What Can we Learn from Observations & Modeling to Inform Inventory Estimates?

Presented to
MARAMA
by
Russell R. Dickerson, Univ. Maryland

Supported by MDE
NASA, NOAA, and DNR
February 27, 2013

UMD/URF Cessna
Photo by J. Stehr

But first
An Announcement

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The NASA Air Quality Applied Sciences Team (AQAST): Exploitation of NASA data

Daniel J. Jacob, Harvard University, AQAST Leader

Annual Meeting: Open to MARAMA
June 4-6, 2013 College Park

AQAST
NASA Air Quality Applied Sciences Team

http://acmg.seas.harvard.edu/aqast

Air Quality Applied Sciences Team (AQAST)

EARTH SCIENCE SERVING AIR QUALITY MANAGEMENT NEEDS

Earth observing system

satellites

suborbital platforms

models

AQAST

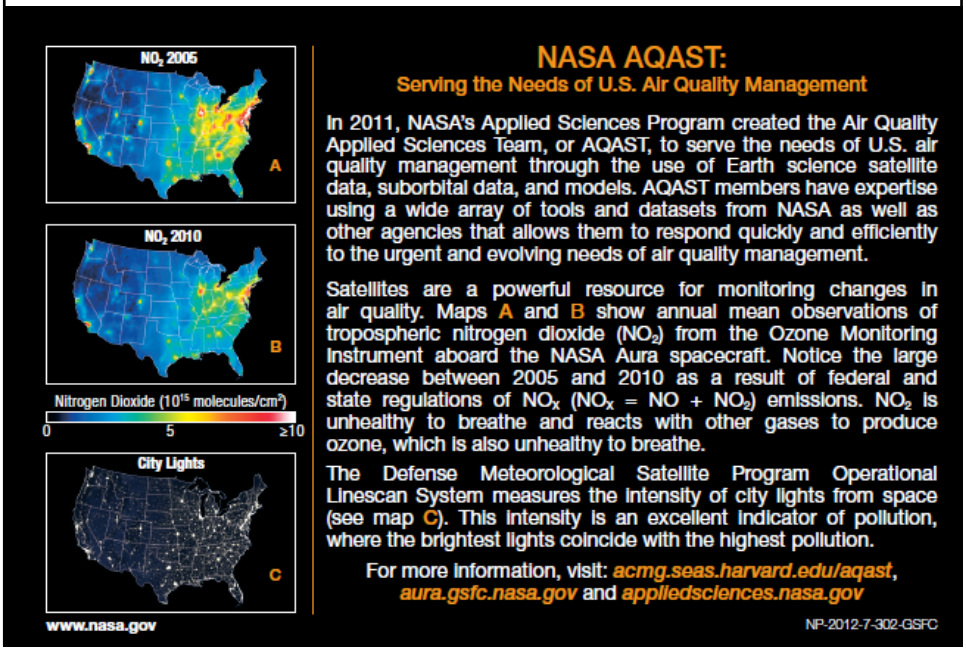
Air Quality Management Needs
- Pollution monitoring
- Exposure assessment
- AQ forecasting
- Source attribution of events
- Quantifying emissions
- Natural & foreign influences
- AQ processes
- Climate-AQ interactions

AQAST

AQAST members

- **Daniel Jacob (leader),** Loretta Mickley (Harvard)
- **Greg Carmichael** (U. Iowa)
- **Dan Cohan** (Rice U.)
- **Russ Dickerson,** Ross Salawitch, Tim Canty (U. Maryland)
- **Bryan Duncan**, Yasuko Yoshida, Melanie Follette-Cook (NASA/GSFC); Jennifer Olson (NASA/LaRC)
- **David Edwards** (NCAR)
- **Arlene Fiore** (NOAA/GFDL); Meiyun Lin (Princeton)
- **Jack Fishman,** Ben de Foy (Saint Louis U.)
- **Daven Henze**, Jana Milford (U. Colorado)
- **Tracey Holloway,** Steve Ackerman (U. Wisconsin); Bart Sponseller (Wisconsin DRC)
- **Edward Hyer,** Jeff Reid, Doug Westphal, Kim Richardson (NRL)
- **Pius Lee,** Tianfeng Chai (NOAA/NESDIS)
- **Yang Liu**, Matthew Strickland (Emory U.), Bin Yu (UC Berkeley)
- **Richard McNider**, Arastoo Biazar (U. Alabama – Huntsville)
- **Brad Pierce** (NOAA/NESDIS)

AQAST lenticular produced by Bryan Duncan (ask for it!!)



Outline

Evaluating emissions estimates: Questions.

1. What is the uncertainty associated with emissions inventories? Plain English: What range of answers do the data allow?

2. If the emissions estimates are wrong does it matter?

3. Are the trends right?

4. Do models get O_3 and NO_2 right for the right reasons?

5. Which sources matter most?

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Outline

Content of this talk:

1. Trends in surface observations; do satellite obs agree?

2. RAMMPP & DISCOVER-AQ

   Model (CMAQ) – measurement comparisons.

3. Ratios of observed concentrations*.

*Point source emissions agree with in situ obs to <20%, but this is a separate talk.

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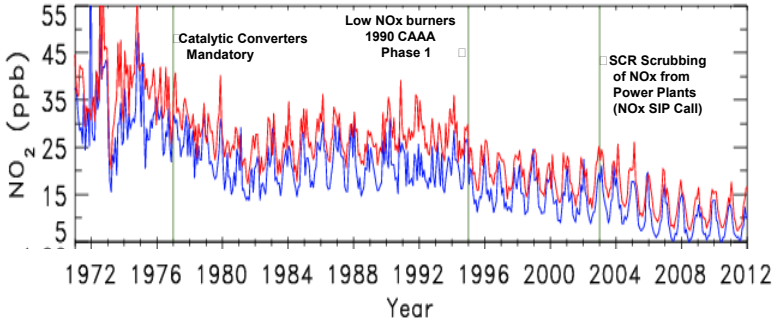
Recent UMD Trend Analysis
Tracking NO_X Reductions and Ozone Improvements

Ambient NO_2 concentration trends: DC, MD and Northern VA



Red - Weekdays
Blue - Weekends

From Hosley, Salawitch, Canty, et al. in preparation, 2012; Preliminary Data. Do not cite!

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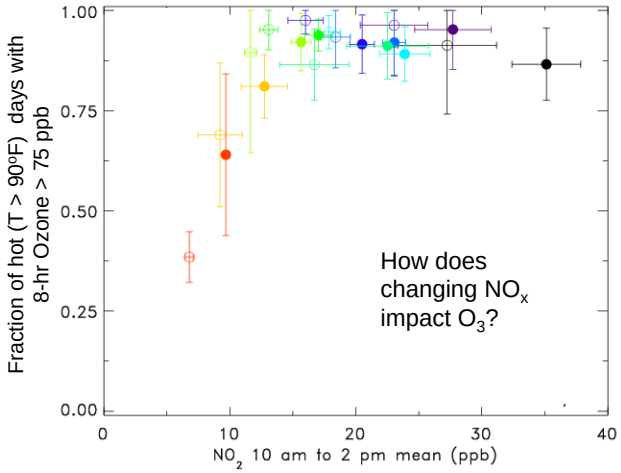
As measured NO_x levels have gone down …
… *So have ambient ozone levels*



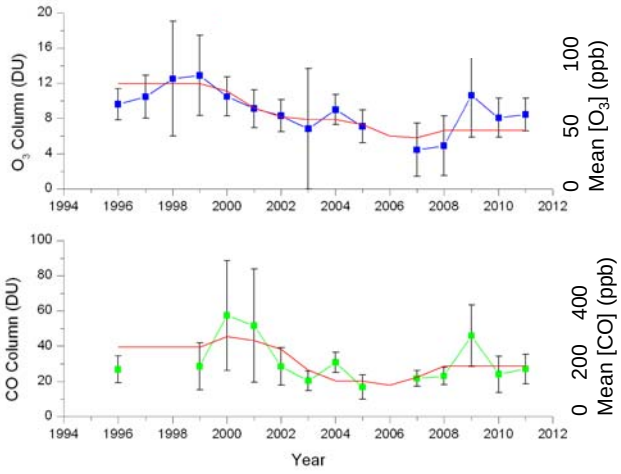
Observations show:
NOx reductions worked, but response is nonlinear;
we had to get over the hump.

From Hosley, Salawitch, Canty, et al. in preparation, 2012; Preliminary Data. Do not cite!

Transported Ozone Aloft Has Also Improved

15 year aircraft record shows that aloft ozone (throughout the lowest 4500' of the atmosphere) has improved.

Carbon monoxide has also cleaned up - NASA satellite confirmation.



Hao He, Ph.D. Dissertation 2012.

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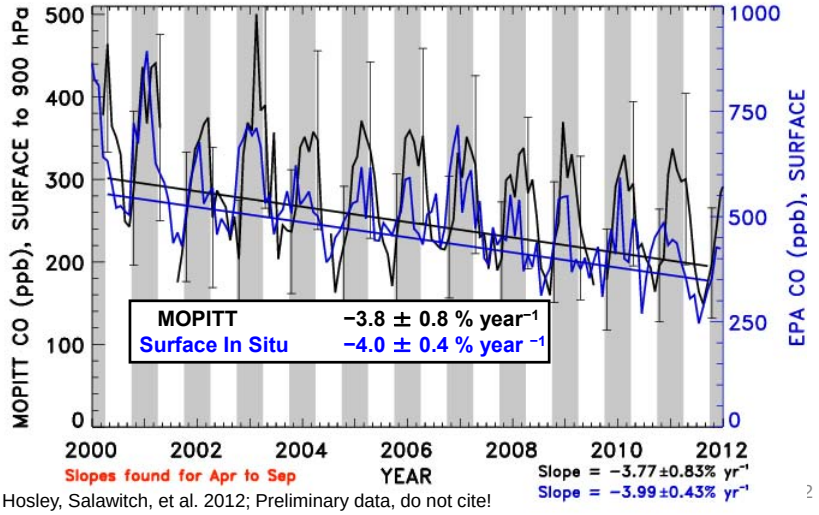
Satellites and Air Quality

An emerging tool

NASA Satellite (MOPITT) CO over Greenbelt, MD
Confirms wide spread improvements in CO



Canty, Hosley, Salawitch, et al. 2012; Preliminary data, do not cite!

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Surface and Satellite observations confirm the trends of concentrations.
But what about the Emissions?

July 2011 as a case study.

RAMMPP

(Regional Atmospheric Measurement Modeling & Prediction Program)

Balanced Theory & Observations

WRF
Dynamical Model
12-0.5 km Resolution
Forecasting

Input
Emissions Inventories
SMOKE
(Chem Engineering)

Chemical Transport Models

CMAQ
Modular
Open Code
Collaborative w/EPA
and
WRF-CHEM
Interactive
Photochem.
Aerosols
Transport
Deposition

Observations

Surface:
- Beltsville
- Greenbelt
- Piney Run

Aloft
- Cessna Aircraft
- NASA P3
- Profiler
- Sondes

Remote (NASA & NOAA)
- AERONET
- GASP (AOD)
- MOPITT (CO)
- MODIS (particles)
- OMI (NO_2, SO_2)
- PANDORA (NO_2, SO_2)
- SCIAMACHY (SO_2, NO_2, H_2CO…)

UMD Research Aircraft

GPS Position (° Lat, ° Long)

Meteorology (T, RH, Pr, P_{alt})

Carbon Monoxide (CO)

Nitrogen Dioxide (NO_2)

Ozone (O_3)

Sulfur Dioxide (SO_2)

Aerosol Optical Properties:

Absorption, b_{ap} (565 nm)

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

Aerosol Chemistry (NIST)

Major ions and SOA.

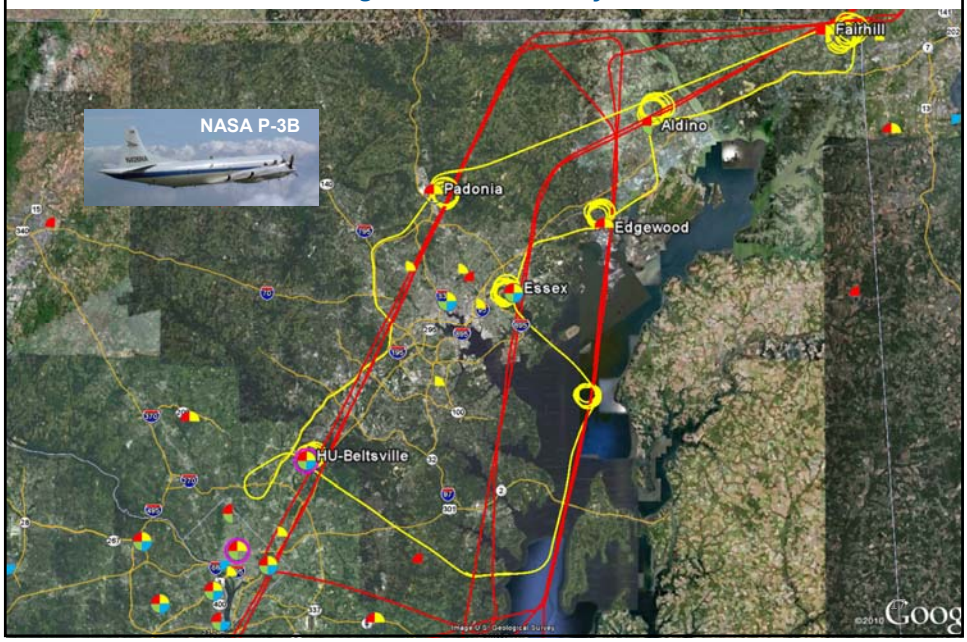
DISCOVER-AQ

July 2011

- 3 Aircraft
- 1 Ship
- 2 satellites
- 2 ground-based remote networks
- Many Ground stations
- $ 30M

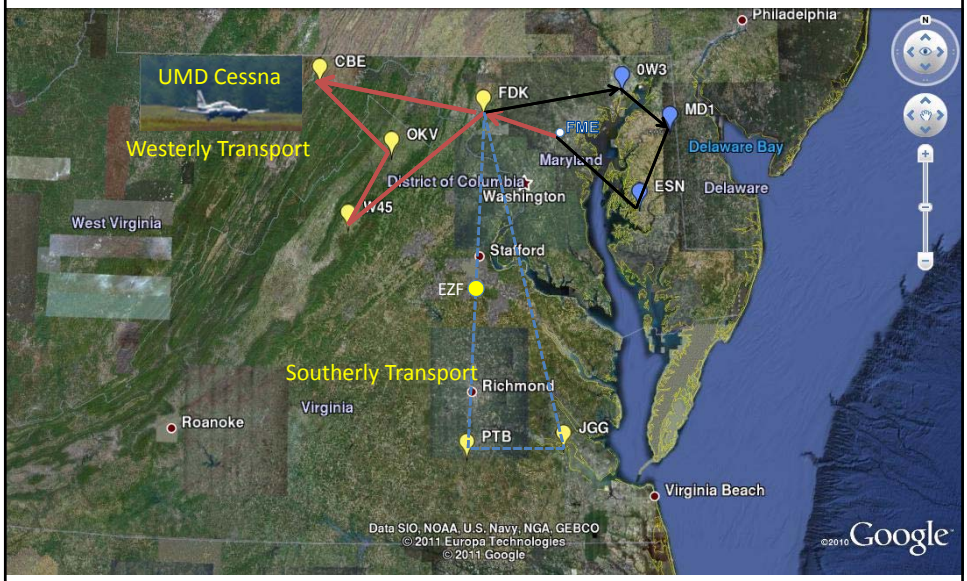
Foremost air pollution field study in the Mid-Atlantic region ever.

NASA Aircraft flew spirals over ground sites all day.



UMD Cessna in RAMMPP during DISCOVER-AQ flew spirals over a larger area.

AM ⟵ ⟶ PM



The Regional Nature of Ozone Formation.

Results from the RAMMPP Aircraft in DISCOVER-AQ show that NOx is already high upwind of Baltimore.

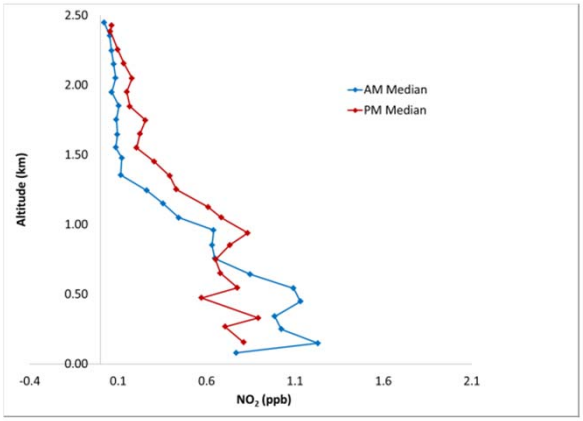


Figure 4.8.3. Morning (upwind of the Baltimore/Washington area) vs. afternoon (downwind) NO_2 profiles from DISCOVER-AQ as measured from the UMD Cessna. Little difference is observed in the observed column content (altitude integral) indicating the regional nature of pollutants [L. Brent et al., manuscript in preparation; 2012].

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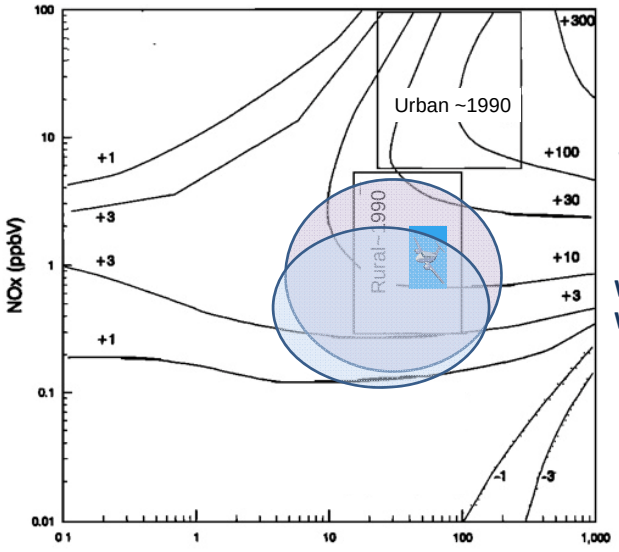
How fast do precursor pollutants make ozone (ppb/hr)?



Where is the Balt/Wash area? (boundary layer)

Where is Western MD?

Smog chamber and modeling results on O_3 formation rates.

Copyright © 2012 R.R. Dickerson based on Chameides et al., 1992

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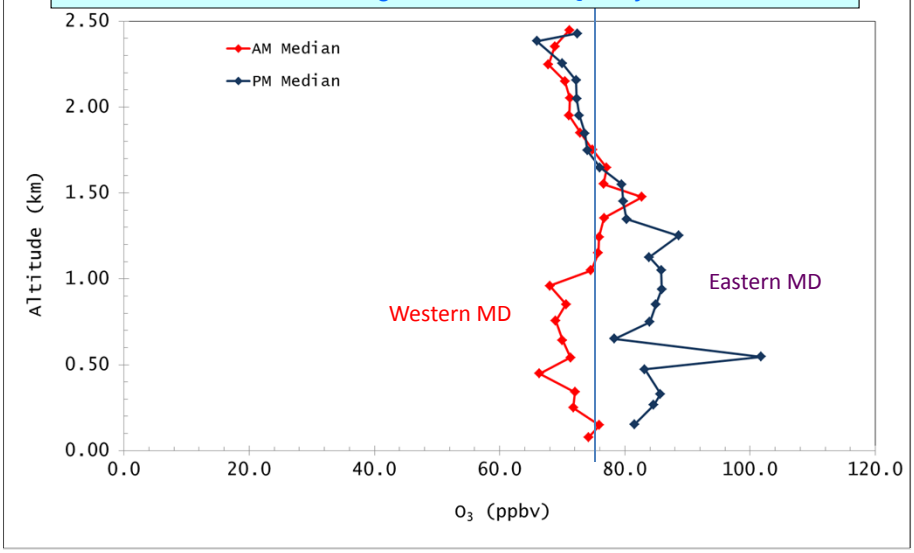
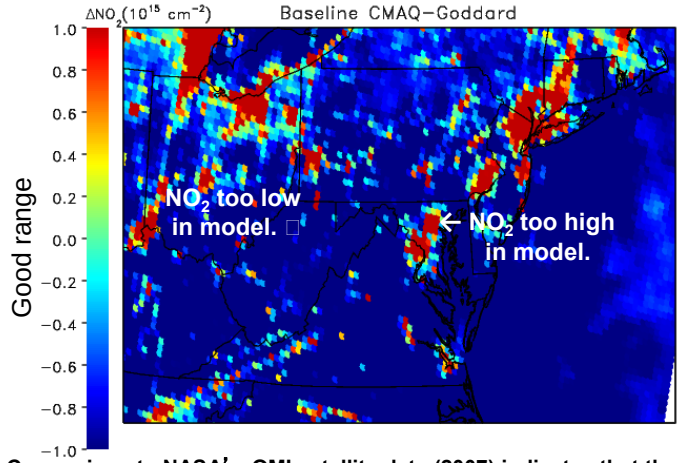
Ozone observed over western Maryland in the morning and over eastern Maryland (downwind of Baltimore and Washington) in the afternoon during DISCOVER-AQ, July 2011



NASA Satellite observations suggest enhancements in CMAQ/CB05 chemistry. The models have improved and we will make them even better.



Comparison to NASA's OMI satellite data (2007) indicates that the ratio of urban/rural NO_2 is too high in the model (CMAQ, L2 emissions) simulation.

Thanks to NASA support through the AURA Science Team and AQAST.

From Canty, et al. in preparation, 2012

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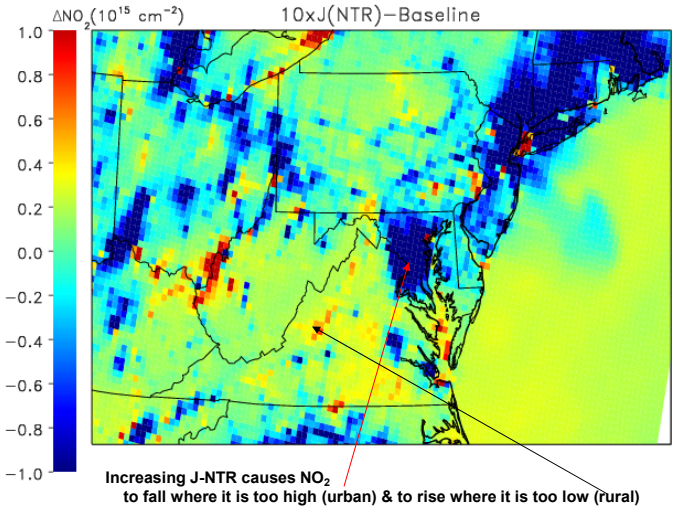
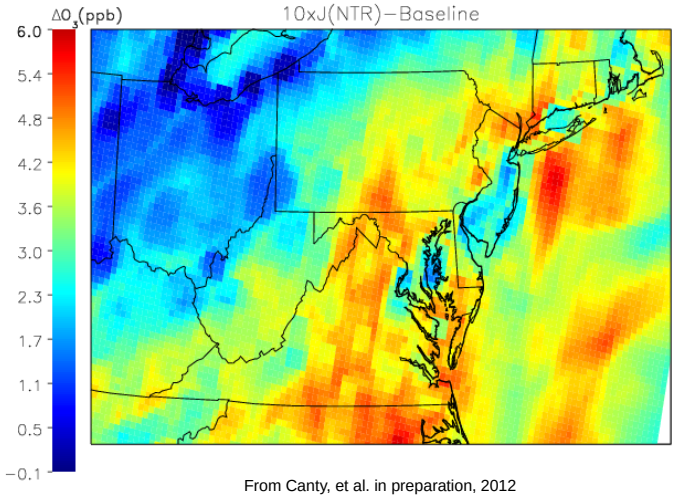
Improving the simulation of chemistry in CMAQ moves more NO_X to rural areas and brings the model closer to NO_2 observations. CB05 was designed with adjustable NOx chemistry.



From Canty, et al. in preparation, 2012

23

Improving the simulation of chemistry in CMAQ moves more NOx to rural areas **and shows more ozone in rural areas in agreement with observations**.



From Canty, et al. in preparation, 2012

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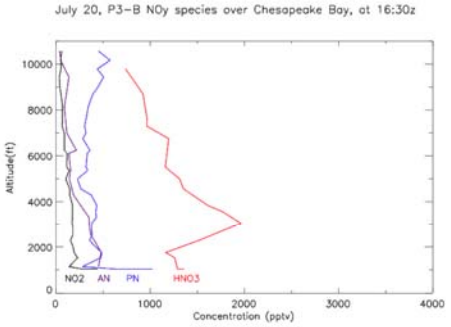

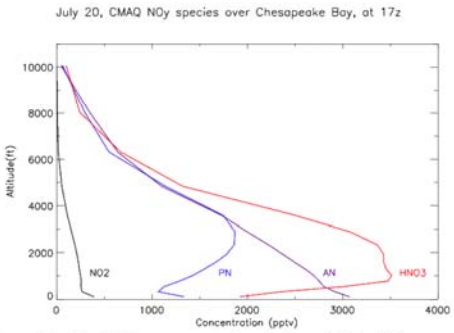

Reactive N Species Over the Chesapeake Bay

From Goldberg et al. in prep., 2013.

Observed

CMAQ modeled

NO_2 agrees, but NOy too high.

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Preliminary Conclusions:

- NOx emissions too high
- Removal rate too fast and/or
- Recycle rate too slow.

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Let's look at ratios

- EPA inventories estimate a NO_y/CO ratio of136 mmol/mol (CO/NOx ~ 7).
- Previous research suggests inventory ratios of NO_y/CO are an overestimate:
  - Fujita (2012) – models overestimate concentrations by 25-40%
  - Parrish (2006) – Inventories are a factor of 2 larger than measurements
- Research questions:
  - What are the emissions ratios of pollutants - particularly NO_y, CO, and CH_4 - to CO_2 and CO in Maryland?
  - How well do emissions inventories represent these ratios?

Preliminary Data. Do not cite.

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Methodology

- Identified 70 spirals from DISCOVER-AQ P3B flights with areas of correlated CO and NO_y concentration.
- Determined mixed layer from vertical profiles of relative humidity and equivalent potential temperature.
- Calculated, for all measured compounds in the mixed layer, $\Delta X/\Delta CO_2$ and $\Delta X/\Delta CO$.
- Included only those correlations with $r^2 > 0.8$ and with > 10 data points.

Preliminary Data. Do not cite.

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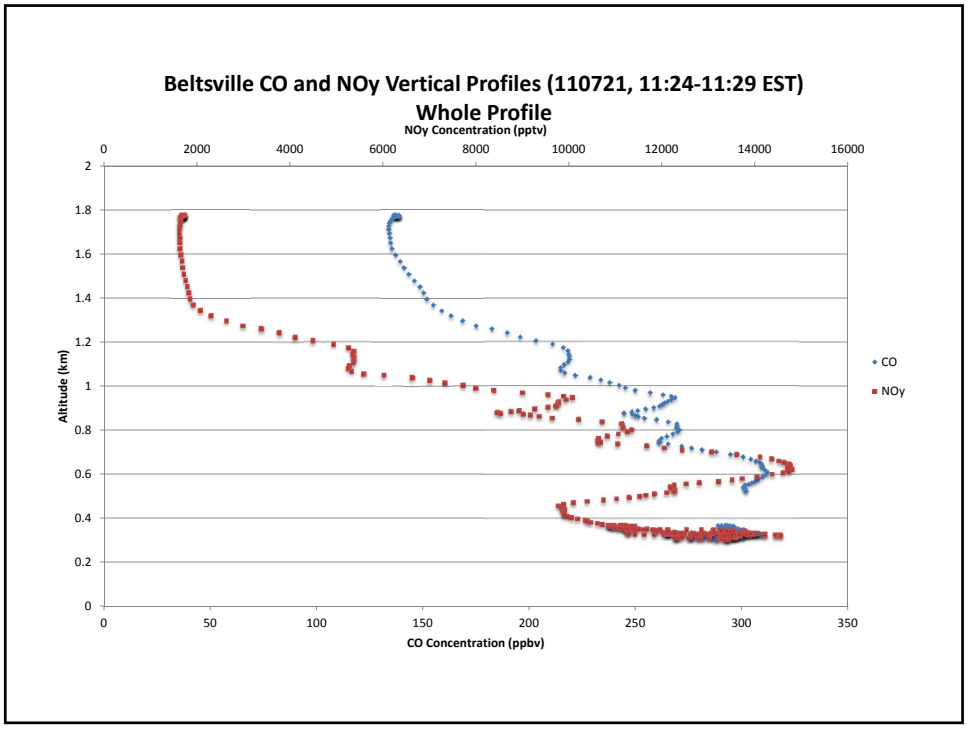
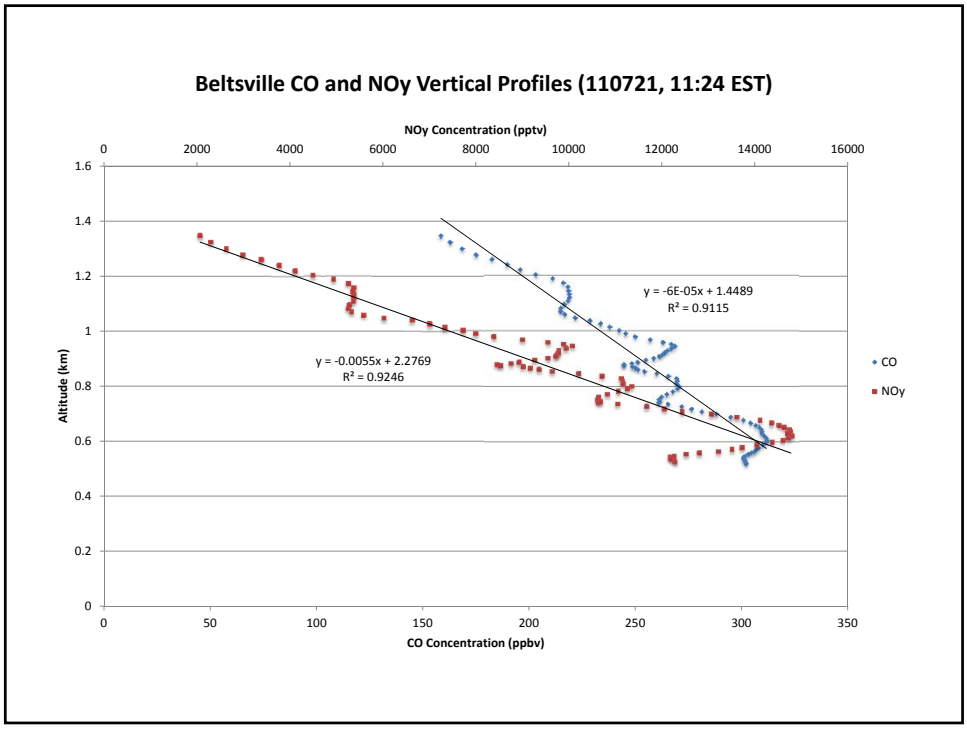
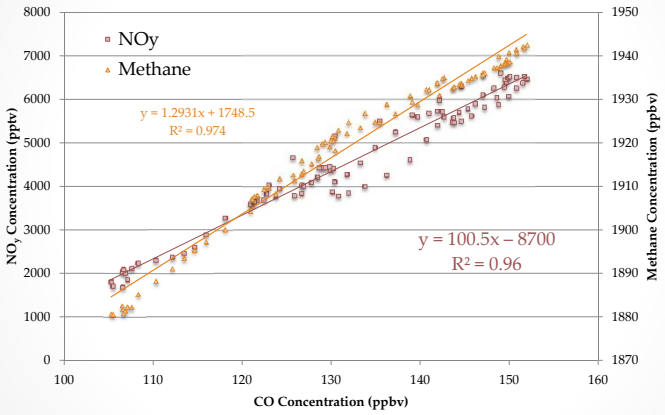

Beltsville CO and NOy Vertical Profiles (110721, 11:24-11:29 EST)
Whole Profile
NOy Concentration (pptv)
0
2000
4000
6000
8000
10000
12000
14000
16000
Altitude (km)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
1.8
2
CO Concentration (ppbv)
0
50
100
150
200
250
300
350
CO
NOy


Beltsville CO and NOy Vertical Profiles (110721, 11:24 EST)
NOy Concentration (pptv)
0
2000
4000
6000
8000
10000
12000
14000
16000
Altitude (km)
0
0.2
0.4
0.6
0.8
1
1.2
1.4
1.6
CO Concentration (ppbv)
0
50
100
150
200
250
300
350
y = -6E-05x + 1.4489
R² = 0.9115
y = -0.0055x + 2.2769
R² = 0.9246
CO
NOy

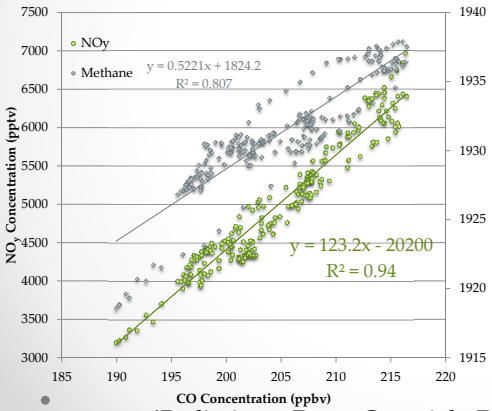
Essex, 07/26, 9:13 EST

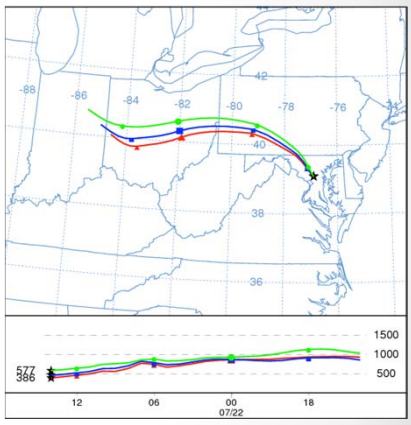


Preliminary Data. Copyright D. Anderson & R. Dickerson, UMD

Beltsville, 110722, 949-979 hPA, 10:05 EST

Air mass from Ohio River Valley. NO_y/CO ratio slightly higher than previous profile.

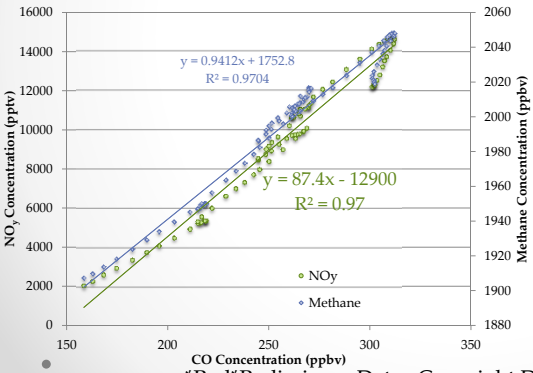


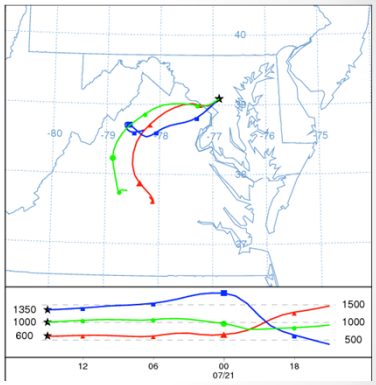


Preliminary Data. Copyright D. Anderson & R. Dickerson, UMD

Beltsville, 110721, 868-953 hPa, 11:27 EST

Air mass from Virginia. NO_y/CO ratio slightly lower than previous profiles.





*Prel*Preliminary Data. Copyright D. Anderson & R. Dickerson, UMD*

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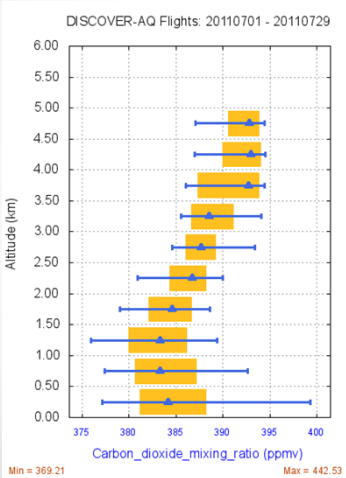
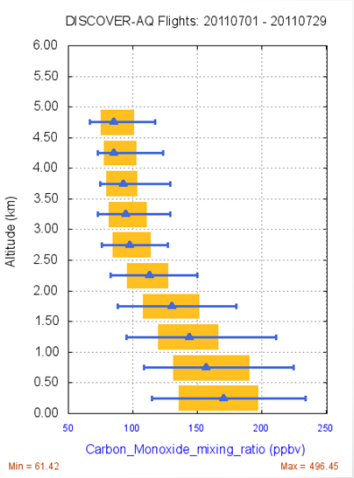
DISCOVER-AQ Average CO_2 and CO Profiles





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Summary of Emissions Ratios

	DISCOVER-AQ Average (mmol/mol) $\pm \sigma/n^{0.5}$	Number of Data Points	Fujita *et al* 2012 (mmol/mol)	EPA (mmol/mol)	EPA/DISCOVER-AQ
NOy/CO	**72.8 ± 7.8**	**60**	**108**	**136⁺**	**1.9**

*: Values for 2010 +: Values for 2011; CO & NO_y data from NEI. CO_2 data from 2012 EPA US GHG Inventory.

- Fujita study only measured emissions from on-road transportation. Values lie in-between that of DISCOVER-AQ and the EPA.
- NOy/CO ratio from DISCOVER-AQ in line with work of Parrish.
- **NEI appears to overestimate NOx emissions by a factor of 1.9.**

Preliminary Data. Copyright D. Anderson & R. Dickerson, UMD

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Has this been seen before?

New paper by the EPA RTP scientists [Yu *et al.*, 2012]
Compares CMAQ (WRF; CB4.2; Mobile 6 and BEIS) to the TEXaqs 2006 observations. They conclude:

"The comparisons with measurements at the AIRNow surface sites revealed that ARW–CMAQ and NMM– CMAQ have a very similar good performance for the high maximum 8–hr O_3 concentration range (>40 ppbv) with slightly better performance for ARW–CMAQ."

The paper shows that CMAQ had a 45% high bias when 8-hr O_3 is <40 ppb and a 9.5% low bias when O_3 is > 75 ppb.

Compared to P3 obs in the lowest 200m, the model:
- Does well for CO (124 observed vs. 117 ppb modeled)
- **Overestimates NOy (9.2 vs. 4.6 ppb) and all NOy constituents.**
- Shows the OPE substantially less than observed from O_3 vs. NOz (8 vs. 3).

Yu, S. C., et al. (2012), Comparative evaluation of the impact of WRF-NMM and WRF-ARW meteorology on CMAQ simulations for O_3 and related species during the 2006 TexAQS/GoMACCS campaign, *Atmospheric Pollution Research*, 3(2), 149-162.

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Conclusions: What can observations tell us about emissions?

- NOx and CO have come down nicely over the past few decades. Emissions *trends* look good.
- CMAQ with CB05 overestimates [NOy] and NOy/CO (factor of ~2) in urban areas, probably due to overestimated NOx emissions.
- Satellites and *in situ* data agree that CMAQ overestimates NOx in urban areas, but underestimates in rural areas, probably because of NOx recycling. Canty is currently rerunning CMAQ with L3 and improved CB05.
- Lower NOx puts the Mid Atlantic States on the steeper part of the ozone production curve: NOx controls more effective!

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Back to Outline

Evaluating emissions estimates: Some answers:

1. What is the uncertainty? CO looks pretty good now, but NOx may be no better than a factor of 2.

2. If the emissions estimates are wrong does it matter? Yes, if you want to model the spatial and temporal scales of ozone events.

3. Are the trends right? They look good.

4. Do models get O_3 and NO_2 right for the right reasons? Overestimated removal rates can compensate for overestimated NO release rates.

5. Which sources matter most? **Longer effective lifetimes for NO_X make interstate transport important.**

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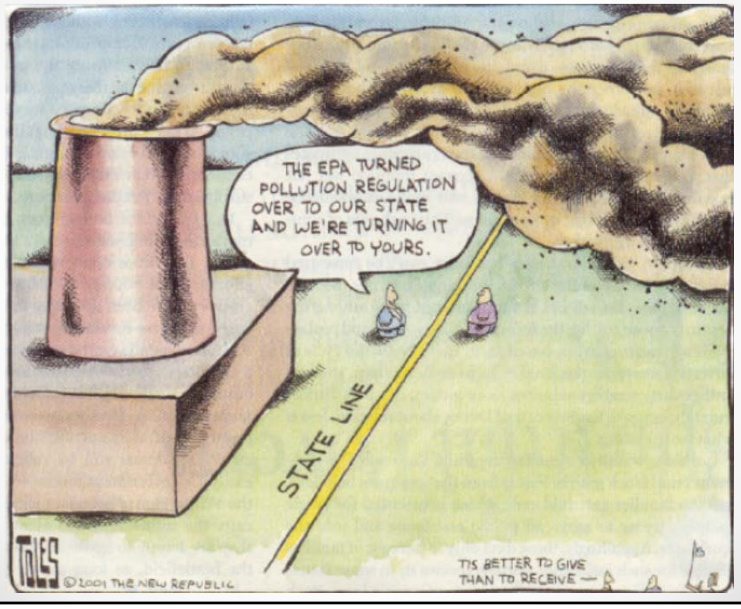
RAMMPP: Policy Relevant Air Quality Science

The MDE/UMD RAMMPP Team
Including NASA, NOAA, PSU, UMBC, DNR.

MDE 39

Life as a Downwind State



End

References, web sites, and acknowledgments

http://www.atmos.umd.edu/~RAMMPP/archives/2011data.html

http://www-air.larc.nasa.gov/missions/discover-aq/discover-aq.html

http://airquality.weather.gov/sectors/conus.php

http://acmg.seas.harvard.edu/aqast/

Comparison of the MOVES2010a, MOBILE6.2, and EMFAC2007 mobile source emission models with on-road traffic tunnel and remote sensing measurements, E.M. Fujita, et al., *J. Air & Waste Manage. Assoc.*, 2012.

Characterization of eastern U.S. air pollution episode using WRF/Chem, E. A. Yegorova, D. J. Allen, C. P. Loughner, K. E. Pickering, R. R. Dickerson, *J. Geophys. Res.*, 116, D17306, doi:10.1029/2010JD015054 , 2011.

Gilliland, A. B., C. Hogrefe, R. W. Pinder, J. M. Godowitch, K. L. Foley, and S. T. Rao (2008), Dynamic evaluation of regional air quality models: Assessing changes in O_3 stemming from changes in emissions and meteorology, Atmos. Environ. 5110-5123.

Thanks: D. Allen, D. Anderson, H. Arkinson, T. Canty, D. Goldberg, D. Krask, D. Baker, S. Ehrman, E. Yegorova, L. Hembeck, Hao He, K. Hosley, R. Hudson J. Hains, K. Vinnikov, Lacey Brent, L. Warren , C. Loughner, M. Woodman, K. Pickering, W. Ryan, R. Salawitch, L. Sparling, J. Stehr, J. Szykman, T. Vinciguerra, D. Willan, DL. Zhang.

Extra slides

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