PSAP
- Black Carbon: Aethalometer

**GPS Position** (Lat, Long, Altitude)
- **Met** (T, RH, P, 3-D wind by BAT)

**Trace gases:**
- \( \text{O}_3 \): UV Absorption, 2B Technology
- \( \text{CH}_4/\text{CO}_2 \): Cavity Ring Down, Picarro
- \( \text{NO}_2 \): Cavity Ring Down, Los Gatos
FLAGG-MD Flights during Winter 2015 and 2016
Different Flight Patterns in Winter 2015 and 2016

Flights on 2/19/2015

FLAGG-MD 2015: Both aircraft did level transects

Flights on 2/19/2016

FLAGG-MD 2016:
- The Cessna did continuous vertical profiles downwind.
- The Duchess did upwind survey & level transects downwind.
Mass Balance Approach to Estimate Emission Rates

Mass Balance Experiment (MBE) approach:

\[ \Delta z = \frac{Z_i - \Delta x}{2} \]

What comes out of the box minus what went in is the flux.

- \([C] : \) concentrations (downwind)
- \([C]_b : \) concentration in background
- \(U_\perp : \) perpendicular wind speed

\( \Delta x \) and \( \Delta z \) represent the changes in position along the x and z axes, respectively.

Background CO\(_2\), CH\(_4\), CO

Urban CO\(_2\), CH\(_4\), CO

\( \Delta x \) and \( \Delta z \) are used to calculate the flux of gases out of the box.

What comes out of the box minus what went in is the flux.
# Estimated GHG Emissions from the Baltimore-Washington Area

## FLAGG-MD winter 2015

<table>
<thead>
<tr>
<th>Flight Date</th>
<th>Flux(CO$_2$) (moles s$^{-1}$)</th>
<th>Flux(CH$_4$) (moles s$^{-1}$)</th>
<th>Flux(CO) (moles s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/6/15</td>
<td>94,500</td>
<td>557</td>
<td>521</td>
</tr>
<tr>
<td>2/13/15</td>
<td>71,000</td>
<td>290</td>
<td>281</td>
</tr>
<tr>
<td>2/18/15</td>
<td>91,200</td>
<td>795</td>
<td>--</td>
</tr>
<tr>
<td>2/19/15</td>
<td>156,800</td>
<td>932</td>
<td>567</td>
</tr>
<tr>
<td>2/20/15</td>
<td>118,100</td>
<td>518</td>
<td>753</td>
</tr>
<tr>
<td>2/23/15</td>
<td>107,900</td>
<td>641</td>
<td>417</td>
</tr>
<tr>
<td>2/24/15</td>
<td>110,300</td>
<td>476</td>
<td>640</td>
</tr>
<tr>
<td>2/25/15</td>
<td>108,500</td>
<td>602</td>
<td>571</td>
</tr>
<tr>
<td>2/27/15</td>
<td>78,800</td>
<td>540</td>
<td>--</td>
</tr>
<tr>
<td>Mean±1σ</td>
<td>104,000 ±25,000</td>
<td><strong>595±184</strong></td>
<td>536±152</td>
</tr>
</tbody>
</table>

## FLAGG-MD winter 2016

<table>
<thead>
<tr>
<th>Date</th>
<th>Flux(CO$_2$) (moles s$^{-1}$)</th>
<th>Flux(CH$_4$) (moles s$^{-1}$)</th>
<th>Flux(CO) (moles s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>02/08/16</td>
<td>73,200</td>
<td>418</td>
<td>430</td>
</tr>
<tr>
<td>02/12/16</td>
<td>93,000</td>
<td>350</td>
<td>510</td>
</tr>
<tr>
<td>02/17/16</td>
<td>107,800</td>
<td>1,078</td>
<td>365</td>
</tr>
<tr>
<td>02/18/16</td>
<td>98,100</td>
<td>373</td>
<td>611</td>
</tr>
<tr>
<td>02/19/16</td>
<td>142,800</td>
<td>722</td>
<td>688</td>
</tr>
<tr>
<td>Mean±1σ</td>
<td>103,000 ±25,600</td>
<td><strong>588±312</strong></td>
<td>521±131</td>
</tr>
</tbody>
</table>

Mean CH$_4$ emission rate: 592 ± 248 moles s$^{-1}$

600 moles/s ~ 30,000 tons/yr
CH₄ Emissions from the SW Marcellus

New Regulations on finishing appear to have improved emissions.

<table>
<thead>
<tr>
<th>Date</th>
<th>CH₄ E.R. (moles s⁻¹)</th>
<th>CH₄ E.R. (ppbv)</th>
<th>PBL Height (m AGL)</th>
<th>WD (deg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8/25/15</td>
<td>2,200</td>
<td>2,391</td>
<td>2,287 ± 120</td>
<td></td>
</tr>
<tr>
<td>8/29/15</td>
<td>1,950</td>
<td>2,315</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9/14/15</td>
<td>1,500</td>
<td>2,156</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

From Marcellus (55x77 km): 2,287 ± 120 moles CH₄ s⁻¹

From Balt-DC (75x95 km): 592 ± 248 moles CH₄ s⁻¹
Example Point Source: CH$_4$ from Brown Station Landfill

Measured 21 times over 7 flights.

<table>
<thead>
<tr>
<th>Flight #</th>
<th>CH$_4$ Emission Rate (moles s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RF1</td>
<td>57.4</td>
</tr>
<tr>
<td>RF2</td>
<td>55.7</td>
</tr>
<tr>
<td>RF4</td>
<td>51.8</td>
</tr>
<tr>
<td>RF5</td>
<td>64.7</td>
</tr>
<tr>
<td>RF6</td>
<td>65.3</td>
</tr>
<tr>
<td>RF7</td>
<td>36.0</td>
</tr>
<tr>
<td>RF8</td>
<td>69.1</td>
</tr>
</tbody>
</table>

---

EPA GHGRP CH$_4$ emission rate for this landfill: 15.5 moles s$^{-1}$

A factor of 3.7 higher than the value in GHGRP
## Mass Balance vs. Inventories

<table>
<thead>
<tr>
<th>Landfill</th>
<th>Mass Balance Flux Range** (moles CH₄/s) - Average of all transects</th>
<th>2015 EPA (moles CH₄/s)</th>
<th>2014 MDE (moles CH₄/s)</th>
<th>2012 Maasakkers (moles CH₄/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Brown Station (21/7)</td>
<td>24.6 – 64.0</td>
<td>3.44 or 15.5*</td>
<td>6.14</td>
<td>8.52</td>
</tr>
<tr>
<td>Eastern Sanitary (3/1)</td>
<td>9.9 – 49.5</td>
<td>4.54</td>
<td>6.85</td>
<td>10.01</td>
</tr>
<tr>
<td>Quarantine Road (7/3)</td>
<td>1.35 – 2.94</td>
<td>3.33</td>
<td>17.6</td>
<td>3.11</td>
</tr>
<tr>
<td>Harford Waste (2/2)</td>
<td>2.22 – 8.50</td>
<td>5.42</td>
<td>3.52</td>
<td>5.52</td>
</tr>
<tr>
<td>Reichs Ford (3/2)</td>
<td>12.8 – 26.2</td>
<td>5.88</td>
<td>1.83</td>
<td>4.96</td>
</tr>
<tr>
<td>Route 40 West (3/3)</td>
<td>2.66 – 10.01</td>
<td>6.89</td>
<td>10.23</td>
<td>6.70</td>
</tr>
</tbody>
</table>

* EPA GHGRP requires landfills with a gas collection system (Brown Station has a gas collection system) to estimate their emissions in two ways. Typically, the higher of the two results is reported as the “official” value, but in Brown Station’s case, the lower number (3.44) was reported instead of the higher number (15.5).

**Flux range computed by varying horizontal transect width by 10% and PBL height by one-sigma.
New gridded US CH₄ NEI for 2012 (0.1° x 0.1°)

Gridded National Inventory of U.S. Methane Emissions

Joannes D. Maasakkers,†,‡ Daniel J. Jacob,† Melissa P. Sulprizio,†,‡ Alexander J. Turner,† Melissa Weitz,‡ Tom Wirth,‡ Cate Hight,‡ Mark DeFigueiredo,‡ Mausami Desai,‡ Rachel Schmeltz,‡ Leif Hockstad,‡ Anthony A. Bloom,‖ Kevin W. Bowman,‖ Seongeun Jeong,§ and Marc L. Fischer§

Gridded EPA Inventory for 2012

Includes all methane emissions included in the National Greenhouse Gas Inventory.

196 moles CH₄/s from Balt-DC
CH$_4$ Emissions from Sources in Balt-DC in CH$_4$ NEI 2012

- Landfills are a major CH$_4$ source.
- CH$_4$ emissions from NG system may be under-estimated.

<table>
<thead>
<tr>
<th>Source</th>
<th>Percentage</th>
<th>Emissions Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Landfills</td>
<td>49%</td>
<td>1.54 kg/s</td>
</tr>
<tr>
<td>EntericFermentation+Manure</td>
<td>12%</td>
<td>0.39 kg/s</td>
</tr>
<tr>
<td>NG Transmission</td>
<td>10%</td>
<td>0.30 kg/s</td>
</tr>
<tr>
<td>NG Distribution</td>
<td>11%</td>
<td>0.36 kg/s</td>
</tr>
<tr>
<td>Combustion</td>
<td>8%</td>
<td>0.24 kg/s</td>
</tr>
<tr>
<td>Waste Water Treatment</td>
<td>8%</td>
<td>0.24 kg/s</td>
</tr>
<tr>
<td>Others</td>
<td>2%</td>
<td>0.07 kg/s</td>
</tr>
</tbody>
</table>
An alternative approach – CO$_2$ and CO emissions well constrained and we can use ratios to learn about the methane flux.
CH\textsubscript{4} Emission Estimate using CH\textsubscript{4}/CO Ratio and CO Emissions

FLAGG-MD winter 2015

- Total CO emissions from Balt-DC in EDGAR v4.3 2010: 0.459 MMtons/yr

- The total CH\textsubscript{4} emissions based on CH\textsubscript{4} to CO ratio: 642 moles CH\textsubscript{4} s\textsuperscript{-1} (436 – 1,003 moles CH\textsubscript{4} s\textsuperscript{-1})
CH₄ Emission Estimate using CH₄/CO₂ Ratio and CO₂ NEI 2014

FLAGG-MD winter 2016

- Total CO₂ emissions from Balt-DC in NEI2014: 89.0 MMtons/yr
- The total CH₄ emissions based on CH₄ to CO₂ ratio: 583 moles CH₄ s⁻¹ (293 – 1,029 moles CH₄ s⁻¹)
CO to CO₂ ratio: Observed vs. EDGAR Emissions

- Observed CO to CO₂ molar ratio = 0.53% (black line)
- CO to CO₂ molar ratio in EDGAR 2010 emission inventory: 0.59% (red dashed line)
CH$_4$ Emissions from the Balt-DC Area: Top-down vs. Bottom-up

CH$_4$ Emissions (moles/s)

Top-down & Bottom-up

CH$_4$ Emissions from the Balt-DC Area: Top-down vs. Bottom-up
Summary

• UMD is working with MDE to fine tune emissions inventories. CO$_2$ is the big player with CH$_4$ ~10% of total.

• Estimated total emissions of CH$_4$ from Balt/Wash area:
  595±184 moles CH$_4$ s$^{-1}$ in winter 2015
  588±312 moles CH$_4$ s$^{-1}$ in winter 2016 (~30,000 tons/yr)

• Major CH$_4$ sources in the area: landfills and broadly, NG system.

• Direct observations of CH$_4$ emissions 1.4 to 3 times higher than inventories.

• Only flew in winter so far; need summer flights.
Extra Slides
UMD is working with MDE to fine tune emissions inventories. CO2 is the big player.

Only flew in winter so far; need summer flights.

Estimated total emissions of CH$_4$ from Balt/Wash area:
- 595±184 moles CH$_4$ s$^{-1}$ in winter 2015
- 588±312 moles CH$_4$ s$^{-1}$ in winter 2016 (~30,000 tons/yr)

Major CH$_4$ sources in the area: landfills and broadly, NG system.

Compared to CH$_4$ emission inventories:
1. Observed CH$_4$ emissions are higher than the US NEI 2012 by a factor of 3
   higher than the state EI by a factor of ~2
   higher than the EDGAR 2010 by a factor of 1.4.
2. Observed CH$_4$ emissions is similar to CH$_4$ emissions inferred from CO and CO$_2$
   NEI with observed CH$_4$/CO and CH$_4$/CO$_2$ ratios.
The total CH$_4$ emissions in the yellow rectangle (an approximately surveyed area) is 421 moles CH$_4$ s$^{-1}$.

Issues with EDGAR emissions: mainly allocated based on population instead of source locations.
### Mass Balance – CH$_4$ from DC/Baltimore

<table>
<thead>
<tr>
<th>Flight Date</th>
<th>Emission Rate (moles CH$_4$ s$^{-1}$)</th>
<th>Emission Uncertainty Range – Horizontal Bounds* (moles CH$_4$ s$^{-1}$)</th>
<th>Flux Uncertainty Range – PBL Depth** (moles CH$_4$ s$^{-1}$)</th>
<th>Flux Uncertainty Range – Horizontal Bounds &amp; PBL Depth Combined*** (moles CH$_4$ s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2/13/15</td>
<td>193</td>
<td>177 – 201</td>
<td>145 – 241</td>
<td>133 – 251</td>
</tr>
<tr>
<td>2/19/15</td>
<td>1260</td>
<td>1230 – 1270</td>
<td>853 – 1670</td>
<td>830 – 1680</td>
</tr>
<tr>
<td>2/20/15</td>
<td>509</td>
<td>452 – 509</td>
<td>351 – 667</td>
<td>311 – 667</td>
</tr>
<tr>
<td>2/23/15</td>
<td>157</td>
<td>141 – 169</td>
<td>110 – 204</td>
<td>99.0 – 219</td>
</tr>
<tr>
<td>2/24/15</td>
<td>951</td>
<td>857 – 1180</td>
<td>705 – 1200</td>
<td>633 – 1490</td>
</tr>
<tr>
<td>2/25/15</td>
<td>183</td>
<td>172 – 187</td>
<td>97.5 – 269</td>
<td>90.6 – 274</td>
</tr>
<tr>
<td>Mean</td>
<td>542</td>
<td>505 – 586 (-6.8% / +8.1%)</td>
<td>377 – 709 (-30.% / + 31%)</td>
<td>349 – 764 (-36% / +41%)</td>
</tr>
</tbody>
</table>

*Flux range computed by varying horizontal transect width by 10%

**Flux range computed by varying PBL height by one-sigma

***Flux range computed by varying horizontal transect width by 10% and PBL height by one-sigma.
Method

A variational data assimilation method is used to find the sources with which HYSPLIT would reproduce concentrations that match best with the observations.

Assumptions

• The two landfills causes for the measured excess CH$_4$.
• The CH$_4$ emissions of the landfills vary each hour.

Two major limitations

• There are other sources that contribute to the excess CH$_4$.
• Meteorological fields in the HYSPLIT dispersion model has uncertainties.

Ongoing work

• To add more sources + constant emissions from landfills
• To use ensemble runs to include uncertainties of meteorological fields
CH₄ Emission Estimate using CH₄/CO₂ Ratio and CO₂ NEI 2014

- Total CO₂ emissions from Balt-DC in NEI2014: **89.0 MMtons/yr**

- The total CH₄ emissions based on CH₄ to CO₂ ratio: **6.6 kg CH₄ /s** (3.5 – 11.6 kg CH₄/s)

- Cold winter in 2015 ➔ expected more CO₂ emissions due to heating
  ➔ A larger inferred CH₄ emission rate
CH₄ Emission Estimate using CH₄/CO₂ Ratio and CO₂ NEI 2014

FLAGG-MD winter 2015

- Total CO₂ emissions from Balt-DC in NEI2014: 89.0 MMtons/yr

- The total CH₄ emissions based on CH₄ to CO₂ ratio: 6.6 kg CH₄/s (3.5 – 11.6 kg CH₄/s)

- Cold winter in 2015 ➔ expected more CO₂ emissions due to heating ➔ A larger inferred CH₄ emission rate
CH₄ Emission Estimate using CH₄/CO Ratio and CO Emissions

FLAGG-MD winter 2016

- Total CO emissions from Balt-DC in EDGAR v4.3 2010: 0.459 MMtons/yr
- The total CH₄ emissions based on CH₄ to CO ratio: 11.8 kg CH₄ /s (6.2– 21.4 kg CH₄/s)
Observed $C_2H_6$-to-$CH_4$ Ratio

- Mean $C_2H_6$-to-$CH_4$ ratio in natural gas of Baltimore Gas & Electric: 0.1045
- Other major $CH_4$ sources (landfills, enteric fermentation, waste water treatment) has little ethane emissions.
- Emissions from the NG system account for 32% (2015) and 41% (2016) of total $CH_4$ emissions.
CH$_4$ Emission Estimate from Unaccount-for NG

- Total NG delivered to the Balt-DC area in February 2015: 54,495 million CF in Feb. 2015.

- Lost & unaccounted-for (LAUF) NG: 3.34% of total NG delivered (PHMSA data)

- Total lost & unaccounted for (LAUF) gas: 12.9 kg CH$_4$/s

- Not all LAUF gas is leaked into the atmosphere because besides leaks, unaccounted-for gas is also due to gas theft, accounting & meter errors, etc.

- The worth of the LAUF gas = 54,495 x 10$^6$ CF x 3.34% x $0.012$/CF
  = $22 M in Feb. 2015
Facility level CH$_4$ Emissions in EPA’s GHGRP (excluding emissions from Petroleum & NG System)

- Mostly landfills
- The total CH$_4$ emissions (other than the NG system) from the yellow rectangle is **1.21 kg/s**

Based on the unaccounted-for natural gas and other CH$_4$ sources in GHGRP, the total CH$_4$ emission rate from the Balt-DC area:

$$12.86 + 1.21 = 14.07 \text{ kg/s}$$

This may overestimate CH$_4$ emissions since not all LAUF gas is emitted into the atmosphere.
## Brown Station – Mass Balance Calculations

<table>
<thead>
<tr>
<th>RF</th>
<th>Mass Balance Flux (moles / s) (Mean of flux calculated on each individual transect)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>30.7</td>
</tr>
<tr>
<td>2</td>
<td>44.7</td>
</tr>
<tr>
<td>4</td>
<td>31.9</td>
</tr>
<tr>
<td>5</td>
<td>59.9</td>
</tr>
<tr>
<td>6</td>
<td>21.1</td>
</tr>
<tr>
<td>7</td>
<td>71.8</td>
</tr>
<tr>
<td>8</td>
<td>66.3</td>
</tr>
<tr>
<td><strong>Mean ± St. Deviation</strong></td>
<td><strong>46.6 ± 18.3</strong></td>
</tr>
</tbody>
</table>
CH$_4$ emissions from King George Landfill

**Map:**
- Longitude (°)
- Latitude (°)
- [CH$_4$] (ppbv)

**Data Points:**
- King George
- Brown Station
- Quarrantine Rd
- Eastern Sanitary

**Graph:**
- [CH$_4$] (ppbv)
- [CH$_4$] (ppbv) vs. Longitude (°)
- 11.5 km
Estimate of CH₄ Emissions from King George Landfill

Mass Balance Experiment (MBE) approach:

\[ E \cdot R_{CH_4} = \int_{z_i}^{z_f} \int_{0}^{x} ([C] - [C]_b) \times U_\perp \, dx \, dz \]

- \([C]\): concentrations (downwind)
- \([C]_b\): concentration in background
- \(U_\perp\): perpendicular wind speed

CH₄ emission rate from King George landfill **based on a single downwind transect**:

24.0 moles CH₄ s⁻¹, or **12,100 tons CH₄ yr⁻¹**

This is close to EPA’s GHGRP CH₄ emission data for this landfill: **11,800 tons CH₄ yr⁻¹**