GHG-Air Quality Synergies

Presented at NIST 7 November 2017 Russell Dickerson, UMD

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OMI SO₂

Li et al. *Scientific Reports* in press 2017.

Who's fallen to 2nd place?



ARIAS Experimental Locations 2016

_ Aircraft Flies Across Hebei Province; Spirals Over Ground Stations







Y12 Instrumentation



GPS Position (Lat, Long, Altitude)

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

Trace gases:

 O_3 : UV Absorption, modified TECO S O_2 : Pulsed Fluorescence, modified TECO $CH_4/CO_2/CO/H_2O$: Cavity Ringdown, Picarro NO_2 : Cavity Ring Down, Los Gatos NO/NO_y : Chemiluminescence, TECO VOCs: grab canisters/GC-FID

Aerosols:

Scattering: b_{scat} (@450, 550, 700 nm), Nephelometer Absorption: b_{ap} (565 nm), PSAP Black Carbon: Aethalometer, 7 λ Cloud Condensation Nuclei (CCN) Single Particle Soot Photometer (SP²) (MPI)

Evaluating emissions: Schematic of ratio method



Plume Detection Algorithm

- Adapted from NASA-LaRC ICARTT plume detection mechanism
- Method:
 - 1) use moving window (1 min of 1-s data) to conduct a linear regression analysis of data collected within the PBL (< 1500 m), i.e., ΔXX/ΔCO₂ ratios.
 - 2) select statistically significant ($R^2 > 0.6$) $\Delta XX / \Delta CO_2$ ratios.
 - 3) calculate the ratio distribution and create the histogram plots.
- This algorithm can automatically detect emissions plumes.

Hao He, UMD



All ARIAs measurements

 $\Delta XX/\Delta CO_2$ ratios histograms

1) $\Delta CO/\Delta CO_2$: high means inefficient combustion

2) ΔSO₂/ΔCO₂: is low
compared with our 2005 &
2008 flights. S/C in coal 13% says Scrubbing!

3) $\Delta CH_4/\Delta CO_2$: isolated plumes with high values observed.

4) $\Delta NO_2/\Delta CO_2$: episodes of large values (>0.1%) observed.

Emission Identification: CO to CO₂ ratios in Hebei



Characteristic Pollutant Emission Ratios

Efficient internal combustion

 $\Delta CO: \Delta CO_2 < 1\%$ $\Delta NOx: \Delta CO_2 < 0.1\%$ $\Delta SO_2: \Delta CO_2 << 0.1\%$

- High Tech coal combustion unscrubbed $\triangle CO: \triangle CO_2 << 1\%$ $\triangle NOx: \triangle CO_2 \sim 1\%$ $\triangle SO_2: \triangle CO_2 = 0.5-2\%$
- Biomass burning $\triangle CO: \triangle CO_2 5-10\%$ $\triangle NOX: \triangle CO_2 = 0.1 \text{ to } 1\%$ $\triangle SO_2: \triangle CO_2 < 0.1\%$ $\triangle CH_4: \triangle CO_2 = 0.2 \text{ to } 2\%$

Evaluating emissions inventories is essential.

- Borrow a classical technique for top down emissions estimates.
- First employed to study biomass burning in the Amazon (Crutzen et al., 1979) later for BC from India (Dickerson et al., 2002) and NOx in Baltimore (Anderson et al., 2014).

$$E_{CO} = \frac{\Delta CO}{\Delta CO_2} \times E_{CO2}$$

$$E_{NOx} = \frac{\Delta NOx}{\Delta CO} \times E_{CO} = \frac{\Delta NOx}{\Delta CO_2} \times E_{CO2}$$

Looking at ratios of short-lived pollutants to CO₂ can

show the use of control equipment,

indicate the efficiency of combustion,

help evaluate emissions inventories.



40% by 2030



Maryland Commission on

Climate Change

DRAFT Annual Report

Prepared for:

Larry Hogan, Governor

State of Maryland

and the Maryland General Assembly



November 2017

<u>Regional Atmospheric Measurement Modeling & Prediction Program</u>

RAMMPP: Balanced Theory & Observations





2016 FLAGG-MD flights in Balt/DC area.

 $\Delta XX/\Delta CO_2$ ratios histogram

- 1) $\Delta CO/\Delta CO_2$: highly efficient combustion
- ΔSO₂/ΔCO₂: Median similar to that measured in China but there are no spikes.
- 3) $\Delta CH_4 / \Delta CO_2$: lower values than in China. (Co-located sources)
- 4) $\Delta NO_2/\Delta CO_2$: similar values observed in China.



2016 FLAGG-MD flights in Indianapolis

 $\Delta XX/\Delta CO_2$ ratios histograms

- 1) $\Delta CO/\Delta CO_2$: similar to Balt/DC, but more spikes.
- ΔSO₂/ΔCO₂: much lower mode than Balt/DC, but spikes
- 3) $\Delta CH_4/\Delta CO_2$: similar as to Balt/DC.
- 4) $\Delta NO_2/\Delta CO_2$: slightly lower values than Balt/DC.

Example of CO₂ plumes captured by UMD Cessna



Overview of FLAGG-MD aircraft camapign 2015



Hourly flux comparison: FLAGG-MD vs. FFDAS



Monthly flux comparison: FLAGG-MD, CarbonTracker, FFDAS, ODIAC



ODIAC2016 (1 km) / Fossil / Feb 2015



FFDAS v2.0 (0.1°) / Fossil / Feb 2015







NASA's DISCOVER-AQ Maryland July 2011 & MDE's I-95 site







- Observations from I-95 NR site for October-November 2016
- Each dot represents an hourly emission ratio (not binned)
- Red line is the ordinary leastsquares regression of the hourly ratios



New York Fight: May 18, 2017 Afternoon CH₄ and CO₂



Primary pollutants CH_4 , CO_2 (& CO) as well as O_3 show higher conc's ENE of NYC/NJ.

Discussion points

CO was a good tracer when cars were dirty. Now isoprene oxidation is comparable to direct emissions in summer in the daytime. Calculating CO from isoprene oxidation is uncertain due to dependence on NOx and complexity of mechanism.

Cost benefit analysis for MD – saves lives thru reduced SO_2 , NOx and VOC's = PM. Success story – Brown Station Landfill CH_4 . States loved NASA's AQAST

Enhanced Ozone Monitoring Plan in the OTR

In open discussion EPA, States, MARAMA judge.robert@epa.gov

Add CO₂ and CH₄ Mixing Height Measurements (PBL vs Residual layer, Lidar etc.) Special Projects such as aircraft and ships.

Discussion Points Continued

What should be monitored? What should be measured? Who are out customers? What do models most need?

Nonattainment: Ozone, but

Epidemiologists tell us that 12 μ g/m³ (35 μ g/m³ for 24 hr) is **not** protective of human health.

Sources SO₂, NH₃, NOx, mineral dust, VOC's SO_2 controlled to ~90% CO controlled to ~90% NOx controlled for ozone but still to high NH₃ going up VOC's critical Uncertainty in US emissions opportunities for synergy CO₂ 6-20% CO 15-150% SO₂ 10-20% NOx Factor of 2 NH₃ Factor of 2+ VOC's 20% to factor of 10 BC = ?

The End



Fear the Turtle!

Reprints can be found at http://www.atmos.umd.edu/~russ/recent_pubs.html





Global NH₃ 2002-2015

Y12 Flight Tracks



Aircraft measurements during ARIAs Campaign



In general higher $[O_3]$ to the north and northwest area (Xingtai and Shijiazhuang) and lower $[O_3]$ over Julu and Quzhou. Up to 6000 ppb of [CO], up to 80 ppbv of $[SO_2]$, and up to 40 ppbv of $[NO_2]$ were observed. A few CH₄ hot spots on the northeast leg.

Power Plant in Hebei, China







From Crutzen and Andreae (1990)

CH4 (ppm)