

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

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Outline

- Aircraft measurements and mass balance approach for CH₄ emissions
- Comparison with CH₄ Emission inventories
- CH₄ emissions based on CO & CO₂ inventories and observed CH₄/CO & CH₄/CO₂ ratios



NIST

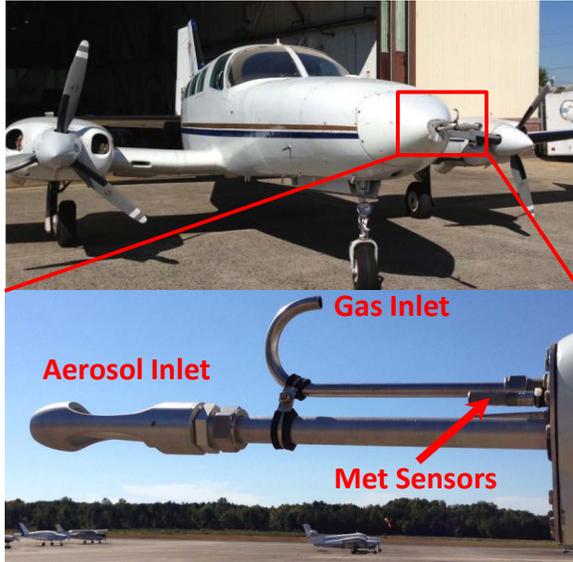


Close collaboration with NIST & MDE.

- CO₂ is the big climate forcing factor.
- Initial results indicate emissions inventories for CO₂ are good to within 10-15%.
- It is not possible to do better with current technology, but we're working on improvements.
- Methane (CH₄) 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 CH₄ emissions estimates.

UMD Cessna & Purdue Duchess Research Aircraft

UMD Cessna



GPS Position (Lat, Long, Altitude)

Met (T, RH, P, wind speed/direction)

Trace gases:

O₃: UV Absorption, modified TECO

SO₂: Pulsed Fluorescence, modified TECO

CH₄/CO₂/CO/H₂O: Cavity Ringdown, Picarro

NO₂: Cavity Ring Down, Los Gatos

NO: Chemiluminescence, modified TECO

VOCs: grab canisters/GC-FID

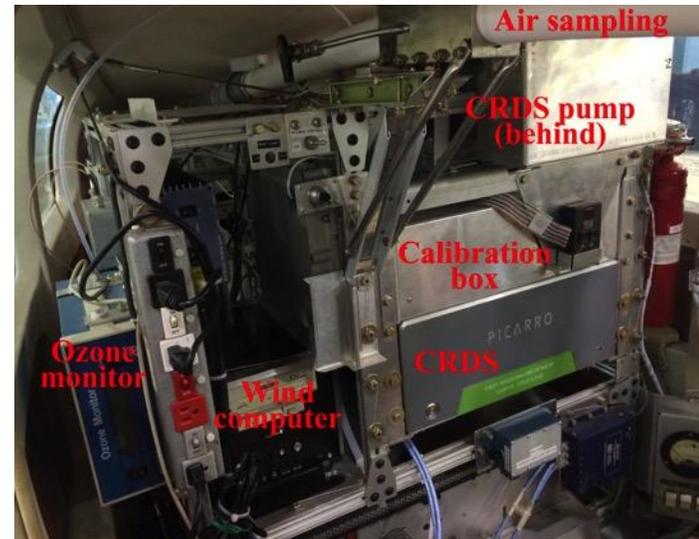
Aerosol Optical Properties:

Scattering: b_{scat} (@450, 550, 700 nm),
Nephelometer

Absorption: b_{ap} (565 nm), PSAP

Black Carbon: Aethalometer

Purdue Duchess



GPS Position (Lat, Long, Altitude)

Met (T, RH, P, 3-D wind by BAT)

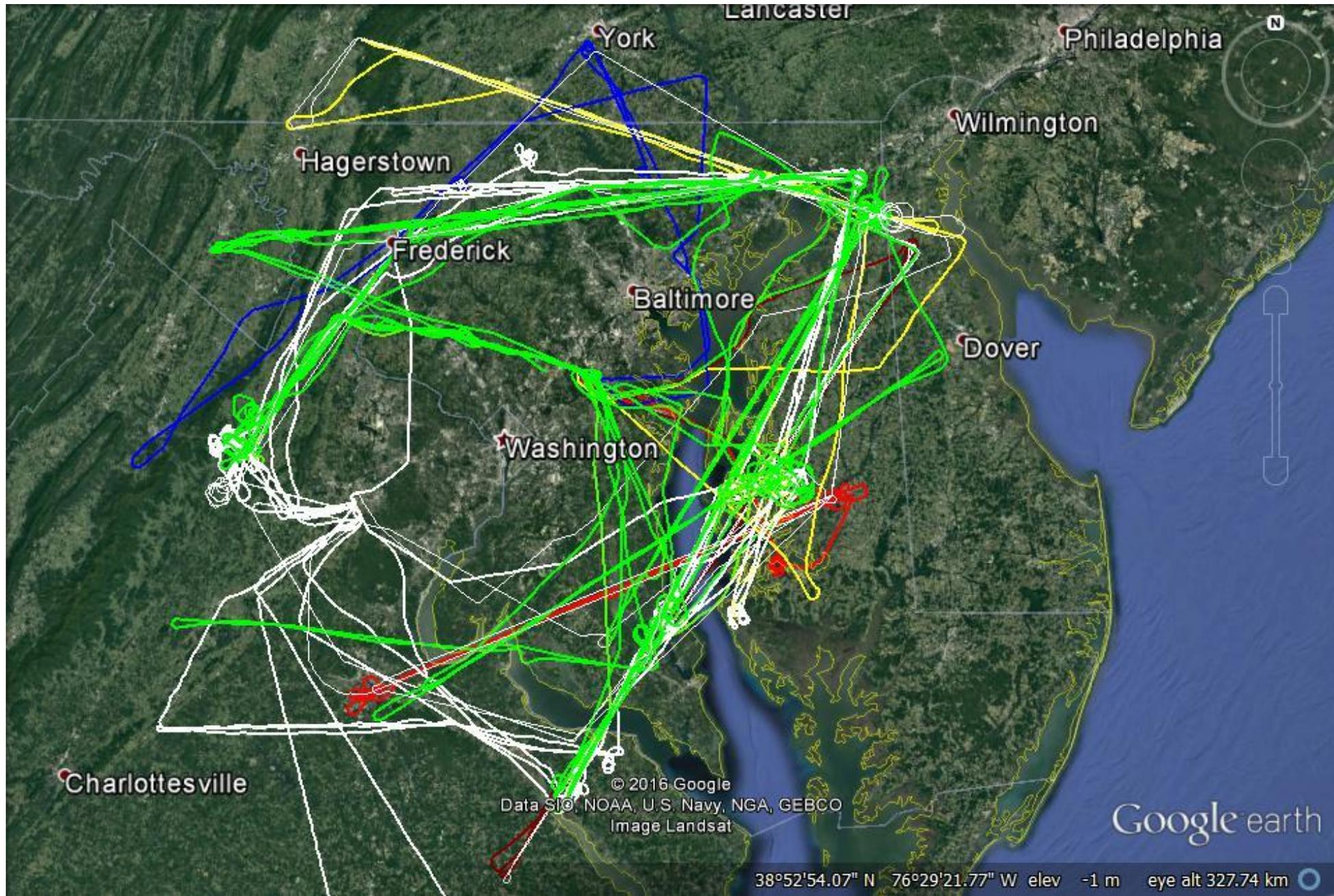
Trace gases:

O₃: UV Absorption, 2B Technology

CH₄/CO₂: Cavity Ring Down, Picarro

NO₂: Cavity Ring Down, Los Gatos

FLAGG-MD Flights during Winter 2015 and 2016

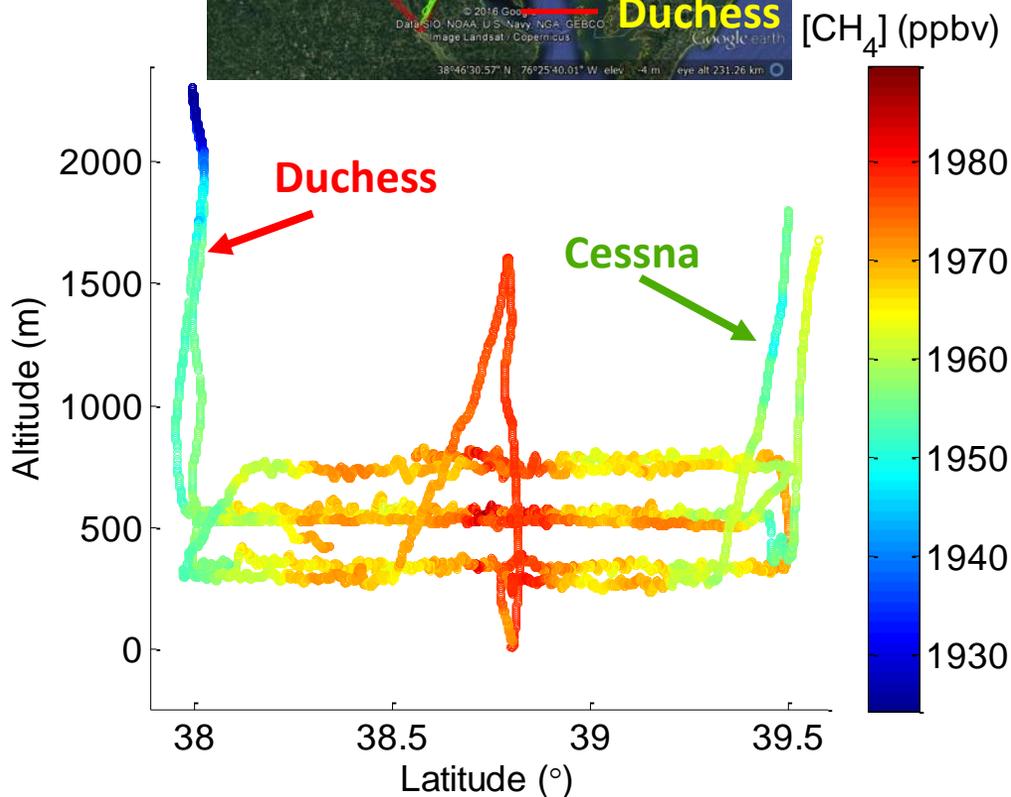


Different Flight Patterns in Winter 2015 and 2016

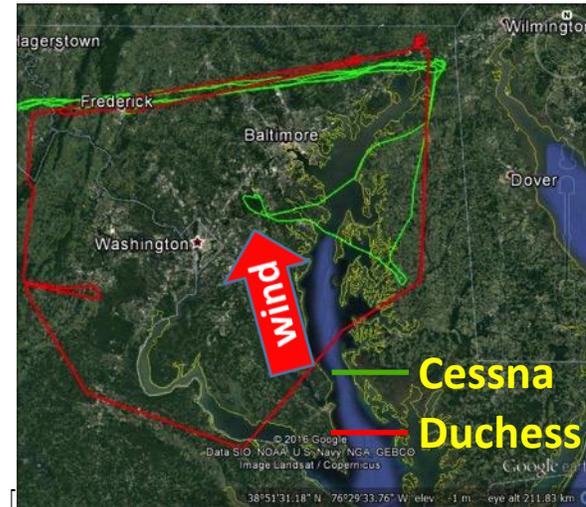
Flights on 2/19/2015



FLAGG-MD 2015:
Both aircraft did level transects

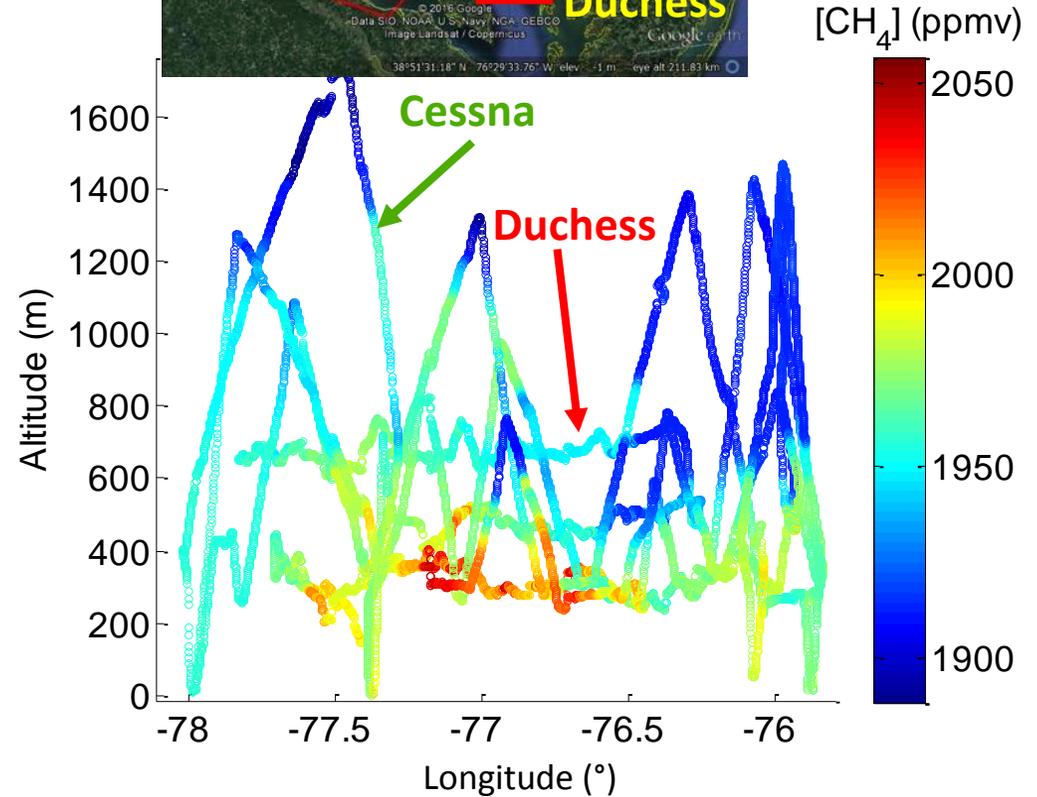


Flights on 2/19/2016



FLAGG-MD 2016:

- The Cessna did continuous vertical profiles downwind.
- The Duchess did upwind survey & level transects downwind.



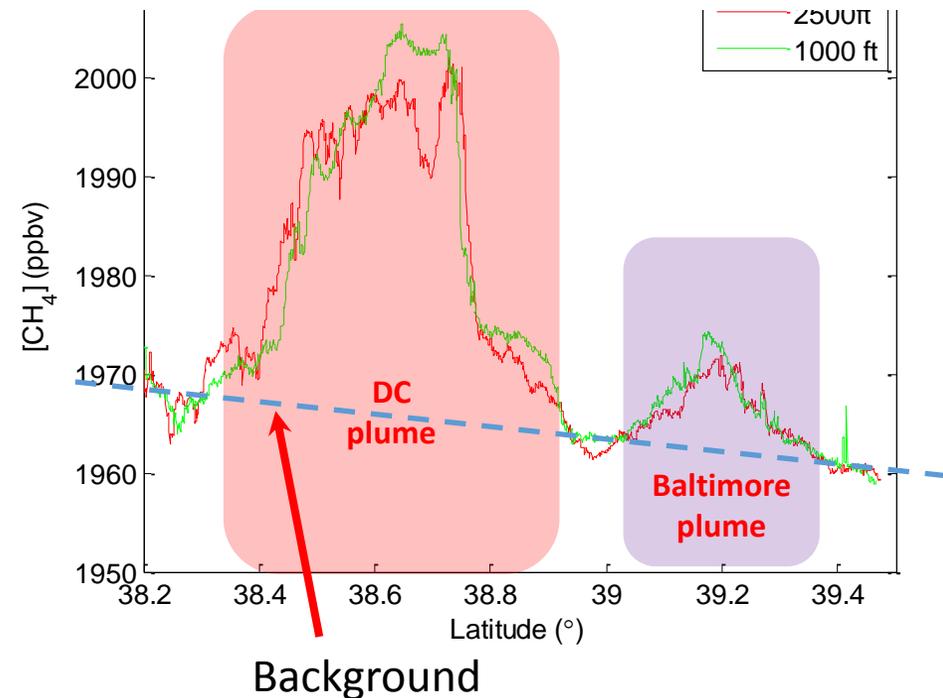
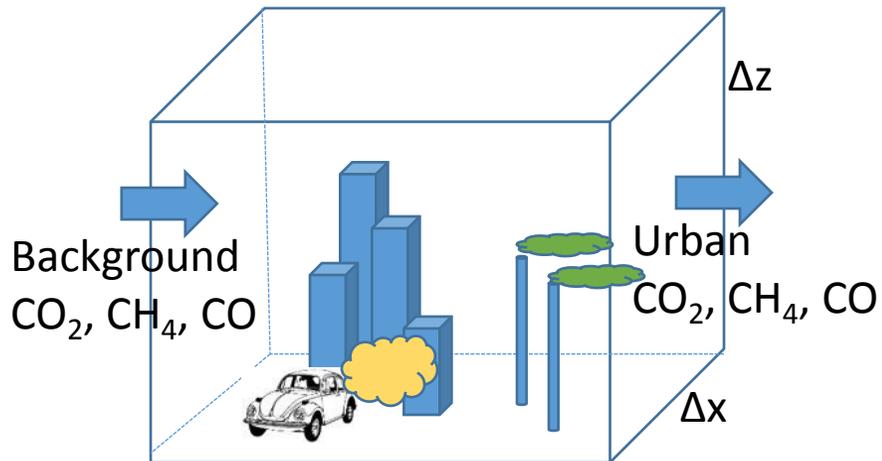
Mass Balance Approach to Estimate Emission Rates

Mass Balance Experiment (MBE) approach:

$$\left[\begin{array}{c} z_i \\ \hat{} \end{array} + x \right]$$

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



Estimated GHG Emissions from the Baltimore-Washington Area

FLAGG-MD winter 2015

Flight Date	Flux(CO ₂) (moles s ⁻¹)	Flux(CH ₄) (moles s ⁻¹)	Flux(CO) (moles s ⁻¹)
2/6/15	94,500	557	521
2/13/15	71,000	290	281
2/18/15	91,200	795	--
2/19/15	156,800	932	567
2/20/15	118,100	518	753
2/23/15	107,900	641	417
2/24/15	110,300	476	640
2/25/15	108,500	602	571
2/27/15	78,800	540	--
Mean±1σ	104,000 ±25,000	595±184	536±152

FLAGG-MD winter 2016

Date	Flux(CO ₂) (moles s ⁻¹)	Flux(CH ₄) (moles s ⁻¹)	Flux(CO) (moles s ⁻¹)
02/08/16	73,200	418	430
02/12/16	93,000	350	510
02/17/16	107,800	1,078	365
02/18/16	98,100	373	611
02/19/16	142,800	722	688
Mean±1σ	103,000 ±25,600	588±312	521±131

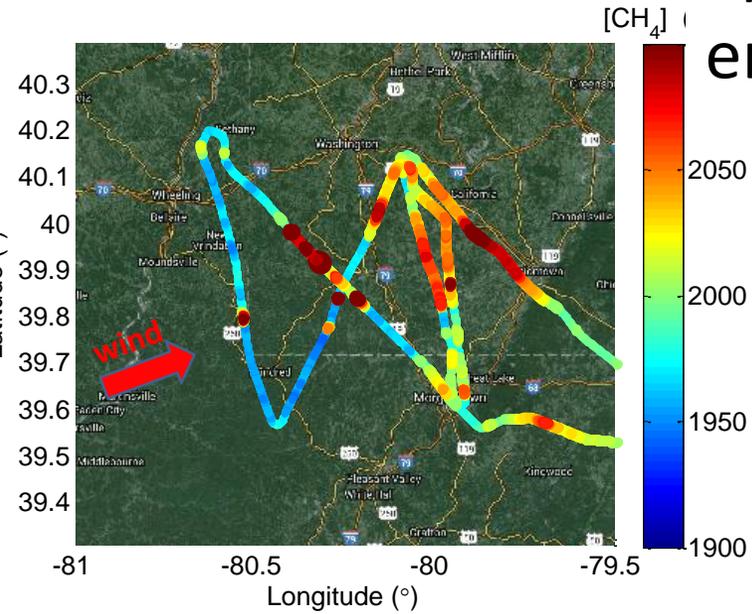
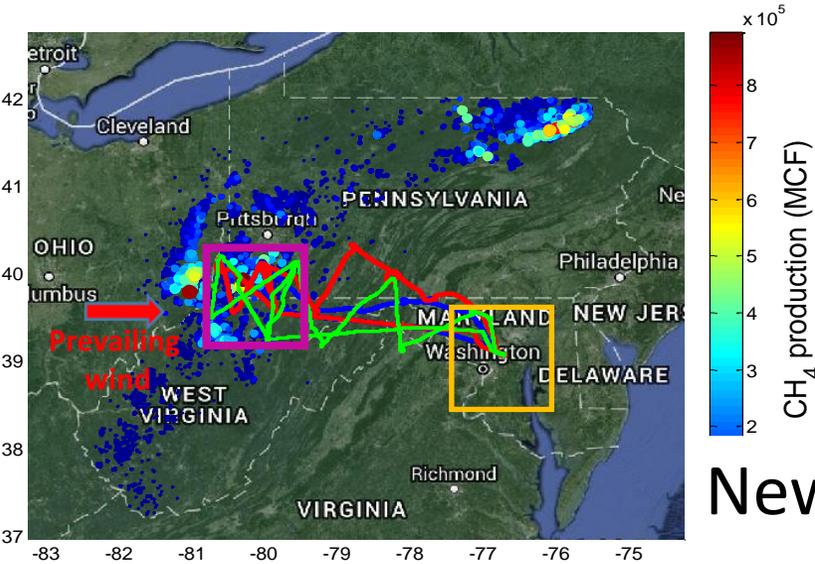
Mean CH₄ emission rate: 592 ± 248 moles s⁻¹

600 moles/s ~ 30,000 tons/yr



CH₄ Emissions from the SW Marcellus

New Regulations on finishing appear to have improved emissions.

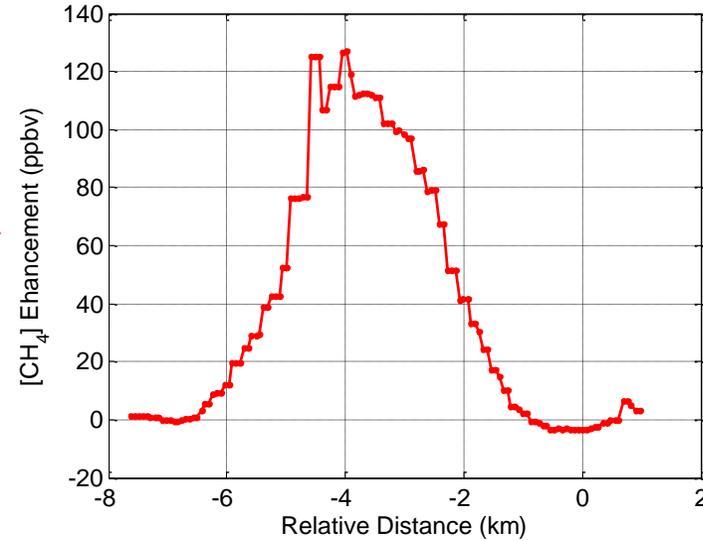
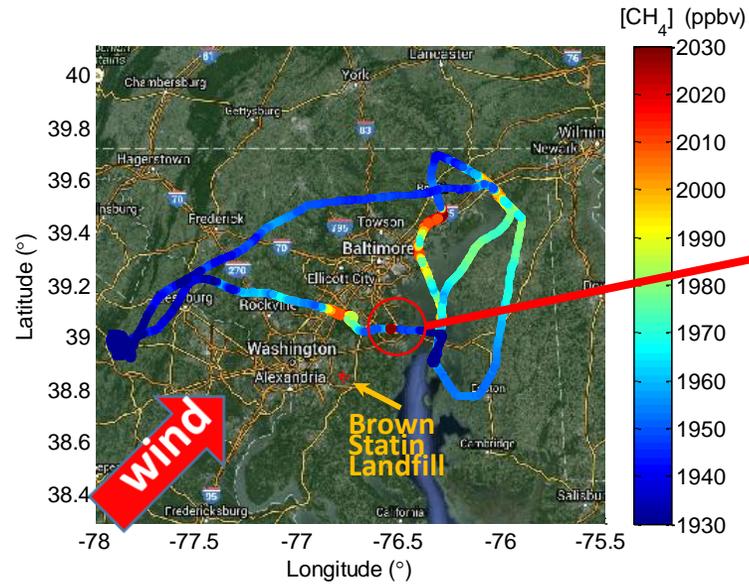


		WD (deg)	PBL Height (m AGL)	CH ₄ E.R. (moles s ⁻¹)		
8/25/15	1,967±22	2,023±39	9.7±1.4	260±12	2,200	2,391
8/29/15	2,016±16	2,119±50	6.6±1.4	226±13	1,950	2,315
9/14/15	1,960±28	2,032±37	9.6±1.2	283±12	1,500	2,156

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: CH₄ from Brown Station Landfill



Measured 21 times
over 7 flights.

<u>Flight #</u>	<u>CH₄ Emission Rate (moles s⁻¹)</u>
RF1	57.4
RF2	55.7
RF4	51.8
RF5	64.7
RF6	65.3
RF7	36.0
RF8	69.1

Mean	57.1 ± 11.1

EPA GHGRP CH₄
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

Landfill (# of transects/flights)	Mass Balance Flux Range** (moles CH ₄ /s) - Average of all transects	2015 EPA (moles CH ₄ /s)	2014 MDE (moles CH ₄ /s)	2012 Maasakkers (moles CH ₄ /s)
Brown Station (21/7)	24.6 – 64.0	3.44 <i>or</i> 15.5*	6.14	8.52
Eastern Sanitary (3/1)	9.9 – 49.5	4.54	6.85	10.01
Quarantine Road (7/3)	1.35 – 2.94	3.33	17.6	3.11
Harford Waste (2/2)	2.22 – 8.50	5.42	3.52	5.52
Reichs Ford (3/2)	12.8 – 26.2	5.88	1.83	4.96
Route 40 West (3/3)	2.66 – 10.01	6.89	10.23	6.70

* 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).

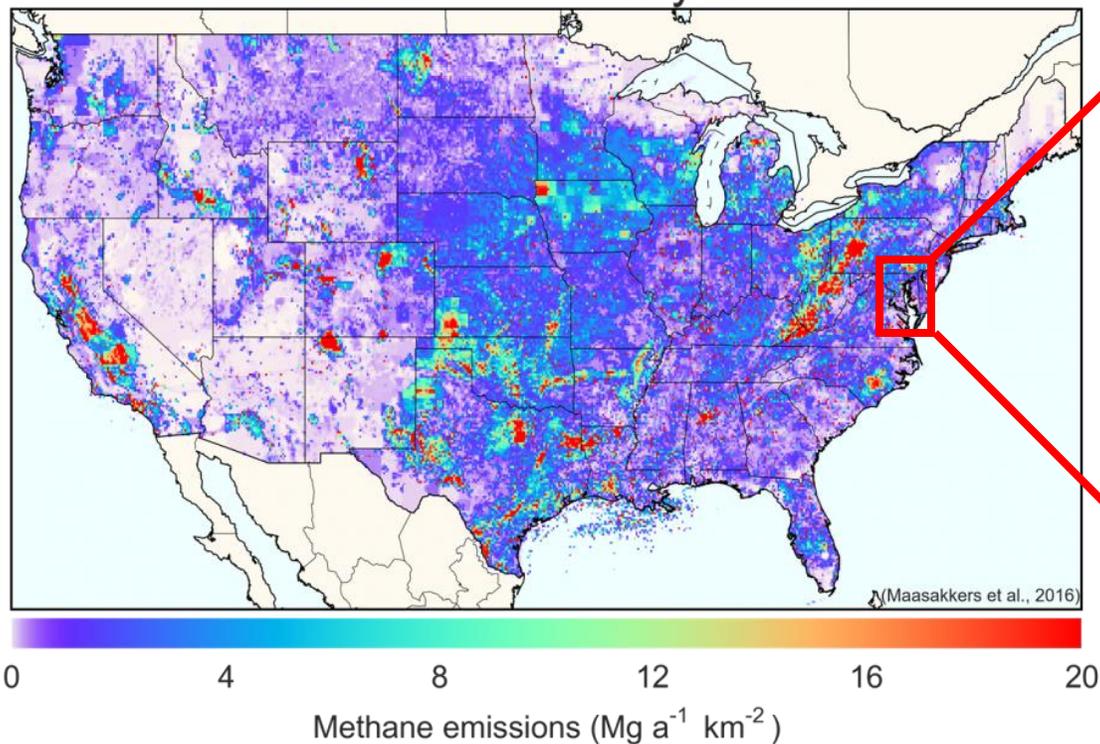
**Flux range computed by varying horizontal transect width by 10% and PBL height by one-sigma.

New gridded US CH₄ NEI for 2012 (0.1° x 0.1°)

Gridded National Inventory of U.S. Methane Emissions

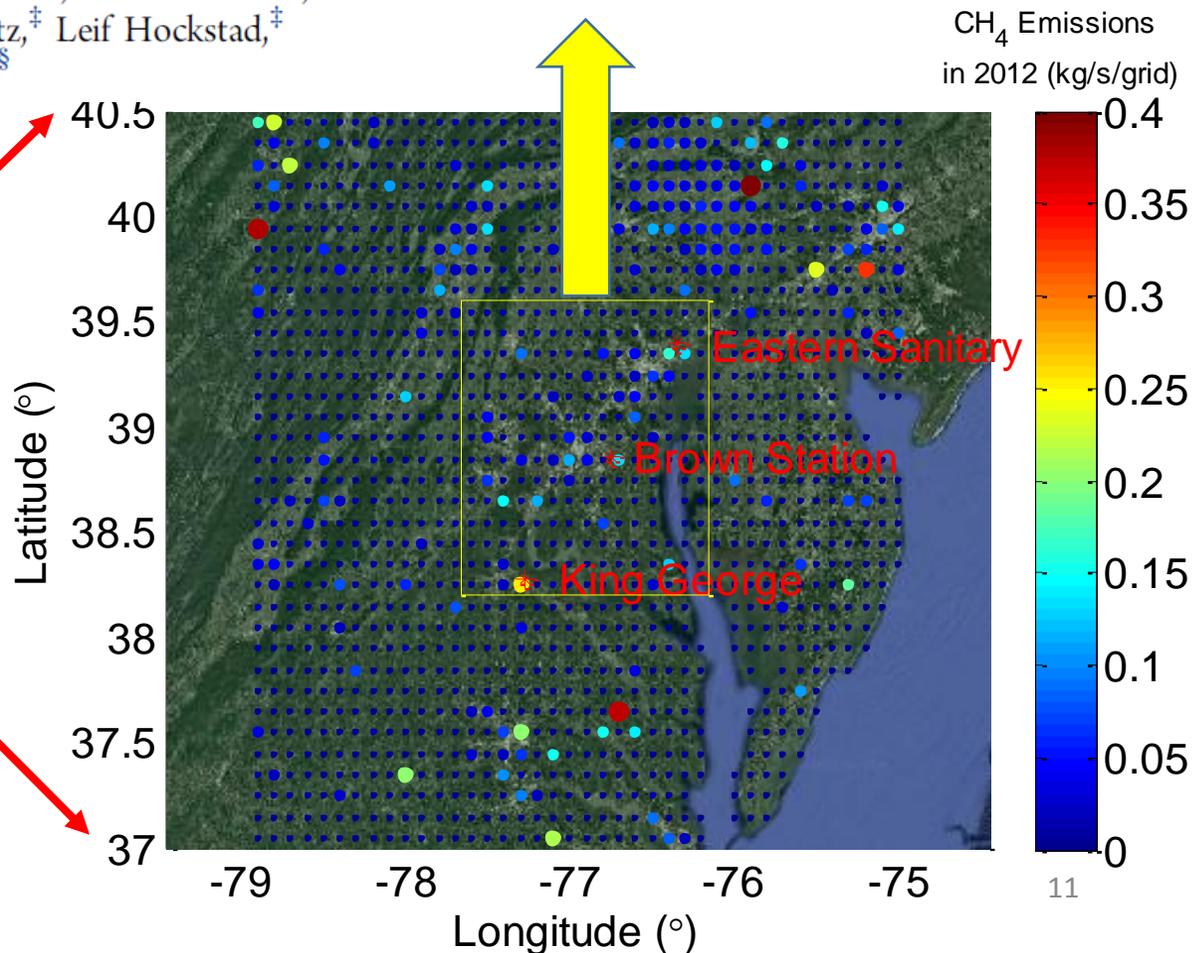
Joannes D. Maasakkers,^{*,†} Daniel J. Jacob,[†] Melissa P. Sulprizio,[†] Alexander J. Turner,[†] Melissa Weitz,[‡] Tom Wirth,[‡] Cate Hight,[‡] Mark DeFigueiredo,[‡] Mausami Desai,[‡] Rachel Schmeltz,[‡] Leif Hockstad,[‡] Anthony A. Bloom,[¶] Kevin W. Bowman,[¶] Seongeun Jeong,[§] and Marc L. Fischer[§]

Gridded EPA Inventory for 2012

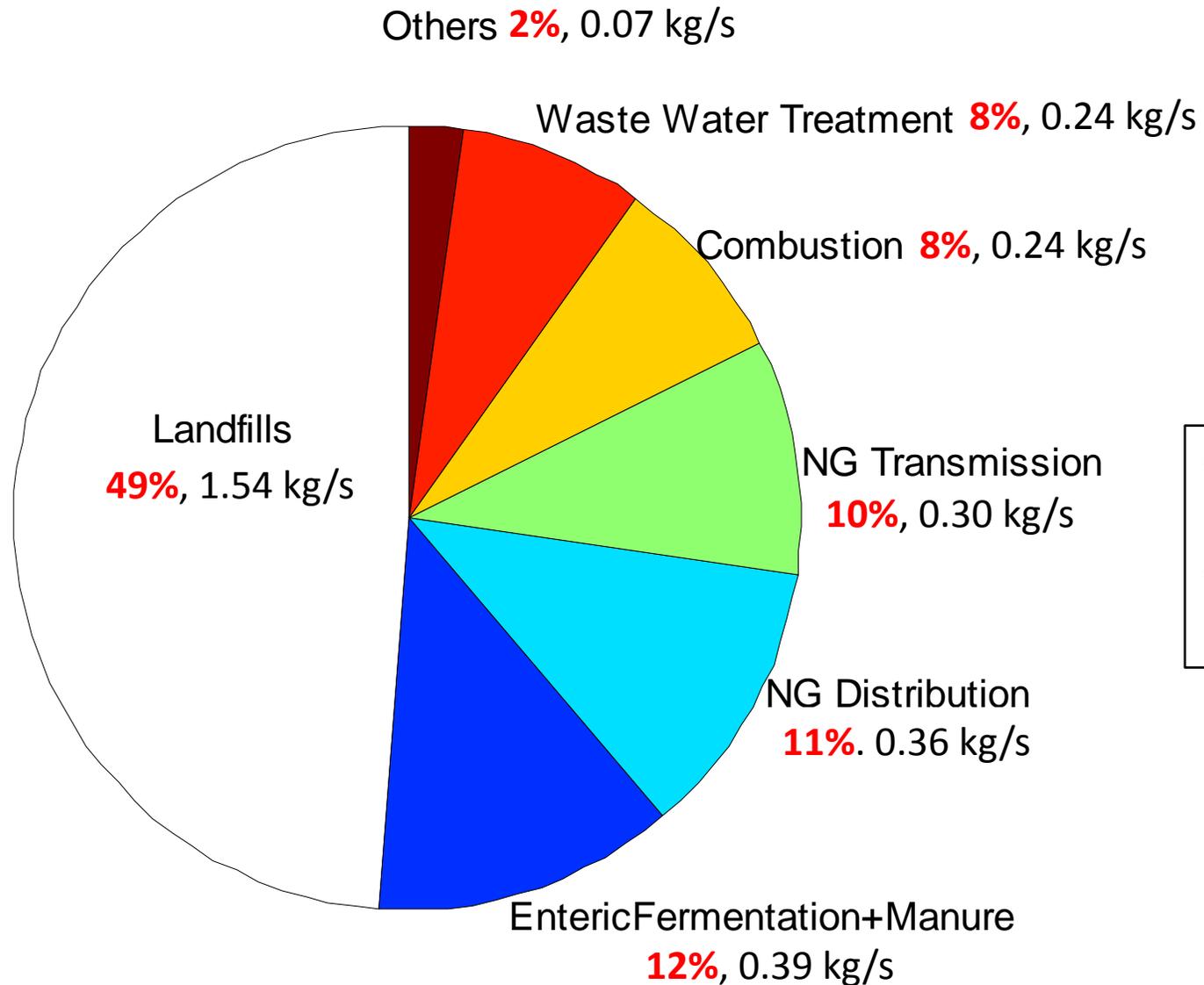


Includes all methane emissions included in the National Greenhouse Gas Inventory.

**196 moles CH₄/s
from Balt-DC**



CH₄ Emissions from Sources in Balt-DC in CH₄ NEI 2012

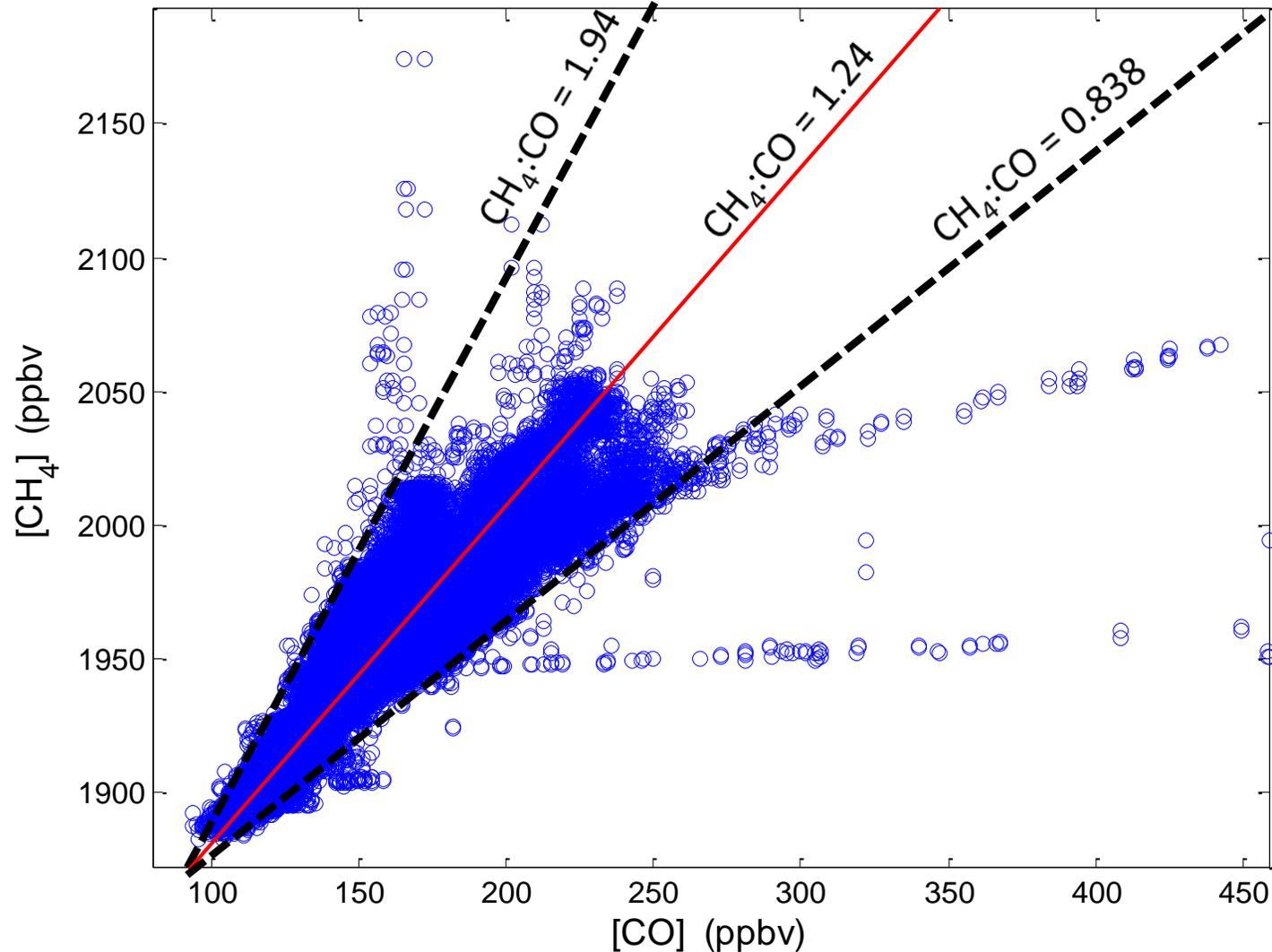


- Landfills are a major CH₄ source.
- CH₄ emissions from NG system may be under-estimated.

An alternative approach – CO₂ and CO emissions well constrained and we can use ratios to learn about the methane flux.

CH₄ Emission Estimate using CH₄/CO Ratio and CO Emissions

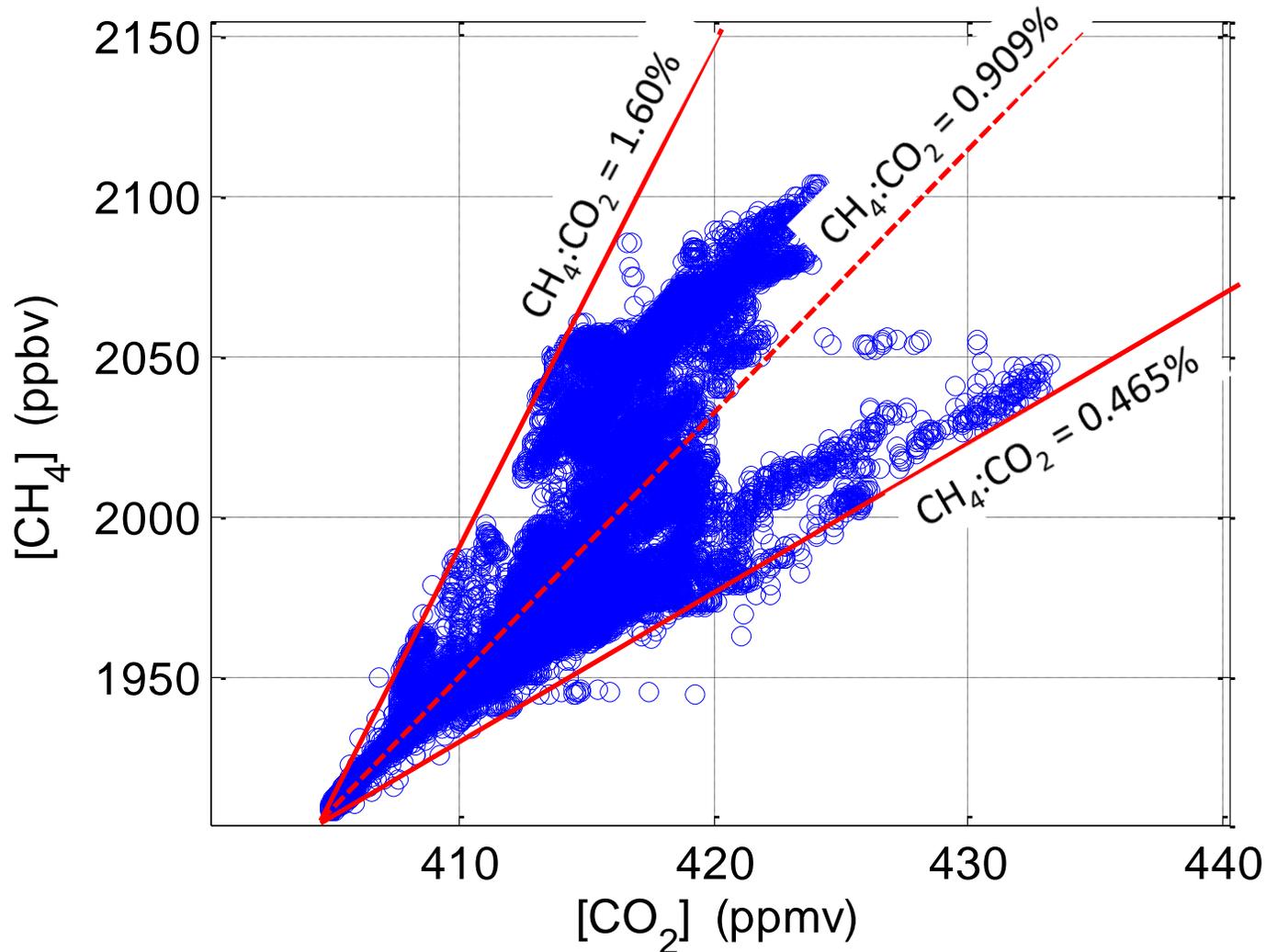
FLAGG-MD winter 2015



- Total CO emissions from Balt-DC in EDGAR v4.3 2010: **0.459 MMtons/yr**
- The total CH₄ emissions based on CH₄ to CO ratio: **642 moles CH₄ s⁻¹** (436 – 1,003 moles CH₄ s⁻¹)

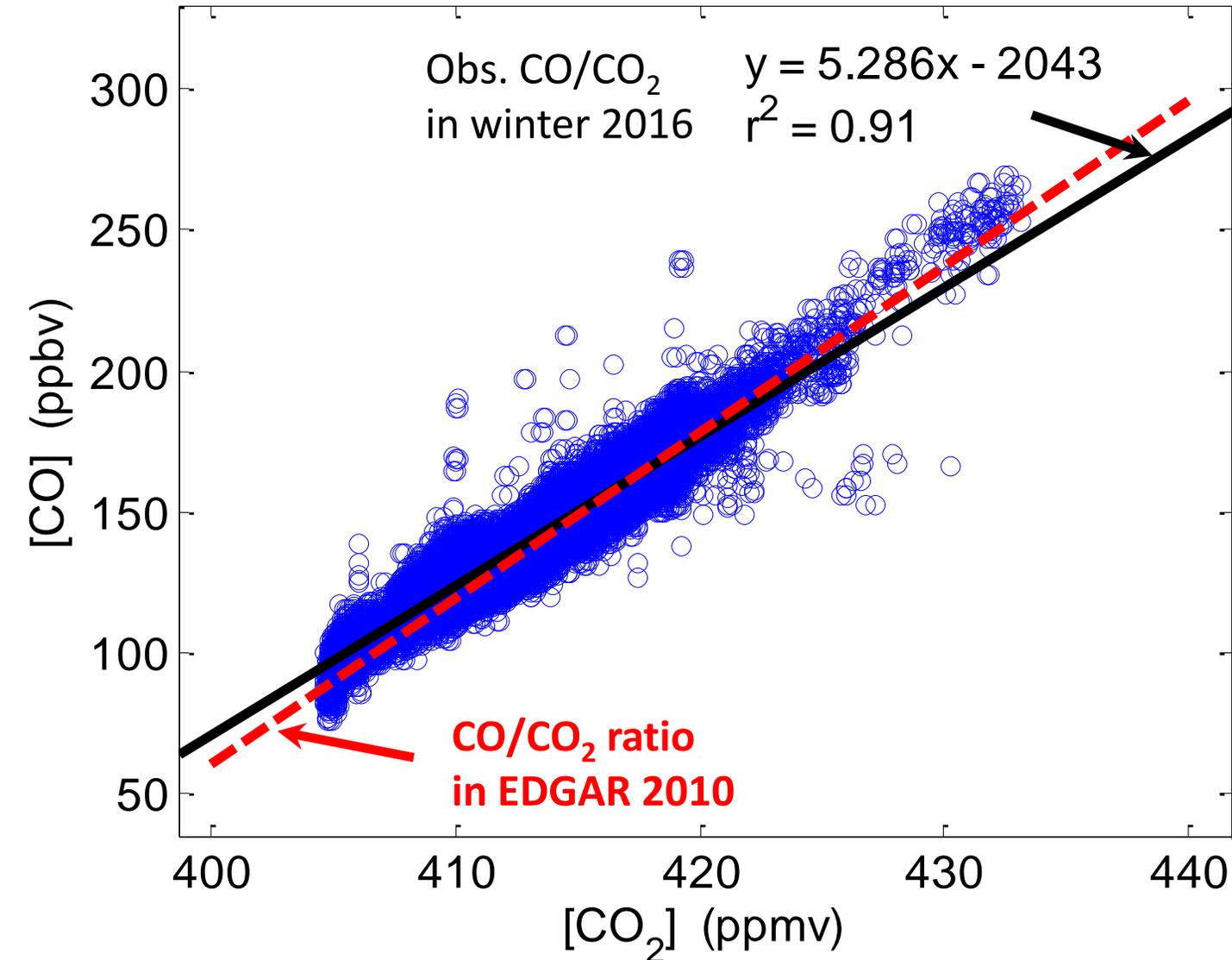
CH₄ Emission Estimate using CH₄/CO₂ Ratio and CO₂ NEI 2014

FLAGG-MD winter 2016



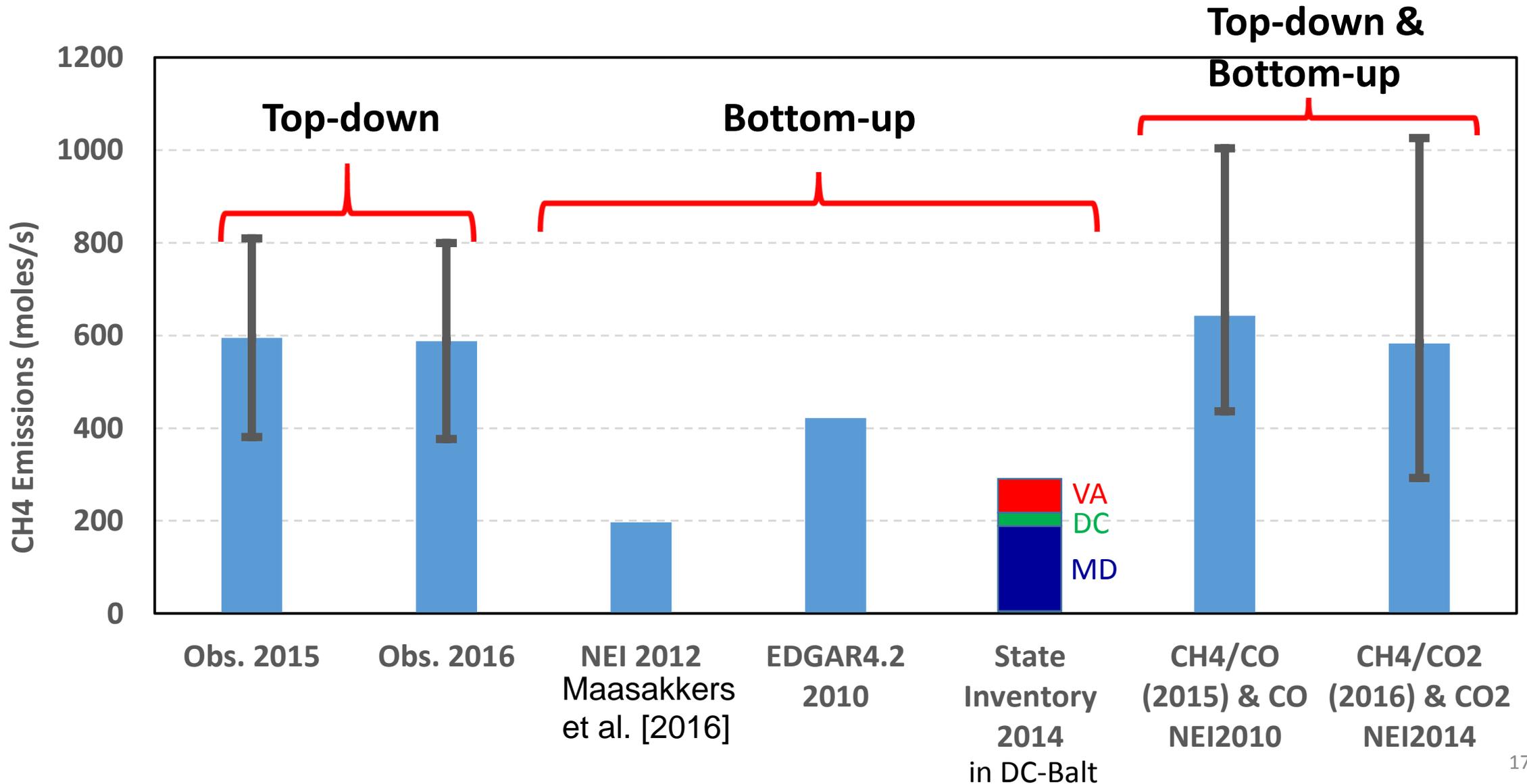
- Total CO₂ emissions from Balt-DC in NEI2014: **89.0 MMtons/yr**
- The total CH₄ emissions based on CH₄ to CO₂ ratio: **583 moles CH₄ s⁻¹** (293 – 1,029 moles CH₄ s⁻¹)

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)

CH₄ Emissions from the Balt-DC Area: Top-down vs. Bottom-up



Summary

- UMD is working with MDE to fine tune emissions inventories. CO₂ is the big player with CH₄ ~10% of total.
- Estimated total emissions of CH₄ from Balt/Wash area:
 - 595±184 moles CH₄ s⁻¹ in winter 2015
 - 588±312 moles CH₄ s⁻¹ in winter 2016 (**~30,000 tons/yr**)
- Major CH₄ sources in the area: landfills and broadly, NG system.
- Direct observations of CH₄ emissions 1.4 to 3 times higher than inventories.
- Only flew in winter so far; need summer flights.

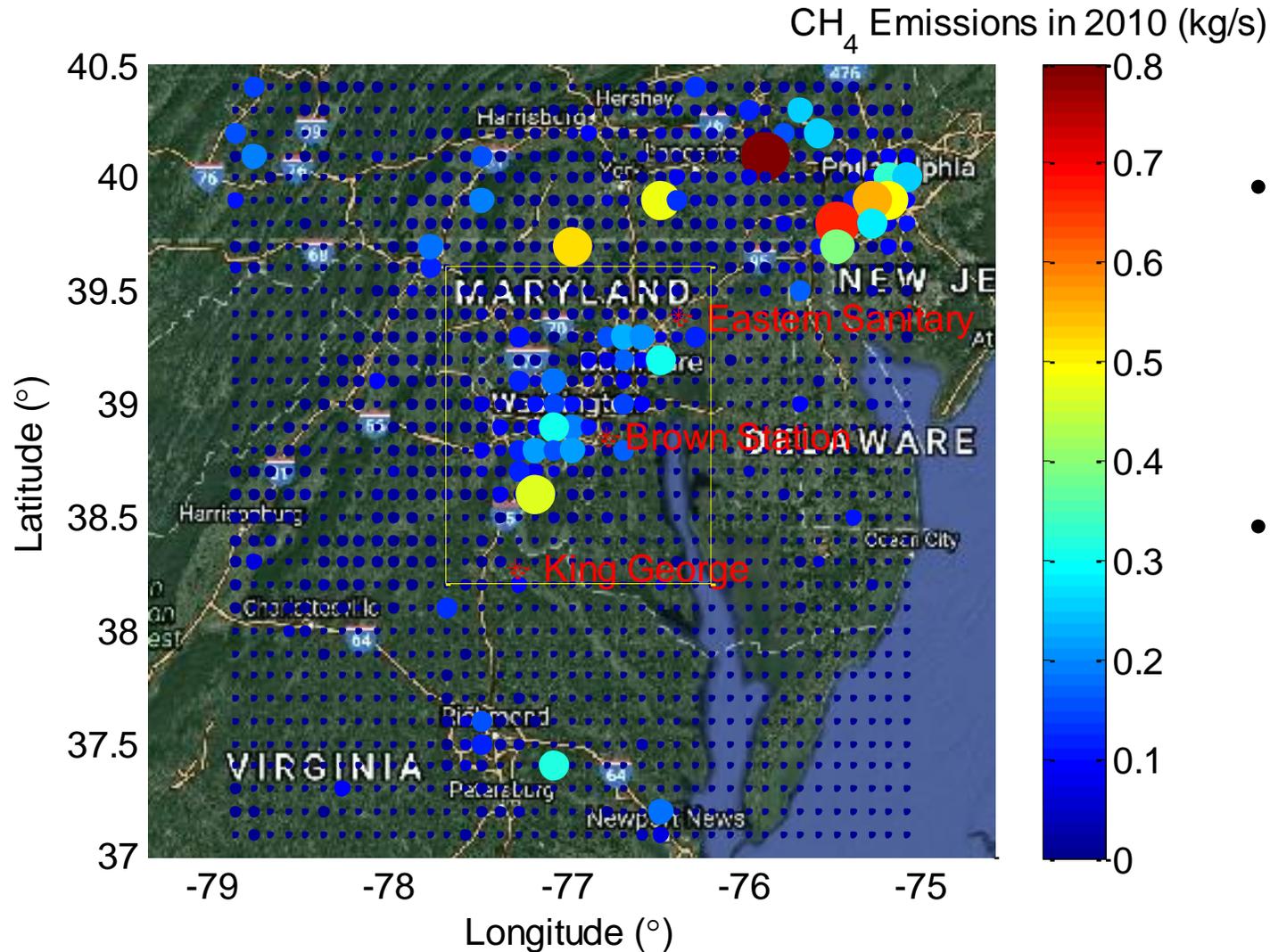
Extra Slides

Summary

- 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 CH₄ from Balt/Wash area:
 - 595±184 moles CH₄ s⁻¹ in winter 2015
 - 588±312 moles CH₄ s⁻¹ in winter 2016 (**~30,000 tons/yr**)
- Major CH₄ sources in the area: landfills and broadly, NG system.
- Compared to CH₄ emission inventories:
 - (1) Observed CH₄ 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 CH₄ emissions is similar to CH₄ emissions inferred from CO and CO₂ NEI with observed CH₄/CO and CH₄/CO₂ ratios.

EDGAR4.2 Global $0.1^\circ \times 0.1^\circ$ CH_4 Emission Inventory 2010

Data source: <http://edgar.jrc.ec.europa.eu/gallery.php?release=v42FT2010&substance=CH4§or=TOTALS>



- The total CH_4 emissions in the yellow rectangle (an approximately surveyed area) is **421 moles $\text{CH}_4 \text{ s}^{-1}$** .
- Issues with EDGAR emissions: mainly allocated based on population instead of source locations.

Mass Balance – CH₄ from DC/Baltimore

Flight Date	Emission Rate (moles CH ₄ s ⁻¹)	Emission Uncertainty Range – Horizontal Bounds* (moles CH ₄ s ⁻¹)	Flux Uncertainty Range – PBL Depth** (moles CH ₄ s ⁻¹)	Flux Uncertainty Range – Horizontal Bounds & PBL Depth Combined*** (moles CH ₄ s ⁻¹)
2/13/15	193	177 – 201	145 – 241	133 – 251
2/19/15	1260	1230 – 1270	853 – 1670	830 – 1680
2/20/15	509	452 – 509	351 – 667	311 – 667
2/23/15	157	141 – 169	110 – 204	99.0 – 219
2/24/15	951	857 – 1180	705 – 1200	633 – 1490
2/25/15	183	172 – 187	97.5 – 269	90.6 – 274
Mean	542	505 – 586 (-6.8% / +8.1%)	377 – 709 (-30.% / + 31%)	349 – 764 (-36% / +41%)

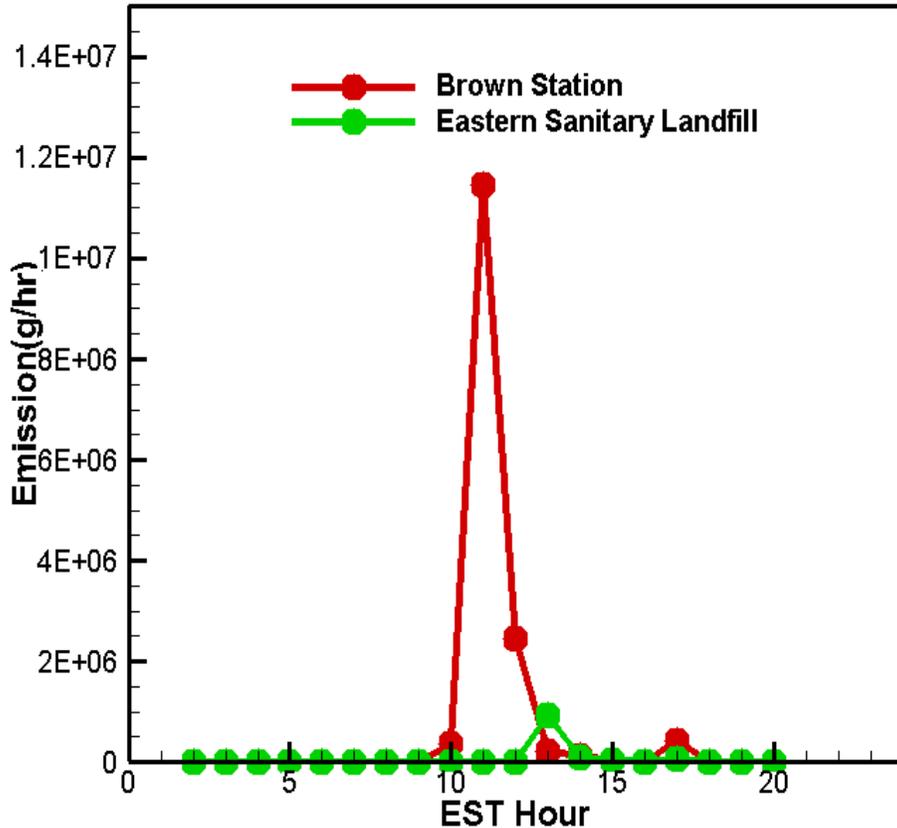
*Flux range computed by varying horizontal transect width by 10%

**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.

Date Assimilation Using HYSPLIT: Collaboration with NOAA/ARL

HYSPLIT Inverse results using Feb. 6, 2015 flight data



Tianfeng Chai and Ariel Stein (NOAA/ARL)

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 CH₄.
- The CH₄ emissions of the landfills vary each hour.

Two major limitations

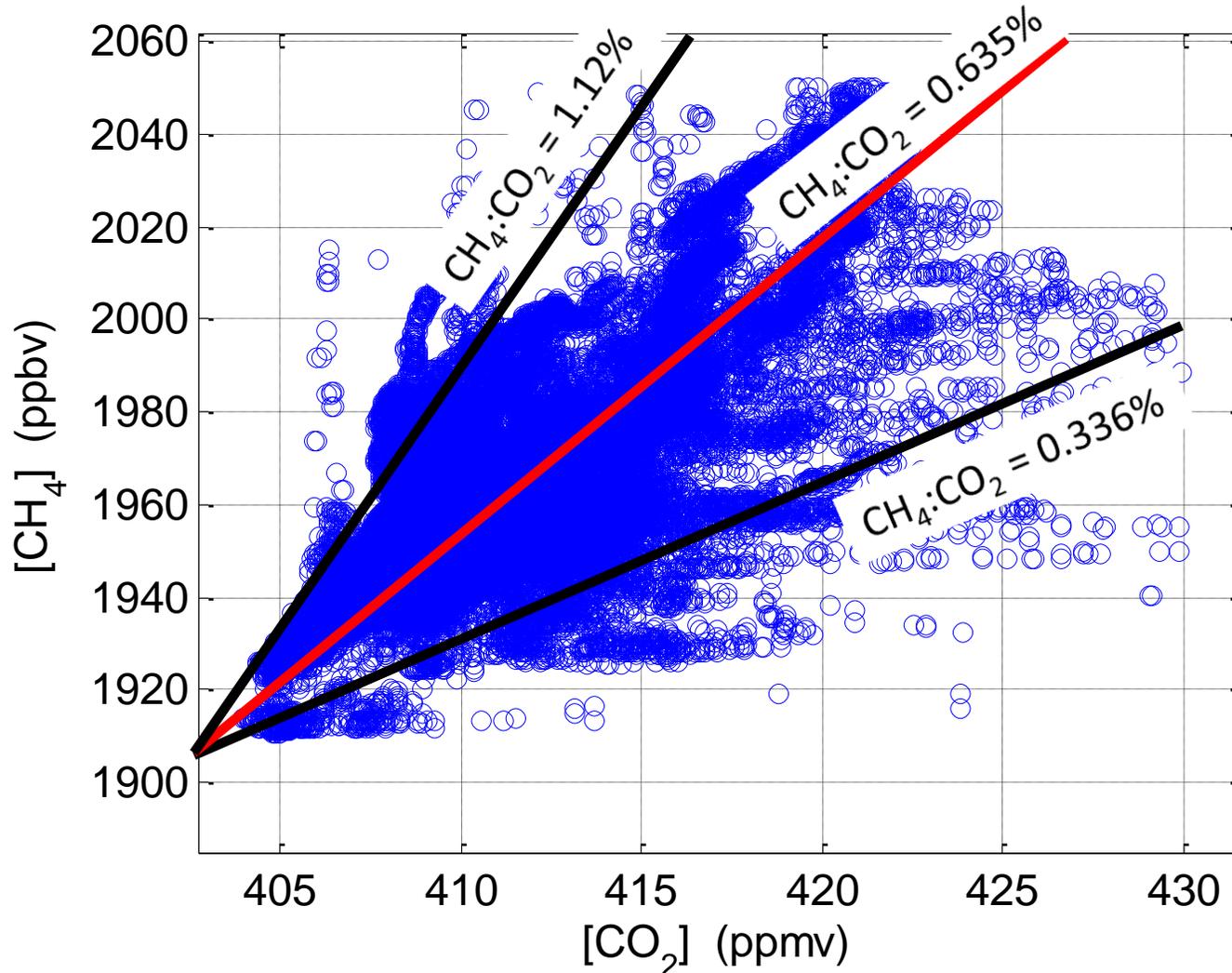
- There are other sources that contribute to the excess CH₄.
- Meteorological fields in the HYSPLIT dispersion model has uncertainties.

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

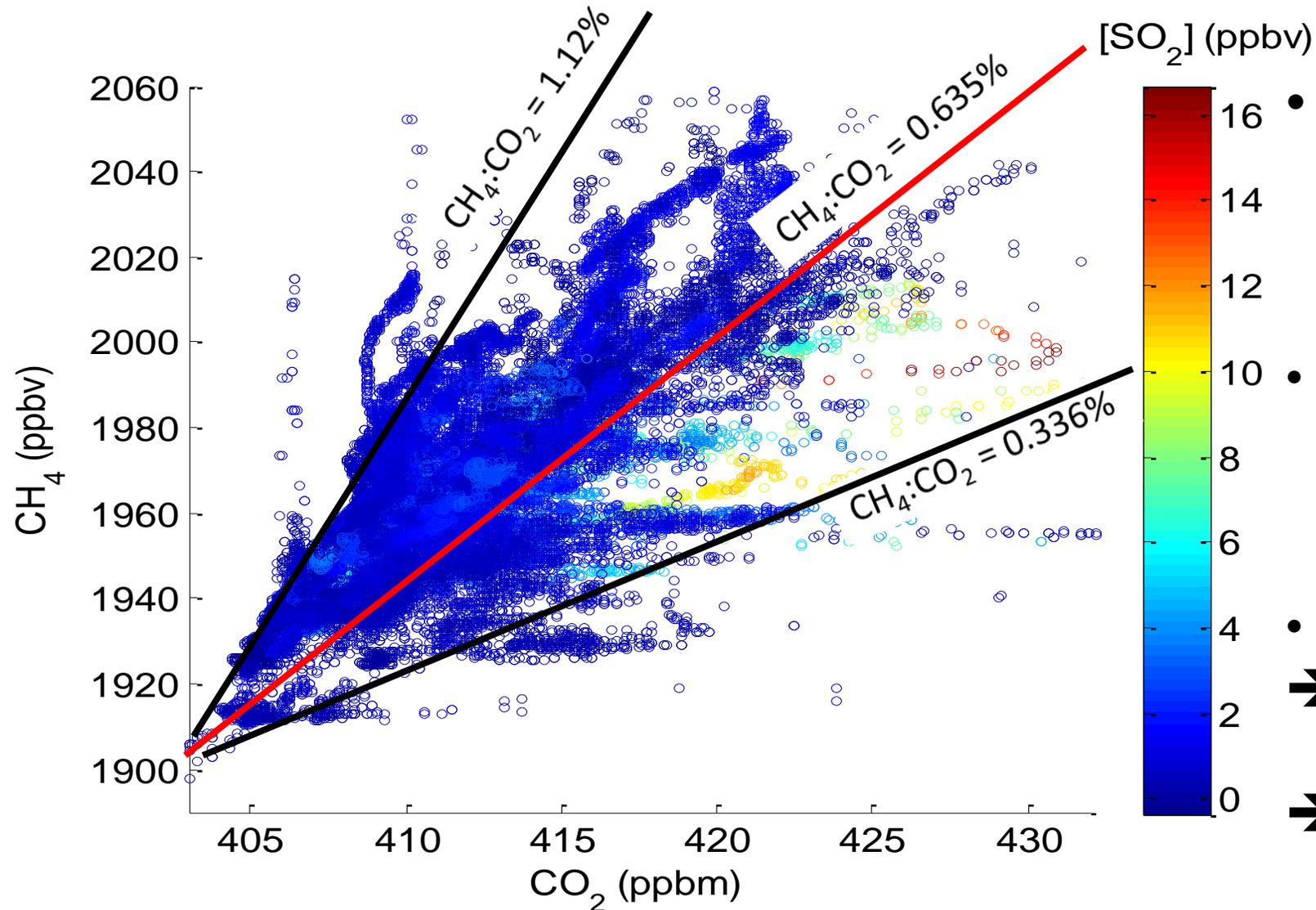
FLAGG-MD winter 2015



- 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/CO₂ Ratio and CO₂ NEI 2014

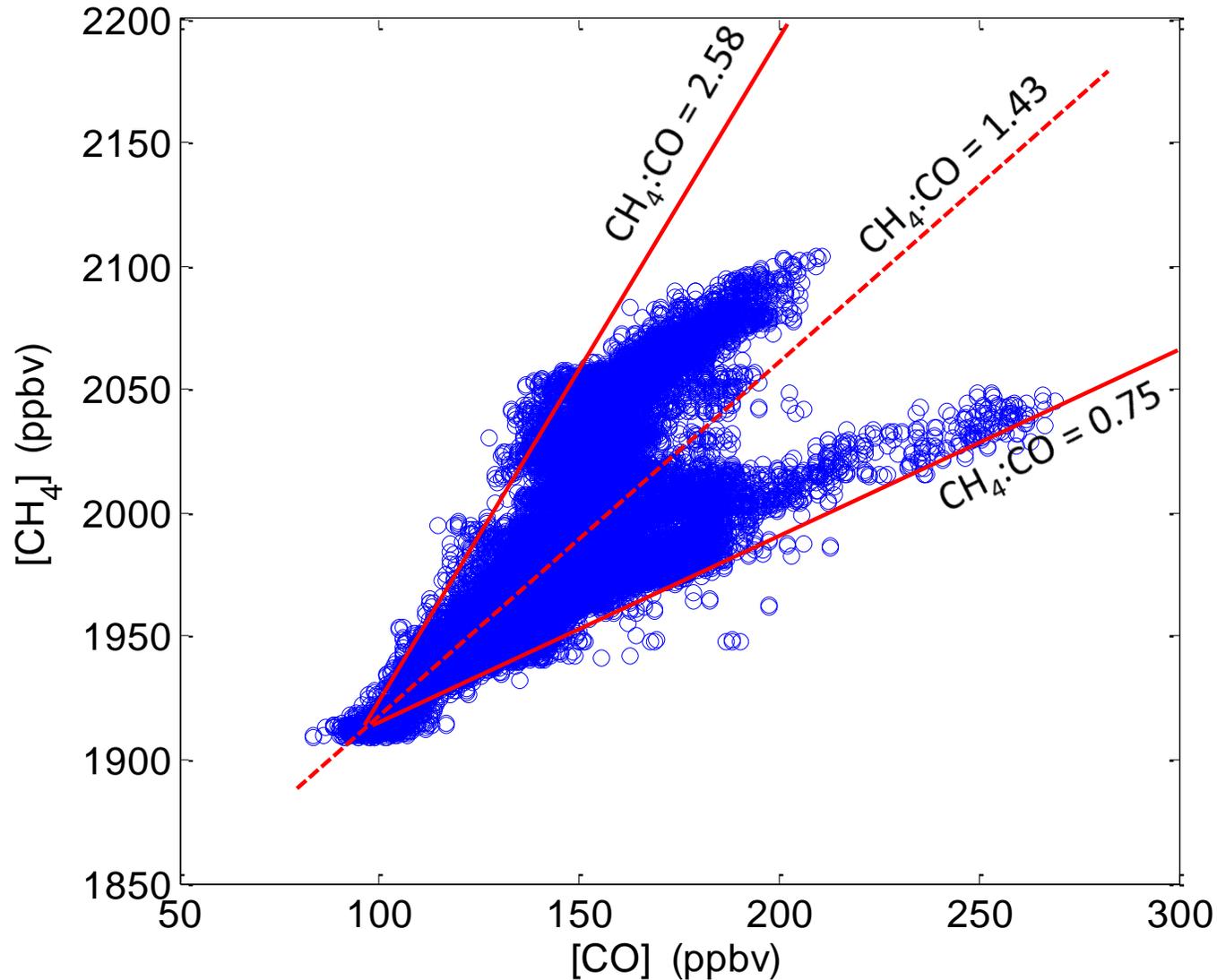
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CH₄ Emission Estimate using CH₄/CO Ratio and CO Emissions

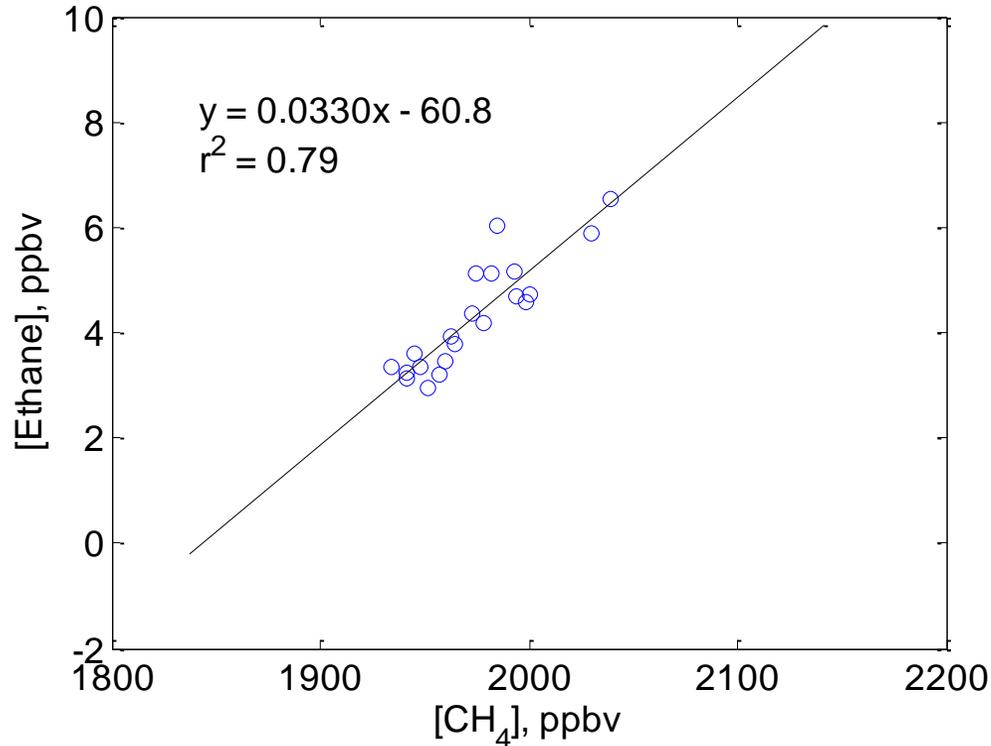
FLAGG-MD winter 2016



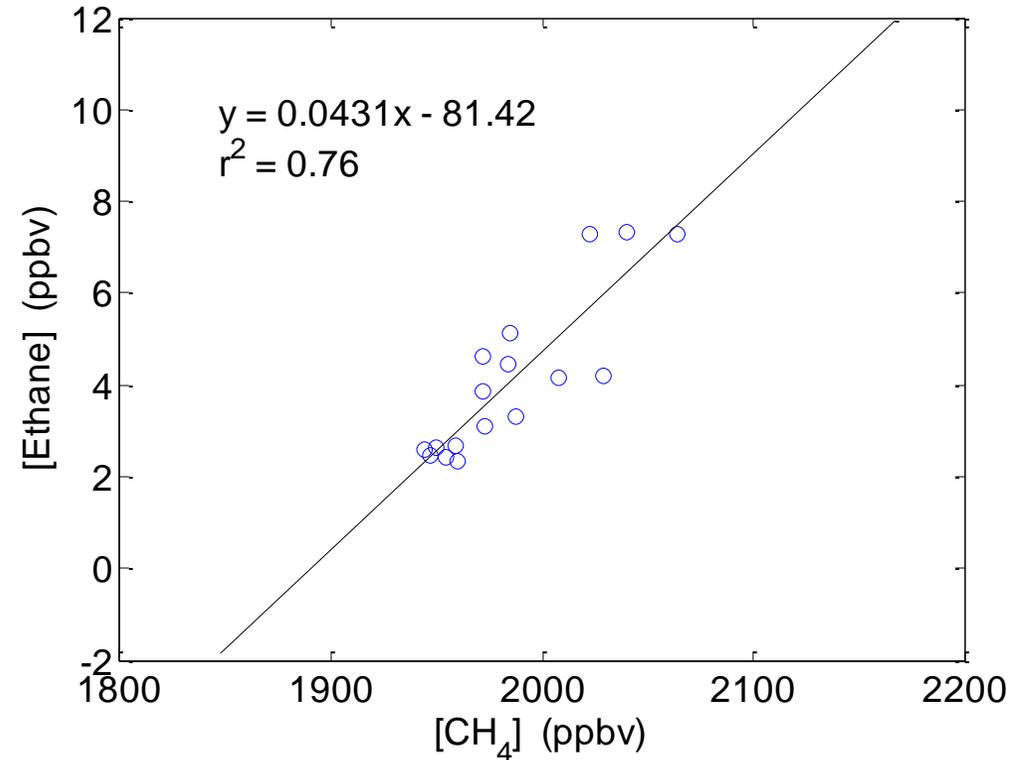
- Total CO emissions from Balt-DC in EDGAR v4.3 2010: **0.459 MMtons/yr**
- The total CH₄ emissions based on CH₄ to CO ratio: **11.8 kg CH₄ /s** (6.2– 21.4 kg CH₄/s)

Observed C₂H₆-to-CH₄ Ratio

FLAGG-MD winter 2015



FLAGG-MD winter 2016

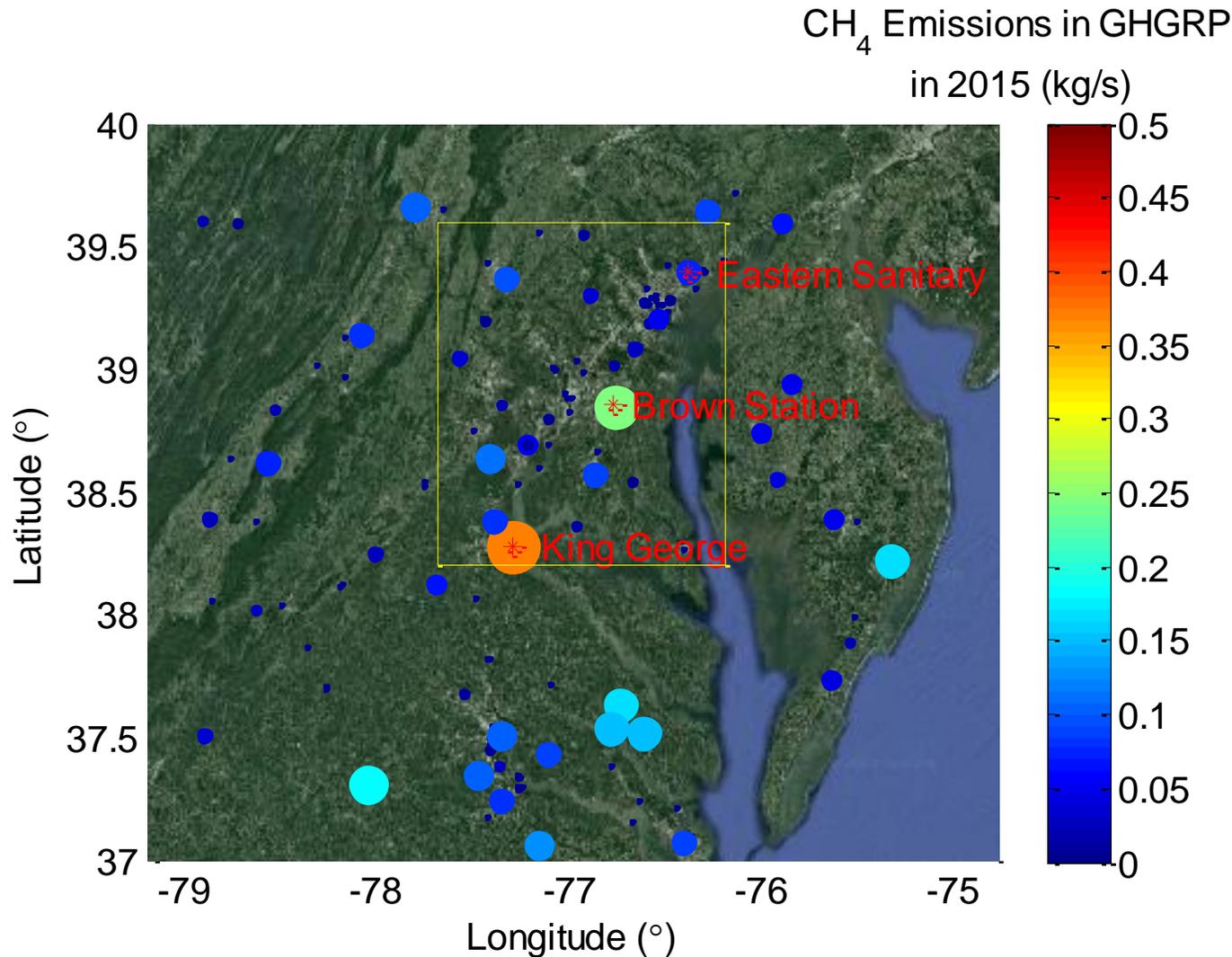


- Mean C₂H₆-to-CH₄ ratio in natural gas of Baltimore Gas & Electric: 0.1045
- Other major CH₄ sources (landfills, enteric fermentation, waste water treatment) has little ethane emissions.
- Emissions from the NG system account for 32% (2015) and 41% (2016) of total CH₄ emissions.

CH₄ 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 CH₄/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 = $54,495 \times 10^6 \text{ CF} \times 3.34\% \times \$0.012/\text{CF}$
= \$22 M in Feb. 2015

Facility level CH₄ Emissions in EPA's GHGRP (excluding emissions from Petroleum & NG System)



- Mostly landfills
- The total CH₄ emissions (other than the NG system) from the yellow rectangle is **1.21 kg/s**

Based on the unaccounted-for natural gas and other CH₄ sources in GHGRP, the total CH₄ emission rate from the Balt-DC area:

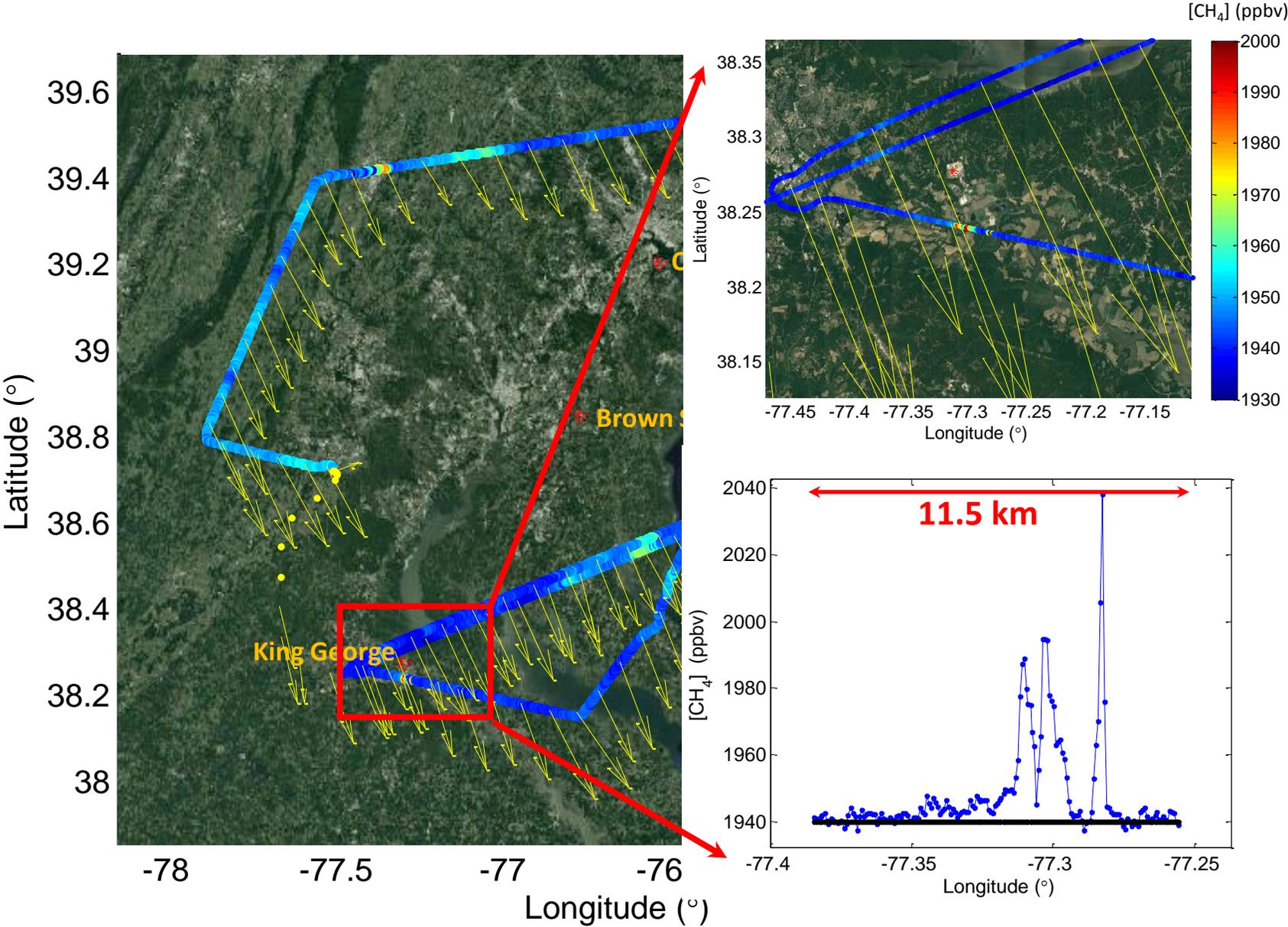
$$12.86 + 1.21 = 14.07 \text{ kg/s}$$

This may overestimate CH₄ emissions since not all LAUF gas is emitted into the atmosphere.

Brown Station – Mass Balance Calculations

RF	Mass Balance Flux (moles / s) (Mean of flux calculated on each individual transect)
1	30.7
2	44.7
4	31.9
5	59.9
6	21.1
7	71.8
8	66.3
Mean ± St. Deviation	46.6 ± 18.3

CH₄ emissions from King George Landfill



Estimate of CH₄ Emissions from King George Landfill

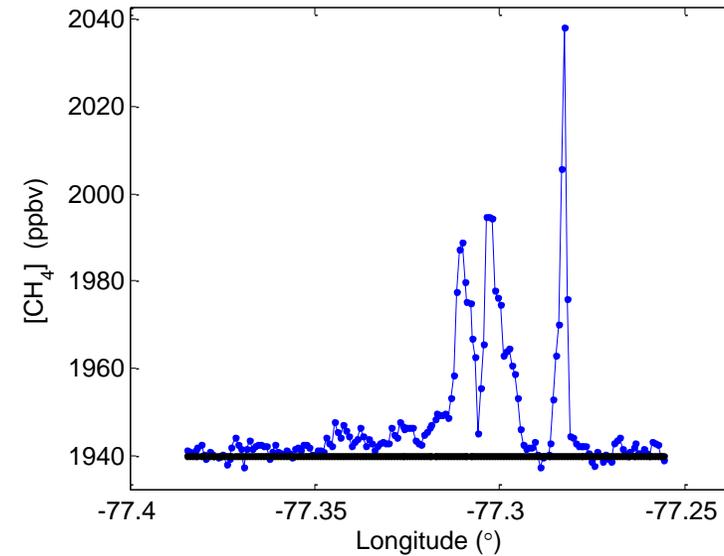
Mass Balance Experiment (MBE) approach:

$$E.R._{CH_4} = \int_0^{z_i} \int_{-x}^{+x} ([C] - [C]_b) \times U_{\perp} dx dz$$

[C] : concentrations (downwind)

[C]_b : concentration in background

U_⊥ : 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