

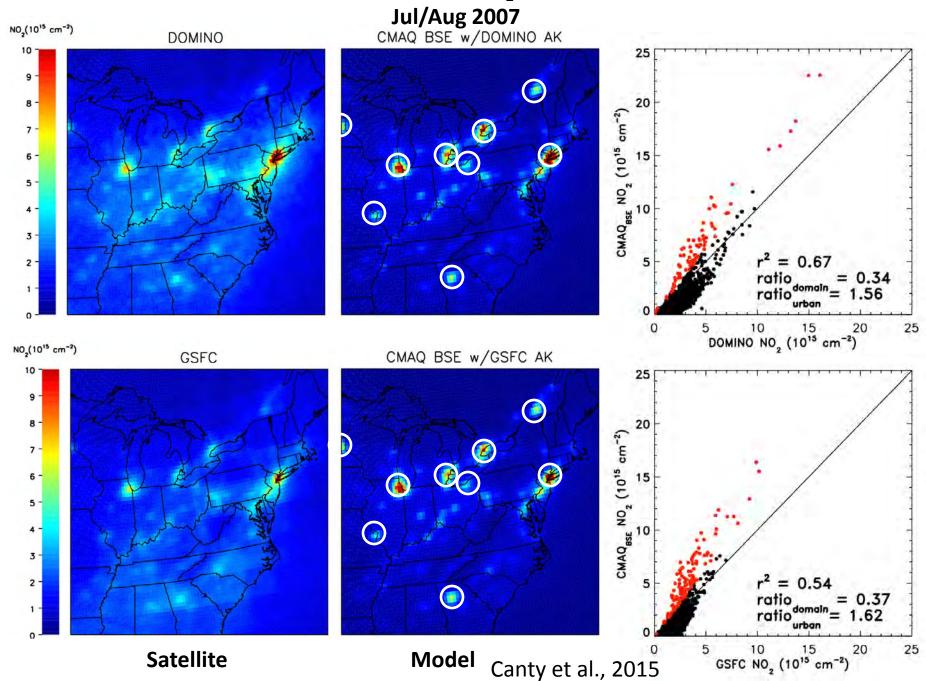


MDE/UMD Air Quality Modeling Update: Improved representation of O_3 precursors

Dan Goldberg, Tim Vinciguerra, Hao He, Tim Canty, Russ Dickerson

EPA Call Preliminary

OMI tropospheric column NO₂ vs CMAQ Baseline



How well are NO_x reservoir species simulated?

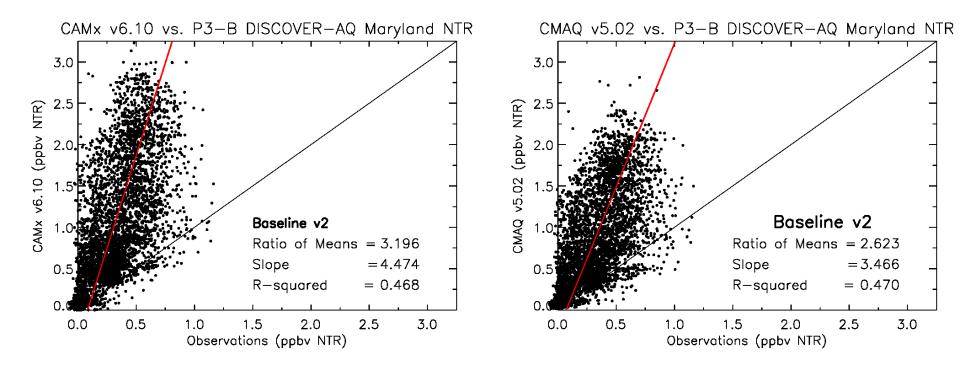
• Alkyl nitrates (AN), including isoprene nitrates, represented as single species (NTR)

NTR, treated as isopropyl nitrate, lost by photolysis & rxn w/ OH

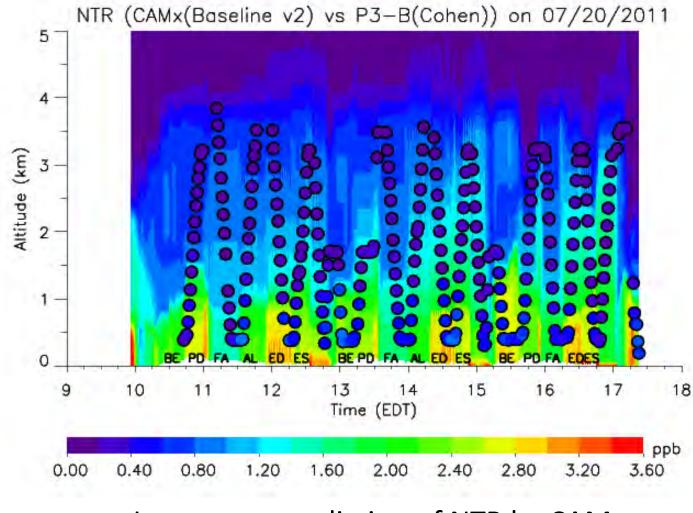
- In CMAQ, ~10 day lifetime
- Evidence that NTR may be comprised of short lived hydroxynitrates with lifetimes ~1 day (Beaver et al., 2012; Perring et al., 2009, and others)

Decrease lifetime of NTR may increase rural NO₂

Fortunately, we can compare aircraft observations during DISCOVER-AQ to CMAQ model run for 2011.

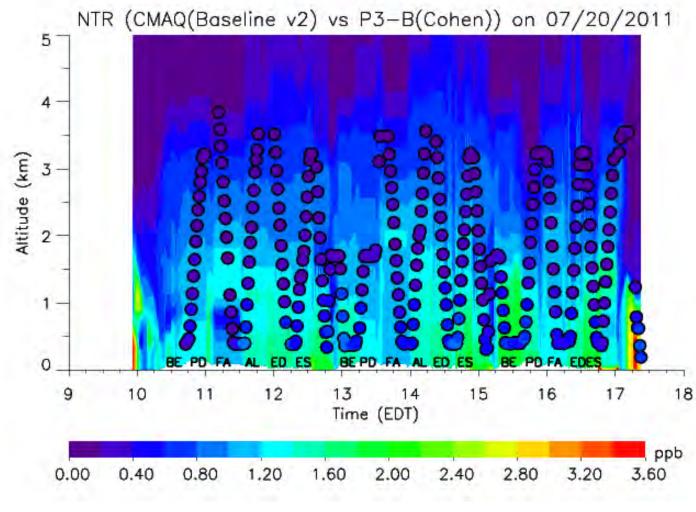


- CAMx v6.10 and CMAQ v5.02, in their baseline model set-up (CB05 and Version 2 emissions), over predict alkyl nitrates mixing ratios.
 - CAMx is 220% too high.
 - CMAQ is 160% too high.



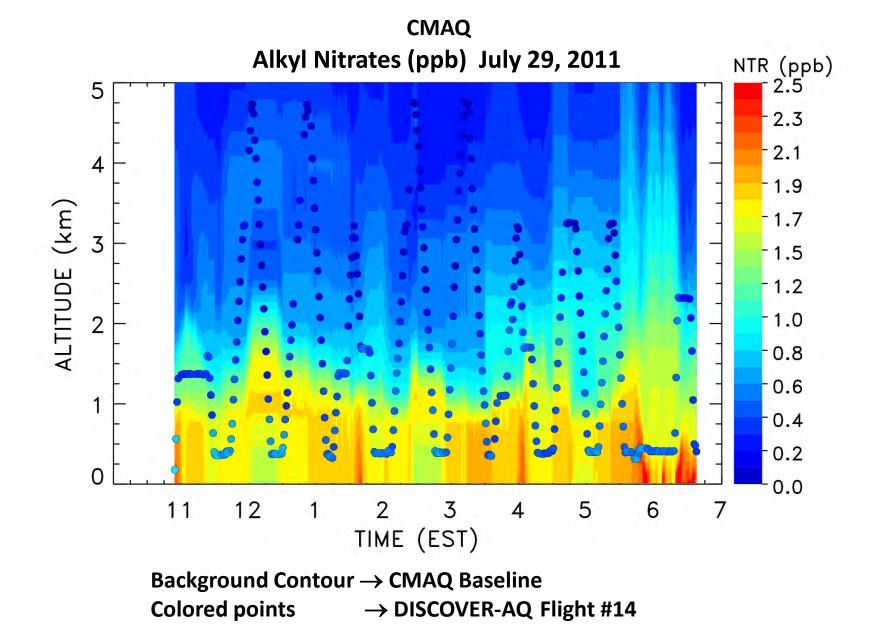
Large over prediction of NTR by CAMx.

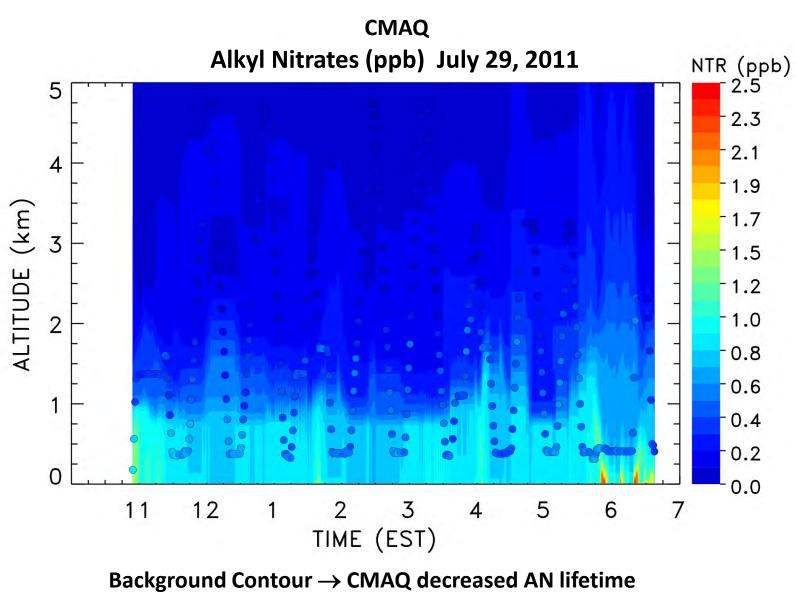
*There are *several* other days during DISCOVER-AQ when there is a similar over prediction.



Large over prediction of NTR by CAMx and CMAQ.

*There are *several* other days during DISCOVER-AQ when there is a similar over prediction.



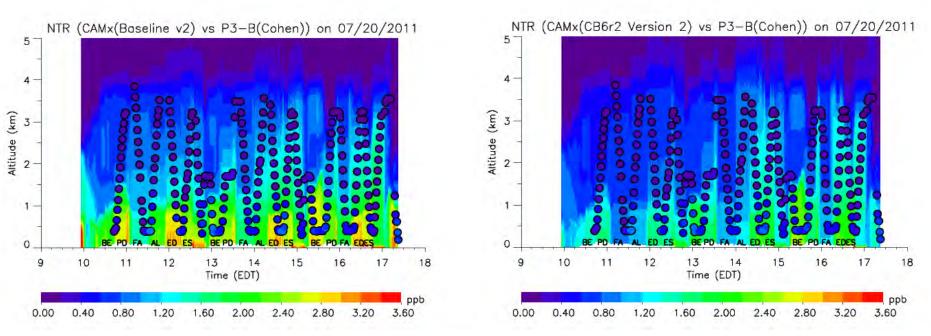


Colored points \rightarrow DISCOVER-AQ Flight #14

Some improvement in NTR simulation when using CB6r2

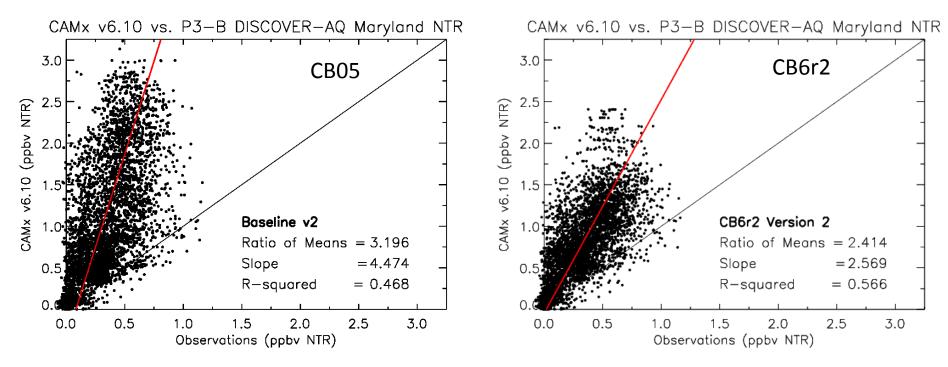
CB05

CB6r2



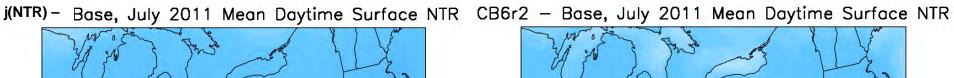
- Overestimate is reduced, but further improvements are still needed
- Some suggestions:
 - Even faster recycling to NO_x, aerosols, etc.
 - Faster NTR dry deposition rates

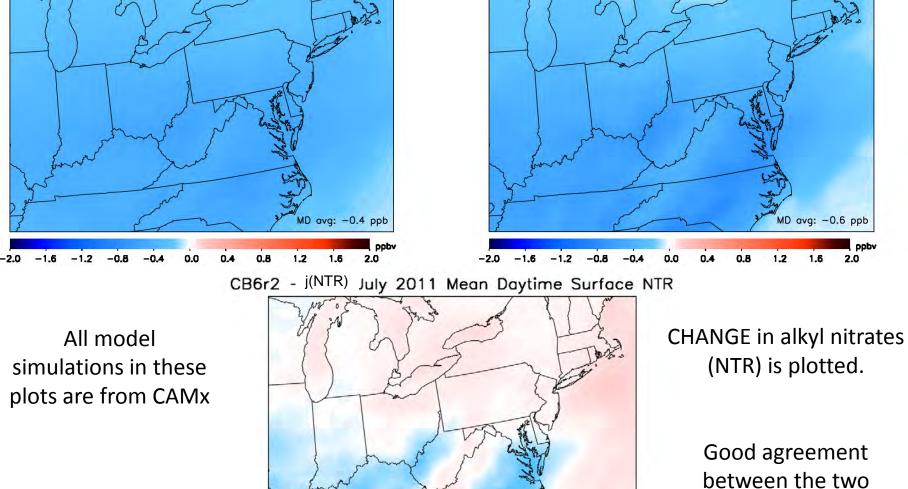
Some improvement in NTR simulation when using CB6r2



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Comparing CB6r2 chemistry to j(NTR)*10 method used in CMAQ/CB05





MD avg: -0.2 ppb

1.6

1.2

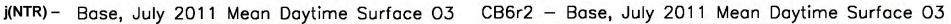
onby

2.0

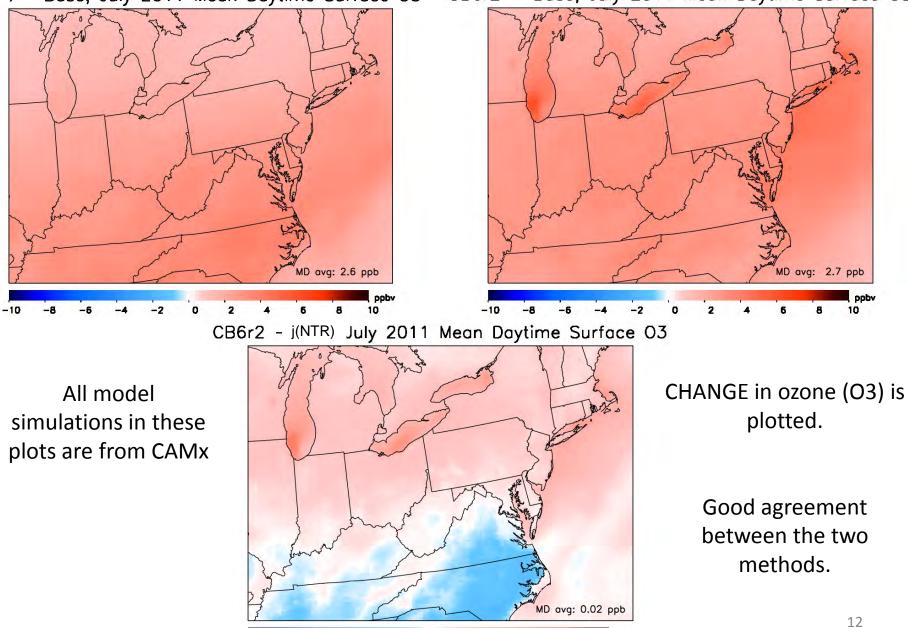
11

methods.

Comparing CB6r2 chemistry to j(NTR)*10 method used in CMAQ/CB05



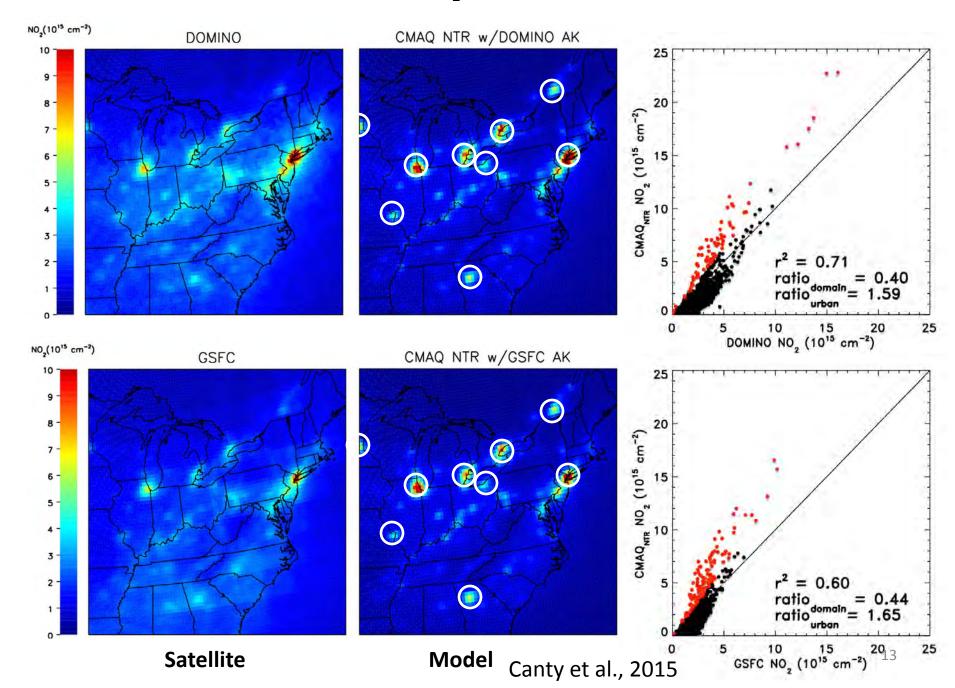
-10



ppbv

10

OMI tropospheric column NO₂ vs CMAQ w/ modified chemistry



Problem:

- NO_v simulation is over predicted by a factor of two
 - Anderson et al., 2014; Goldberg et al., 2014; Yu et al., 2012; Brioude et al., 2013; Doraiswamy et al., 2009; Fujita et al., 2012. Figure from <u>Anderson et al., 2014</u>:

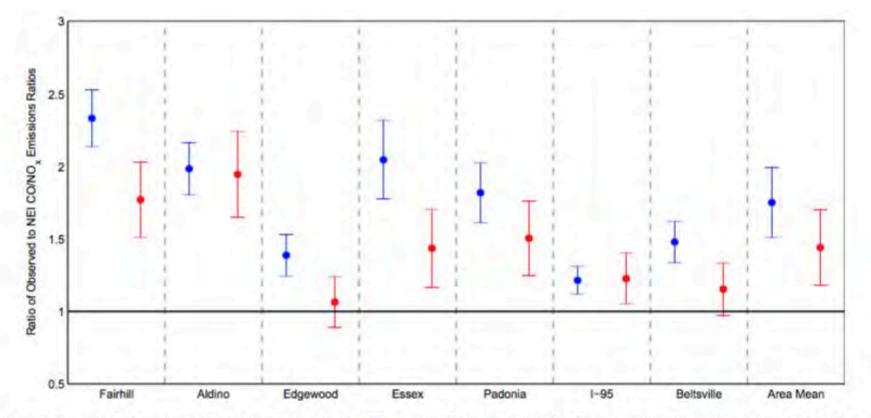


Fig. 10. Ratio of observed CO/NO_x emissions ratios to those predicted by the NEI by location. Values are corrected for CO uncertainties. Blue are derived using the linear least squares method, and red are derived with an orthogonal linear regression. Uncertainties are the 1*a* uncertainties in the NEI values and observations added in quadrature. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

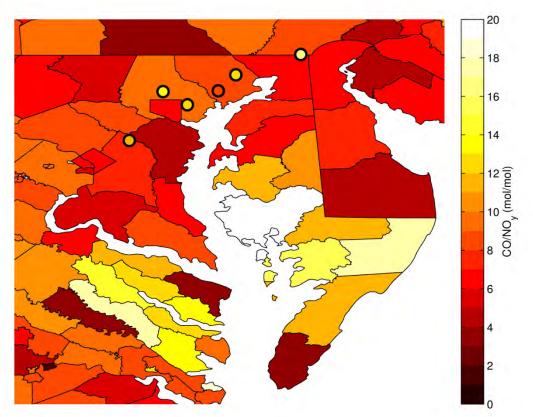
Emissions inventories

Observations of CO/NO_y (colored points) during DISCOVER-AQ ~1.75 times larger than National Emissions Inventory (NEI, colored counties) data used in CMAQ.

Modeled CO 25% larger than observations.

Indicates emissions inventoires may overestimate NO_v

Correct for this by reducing mobile NO_x emissions by 50%



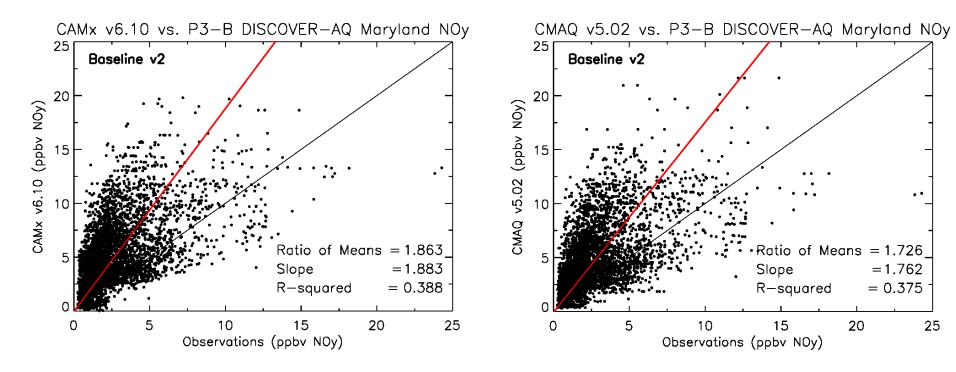
Anderson et al., 2014

Emissions inventories

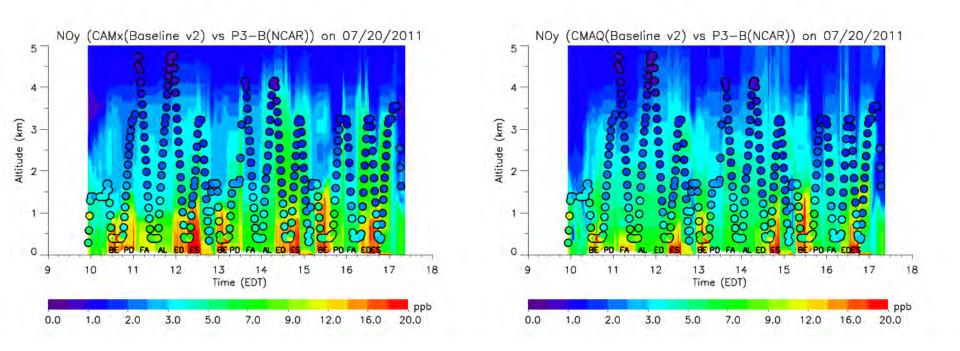
Table 4. Observations and models (NMM–CMAQ and ARW–CMAQ) for different gaseous species (O₃, CO, PAN, NOx, NO, NO₂, HNO₃, NO_n, ethylene, NO₄, air temperature (°C), water vapor (g/kg) and NO₂+O₃ (for lowest 4 layers only)) on the basis of all NOAA P=3 aircraft measurements over the Texas during the 2006 TexAQS (mean ± standard deviation, all units are ppbv except that PAN unit is pptv). Correlations between O₃ and NO₂ for the NO₄-limited conditions indicated by the observational data with (O₃)/(NO₄)>46 (aged air masses) (see text for explanation)

	Mean ± standard deviation			NMB (%)	
	Obs	NMM-CMAQ	ARW-CMAQ	NMM-CMAQ	ARW-CMAQ
03	53.27±17.68	58.27±10.39	56.94±11.39	9.4	6.9
со	124.05±42.8	118.05±49.24	115.87±48.87	-4.8	-6.6
PAN	448.30±316.8	805.17±556.84	781.99±572.24	79.6	74.4
NOz	1.51±2.05	3.76±7.05	4.11±8.46	149.0	172.2
NO ₂	1.24±1.74	3.15±5.97	3.26±6.44	154.0	162.9
NO	0.24±0.41	0.58±1.26	0.81±2.61	141.7	237.5
HNO ₃	1.33±1.12	1.89±1.50	1.79±1.42	42.1	34.6
NOv	4.61±3.33	9.01±8.17	9.35±9.87	95.4	102.8
Ethylene	0.73±0.87	0.41±0.59	0.40±0.61	-43.8	-45.2
NO,	2.57±1.70	4.20±2.44	4.01±2.41	63.4	56.0
NO ₂ +O ₃	57.13±26.26	60.71±11.71	60.62±13.39	6.3	6.1
Temperature	20.02±7.18	19.58±7.16	19.09±7.09	-2.2	-4.6
QV	10.13±5.40	9.89±5.32	9.56±4.75	-2.4	-5.6
Obs:	(Os)=8.4(NOz)+36.9	r=0.65			
ARW-CMAQ:	(Os)=3.4(NOz)+47.3	r=0.86			
NMM-CMAQ:	(O3)=2.7(NOz)+50.3	r=0.82			

Observed CO/NO_y = 26.9 Modeled CO/NO_y = 13.1



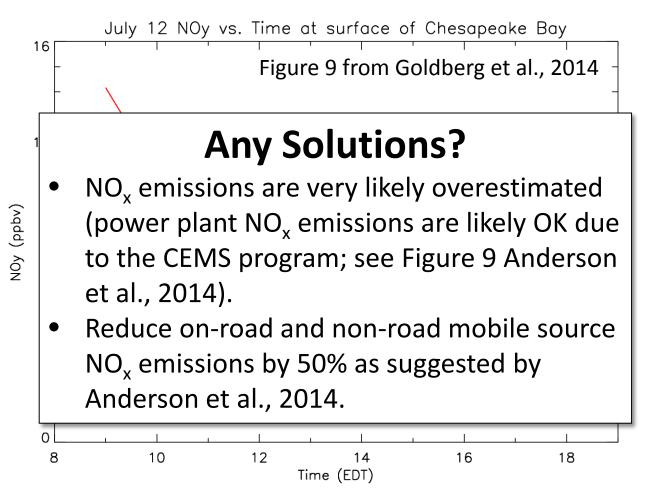
- CAMx v6.10 and CMAQ v5.02, in their baseline model set-up (CB05 and Version 2 emissions), over predict NO_v mixing ratios.
 - CAMx is 86% too high.
 - CMAQ is 73% too high.



NO_v is over predicted, especially in the lower-most layers of the PBL.

*There are <u>several</u> other days during DISCOVER-AQ when there is a similar over prediction.

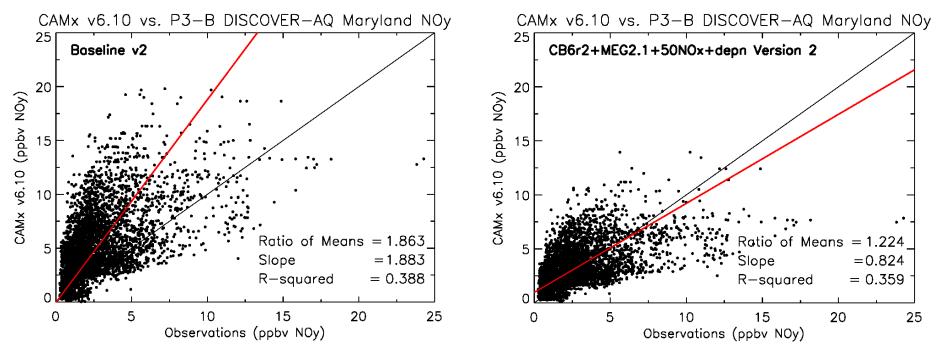
Problem:



NO_y is over predicted by roughly a factor of two, <u>Goldberg et al.</u>, <u>2014</u>

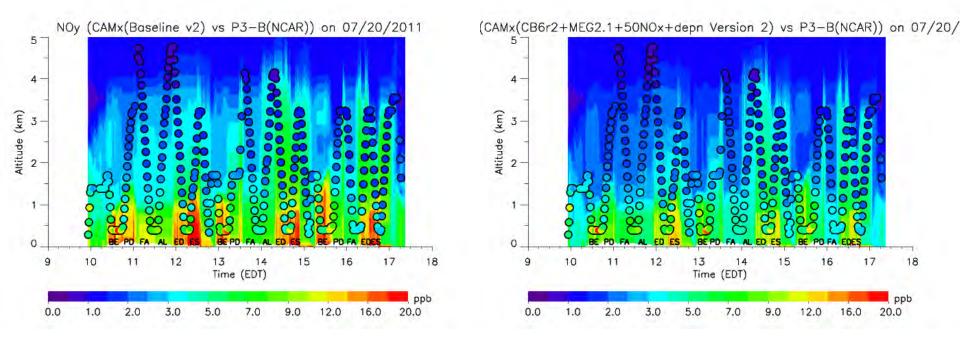
*There are *several* other days during DISCOVER-AQ when there is a similar over prediction.

Better agreement with NO_y observations taken during DISCOVER-AQ



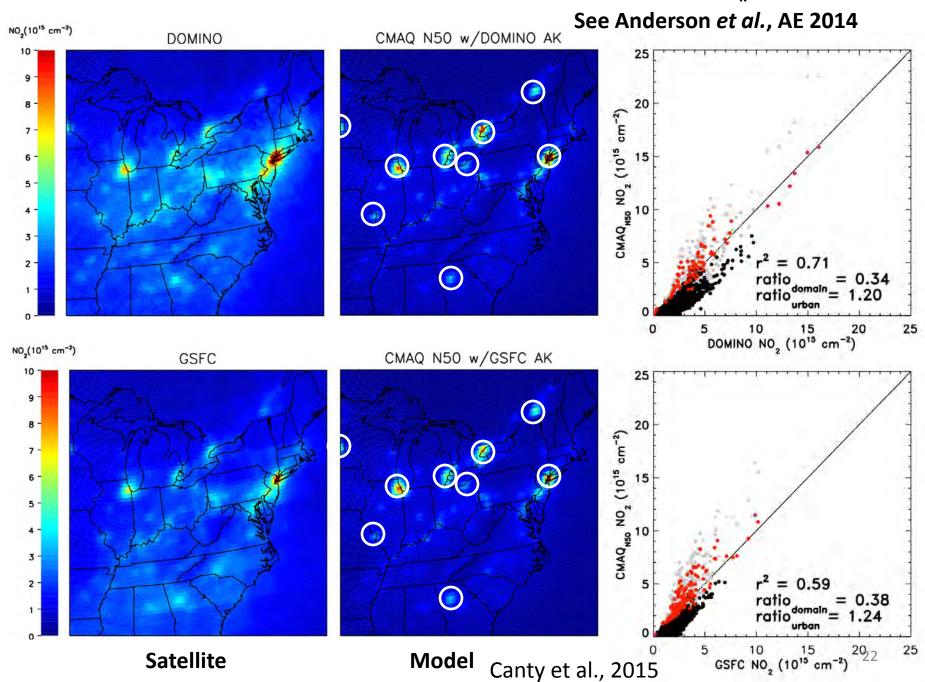
- Overestimate has been reduced, but still a 22% overestimation (NO_y observations > 15 ppbv are likely small-scale plumes which cannot be resolved by a 12 km simulation; thus slope is skewed low); Goldberg et al., in preparation.
- CAMx simulation also includes changes to biogenics and CB6r2 (right panel)

Better agreement with NO_y observations taken during DISCOVER-AQ



 Better prediction of NO_y especially in the PBL, when reducing onroad and non-road mobile emissions by 50%.

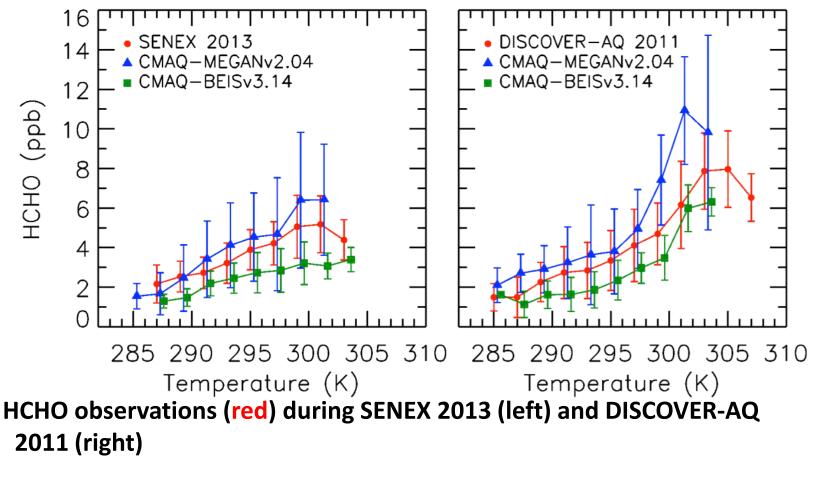
OMI vs CMAQ w/ modified chemistry + 50% \downarrow mobile NO_x



Problem:

- Formaldehyde and Isoprene are under predicted by CAMx & CMAQ when biogenic emissions are initialized using BEIS
- Overpredicted when emissions initialized by MEGAN

Uncertainties in biogenic emissions: HCHO

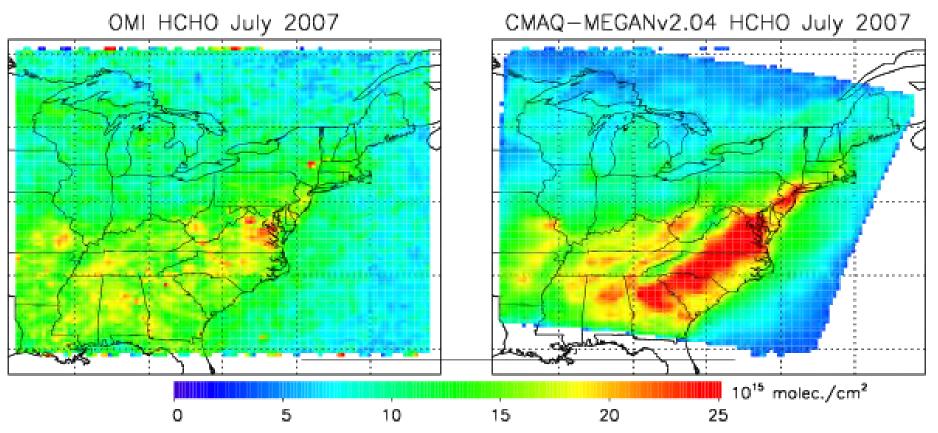


CMAQ using MEGANv2.04 (blue) HIGHER than obs.

CMAQ using BEISv3.14 (green) LOWER than obs.

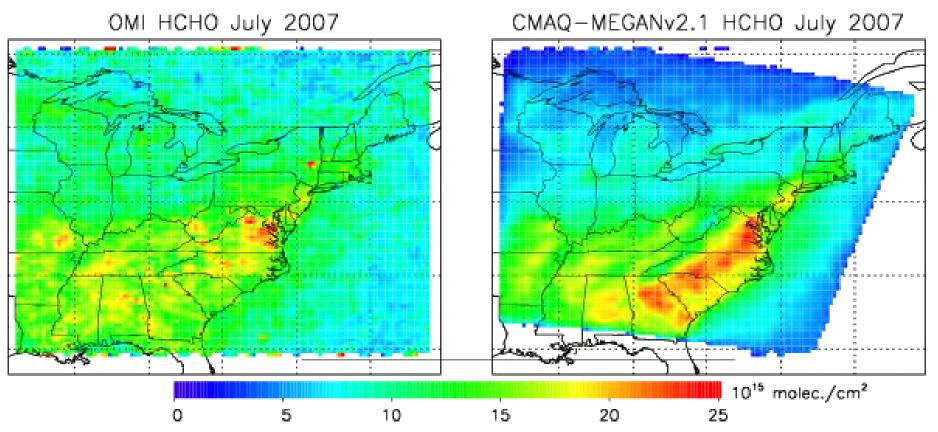
Similar results when comparing to NASA D–AQ isoprene

Satellite HCHO

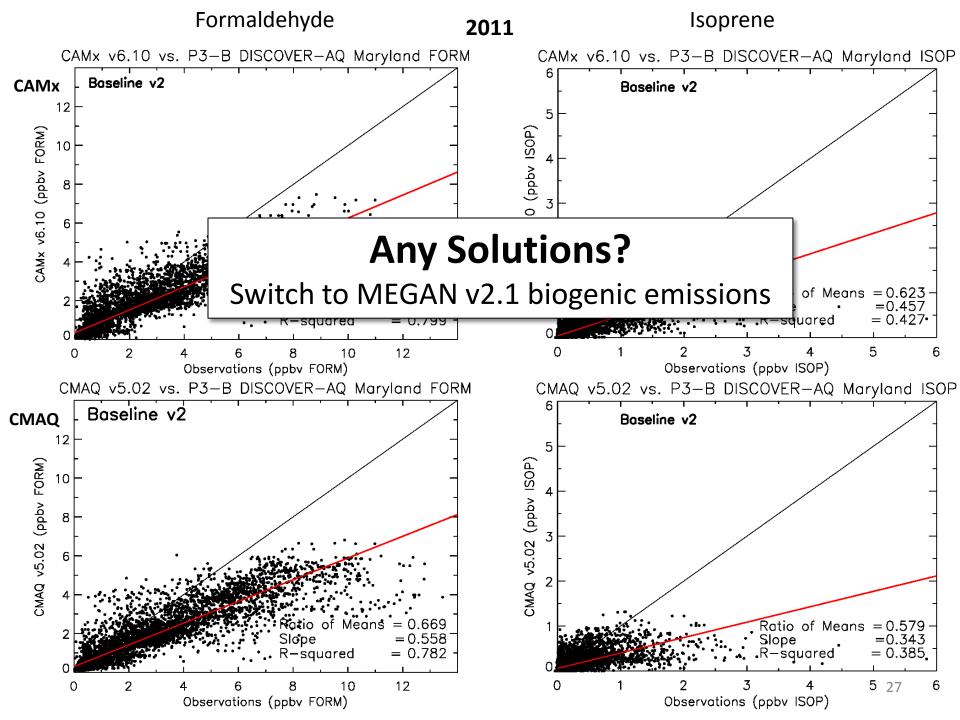


- HCHO product of isoprene oxidation
- OMI HCHO (left) lower than CMAQ HCHO (right)
- New version of MEGAN available (v2.10)

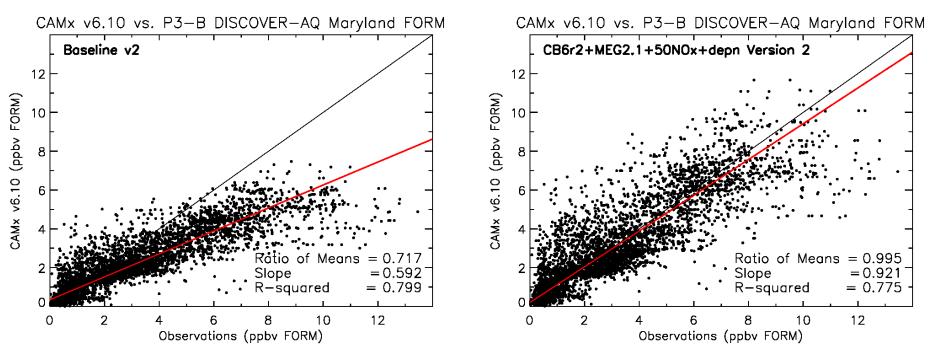
Satellite HCHO



- HCHO product of isoprene oxidation
- OMI HCHO (left) lower than CMAQ HCHO (right)
- New version of MEGAN available (v2.10)
- CMAQ HCHO using update biogenic emissions closer to OMI

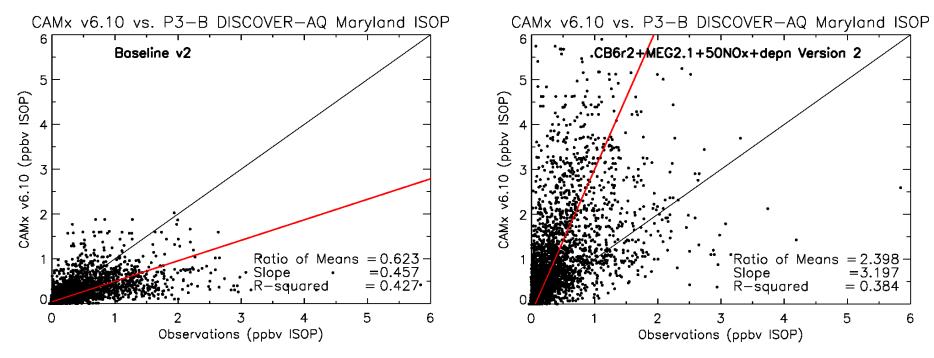


Formaldehyde agrees much better when using MEGAN v2.1 biogenics



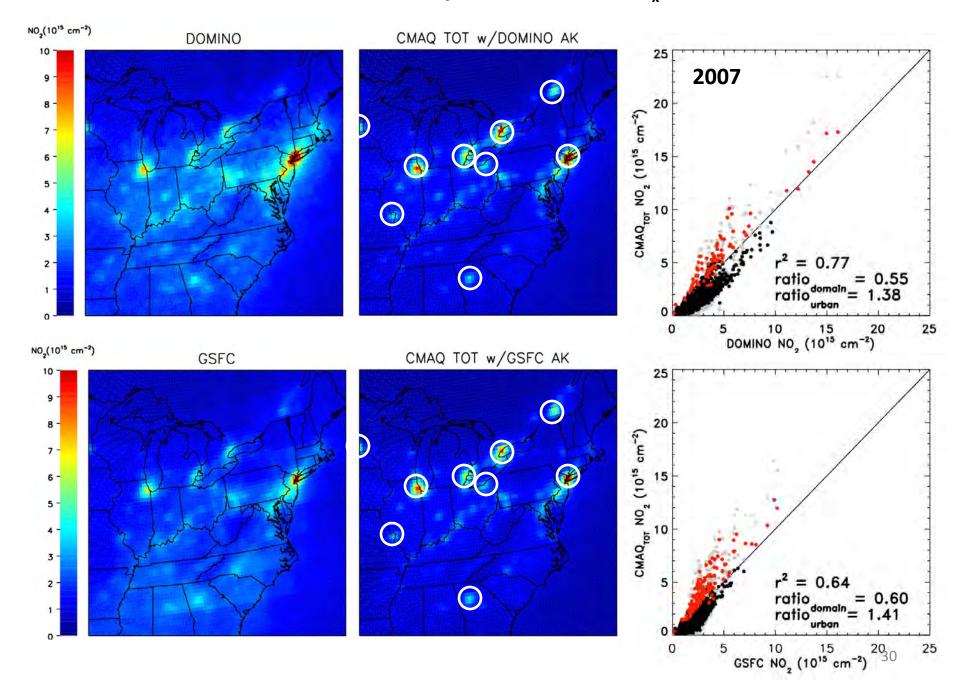
- Underestimate is essentially eliminated; Goldberg et al., in preparation
- Accurate prediction of formaldehyde is essential because it is a major source of the HO₂ radical. Affects OPE (Hembeck et al, in prep.)
- CAMx simulation also includes reductions in mobile NOx and CB6r2 (right panel)

Equally poor prediction for isoprene when using MEGAN v2.1 biogenics



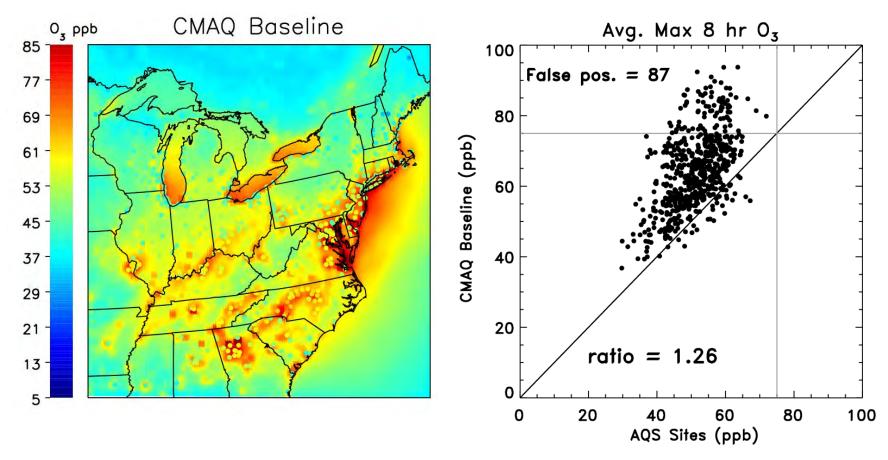
- CAMx with BEIS v3.6 biogenics shows a large underestimation, while CAMx with MEGAN v2.1 biogenics shows a large overestimation.
- CAMx simulation also includes reductions in mobile NOx and CB6r2 (right panel)

OMI vs CMAQ w/ modified chemistry + 50% \downarrow mobile NO_x + new bio emissions



Effect of model changes on O₃

Average Maximum 8 hr O₃, July & August : Model and Observations

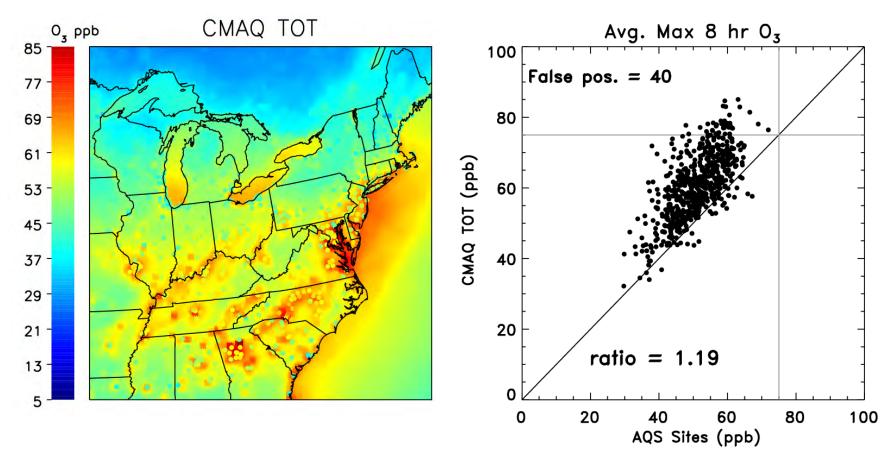


"Standard" model much higher than observations for July+August 2007

Colored points are AQS sites

Canty et al., 2015

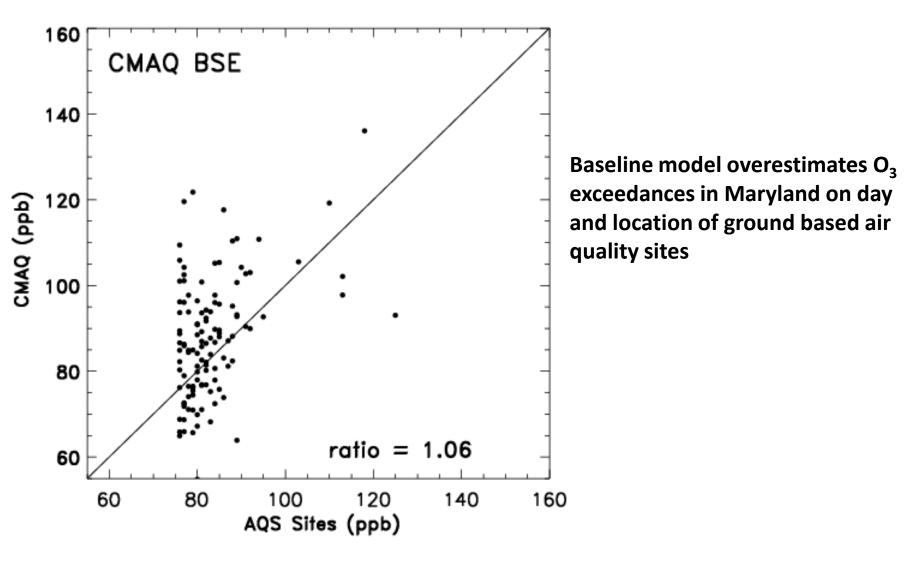
Average Maximum 8 hr O₃, July & August : Model and Observations



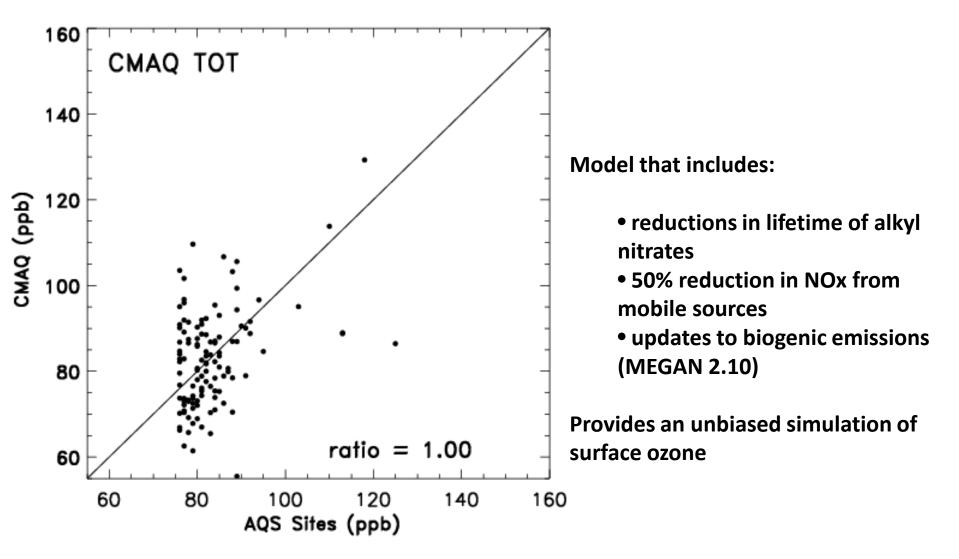
"Standard" model much higher than observations for July+August 2007

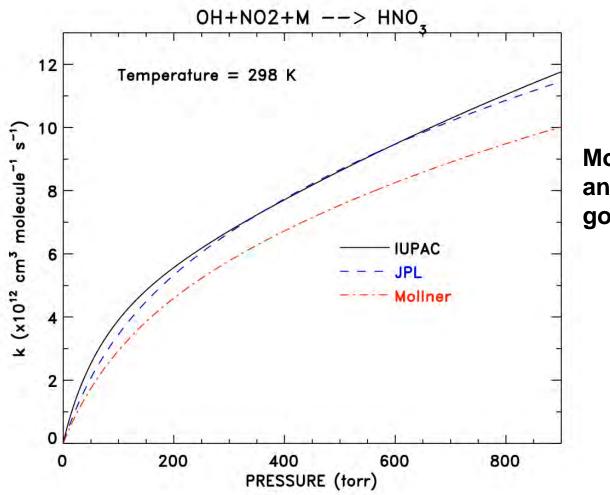
Model that includes modified chemistry, $50\% \checkmark$ in mobile NOx, and updated biogenic emissions (MEGAN 2.10) in better agreement with obs.

July/August 2007 Maryland Ozone Exceedances



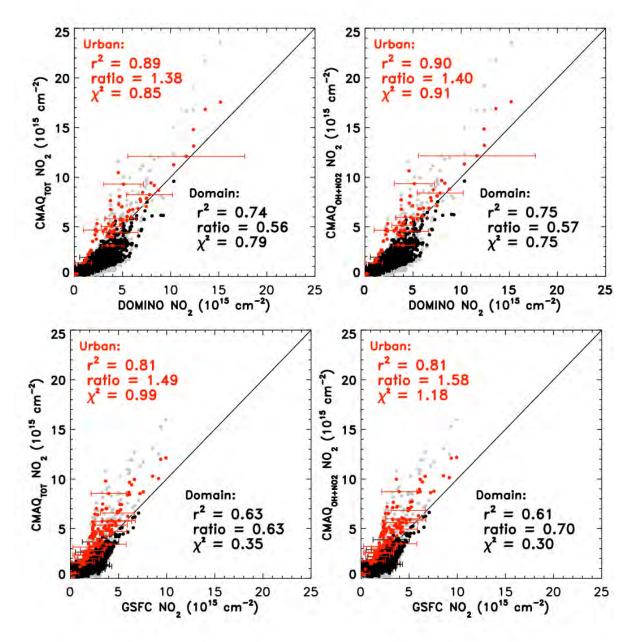
July/August 2007 Maryland Ozone Exceedances





Mollner et al., 2010 reported an update to the kinetics that govern the reaction rate of

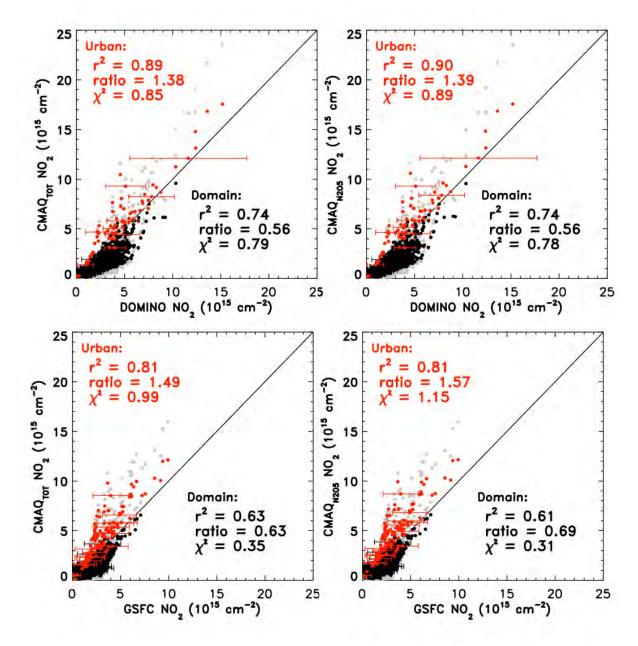
Canty et al., 2015



Left Panels: Trop. NO₂ from CMAQ output that considers update to NTR chemistry, 50% reduction in on-road mobile NO_x, and reduction in isoprene from biogenic sources.

Right Panels: CMAQ output also considers update to OH+NO₂.

Canty et al., 2015



Left Panels: Trop. NO₂ from CMAQ output that considers update to NTR chemistry, 50% reduction in on-road mobile NO_x, and reduction in isoprene from biogenic sources.

Right Panels: CMAQ output also does not include heterogenous loss of N_2O_5 .

Canty et al., 2015

CMAQ SIP Scenarios: 2011 Emissions v1, Series 3

- 3A Using IPM results, reduce SCR/SNCR units to there lowest rates as seen in CAMD data (2005 -2012).
- 3B— Using IPM results, increase SCR/SNCR units to there worst rates as seen in CAMD data (2005 2012).
- 3C— Increase NOX at coal fired SCR/SNCR units to emissions as seen in 2011 CAMD data.
- 3D— Uncontrolled units modeled as if they are controlled by an SCR.

UMD ran tests cases of these scenarios with 2x Biogenic Emissions Following slides show CMAQ output

CMAQ Model Scenarios (2011) Vers 1 Emissions

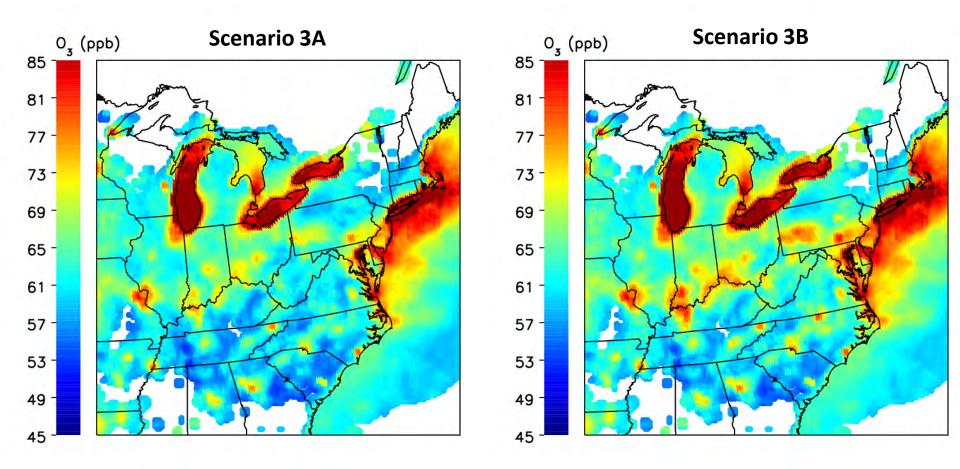
"Best Case"- SCR's Running at Lowest Rate "Worse Case"- SCR's Running at Worst Rate

Scenario 3A O₃ (ppb) 85 81 -77 -73 -69 -65 -61 -57 -53 -49 45

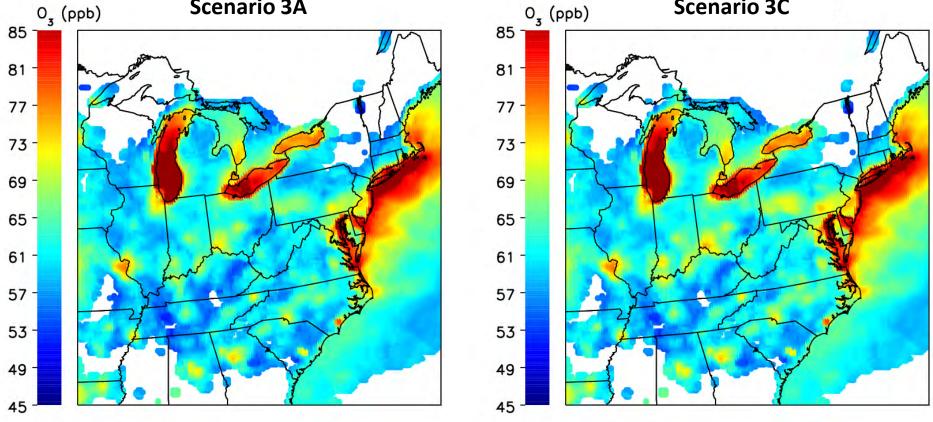
Scenario 3B

CMAQ Model Scenarios (2011) 2 x Biogenics

"Best Case"- SCR's Running at Lowest Rate "Worse Case"- SCR's Running at Worst Rate



CMAQ Model Scenarios (2011) Vers 1 Emissions "Best Case"- SCR's Running at Lowest Rate (nob) Scenario 3A O (nob) Scenario 3C

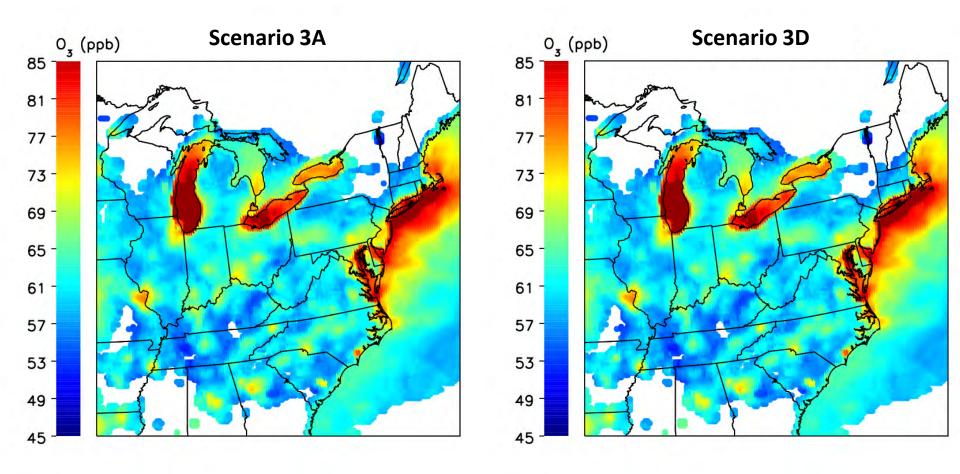


CMAQ Model Scenarios (2011) 2 x Biogenics "Best Case"- SCR's Running at Lowest Rate "Real Case"- SCR's Running at 2011 Rate

Scenario 3A Scenario 3C O₃ (ppb) O₃ (ppb) 85 85 81 -81 -77 -77 -73 -73 -69 -69 -65 -65 -61 -61 -57 -57 -53 -53 -49 49 -45 45

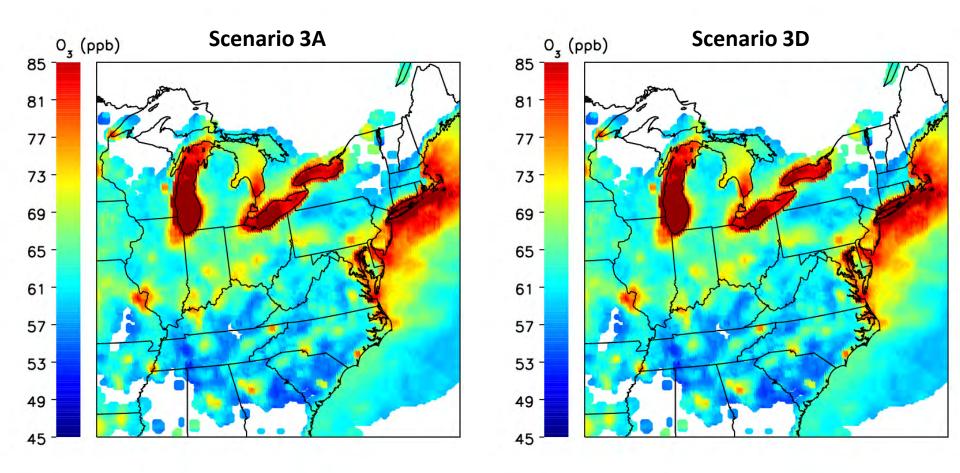
CMAQ Model Scenarios (2011) Vers 1 Emissions

"Best Case"- SCR's Running at Lowest Rate "Better Case"- SCR's on uncontrolled Units



CMAQ Model Scenarios (2011) 2 x Biogenics

"Best Case"- SCR's Running at Lowest Rate "Better Case"- SCR's on uncontrolled Units



All model results for July only (2011 Platform)

County	Site	DV 2011	3A (ATT-1)	3B	3C	3D
Anne Arundel	Davidsonville	83	65.7	67.1	66.6	65.4
Baltimore	Padonia	79	64.0	66.1	65.4	63.6
Baltimore	Essex	80.7	60.8	62.2	61.7	60.6
Calvert	Calvert	79.7	64.8	67.2	66.0	64.7
Carroll	South Carroll	76.3	63.6	66.5	65.6	63.1
Cecil	Fair Hill	83	66.3	68.8	67.9	66.0
Calvert	S.Maryland	79	64.2	66.6	65.5	63.9
Cambridge	Blackwater	75	61.8	63.4	62.7	61.5
Frederick	Frederick Airport	76.3	63.6	66.7	65.7	63.1
Garrett	Piney Run	72	57.5	59.9	59.0	55.2
Harford	Edgewood	90	68.9	70.7	70.0	68.6
Harford	Aldino	79.3	61.5	63.5	62.8	61.2
Kent	Millington	78.7	61.7	63.9	63.2	61.4
Montgomery	Rockville	75.7	61.5	63.1	62.5	61.2
PG	HU-Beltsville	79	63.0	64.4	63.9	62.6
PG	PG Equest.	82.3	65.4	67.1	66.4	65.1
PG	Beltsville	80	63.2	64.7	64.2	63.0
Washington	Hagerstown	72.7	60.4	63.3	62.4	59.8
Baltimore City	Furley	73.7	55.5	56.8	56.4	55.3

All model results for July only (2011 Platform) 2xBiogenic

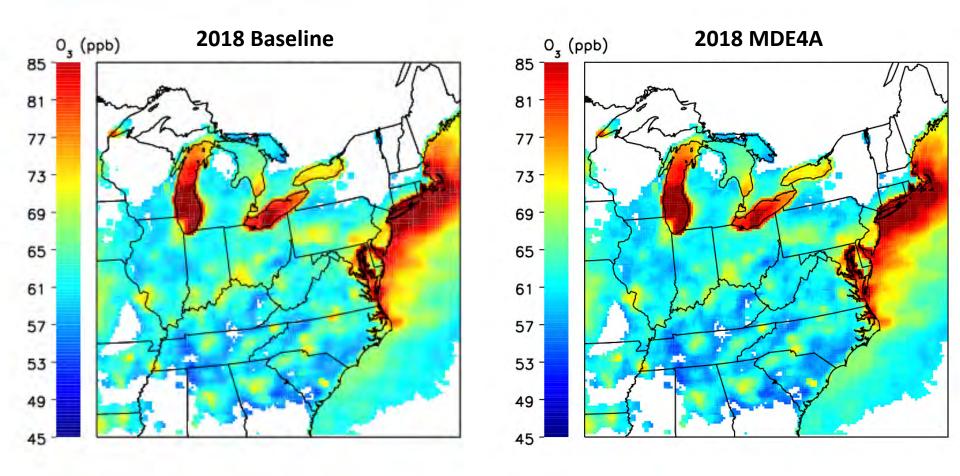
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Baltimore	Padonia	79	66.7	69.2	68.4	66.3
Baltimore	Essex	80.7	68.3	70.1	69.5	68.0
Calvert	Calvert	79.7	67.6	70.4	68.9	67.4
Carroll	South Carroll	76.3	65.2	68.7	67.6	64.7
Cecil	Fair Hill	83	68.4	71.4	70.4	68.0
Calvert	S.Maryland	79	65.7	68.7	67.2	65.4
Cambridge	Blackwater	75	64.2	66.0	65.2	63.9
Frederick	Frederick Airport	76.3	65.3	68.8	67.7	64.7
Garrett	Piney Run	72	58.4	61.5	60.3	55.5
Harford	Edgewood	90	74.8	77.0	76.2	74.4
Harford	Aldino	79.3	64.8	67.2	66.3	64.5
Kent	Millington	78.7	64.3	67.0	66.1	63.9
Montgomery	Rockville	75.7	63.6	65.5	64.8	63.2
PG	HU-Beltsville	79	64.9	66.7	66.1	64.6
PG	PG Equest.	82.3	67.5	69.6	68.8	67.2
PG	Beltsville	80	65.4	67.2	66.6	65.1
Washington	Hagerstown	72.7	61.8	65.0	64.0	61.0
Baltimore City	Furley	73.7	62.5	64.2	63.7	62.3

CMAQ SIP Scenarios: 2011 Emissions v2, Series 4

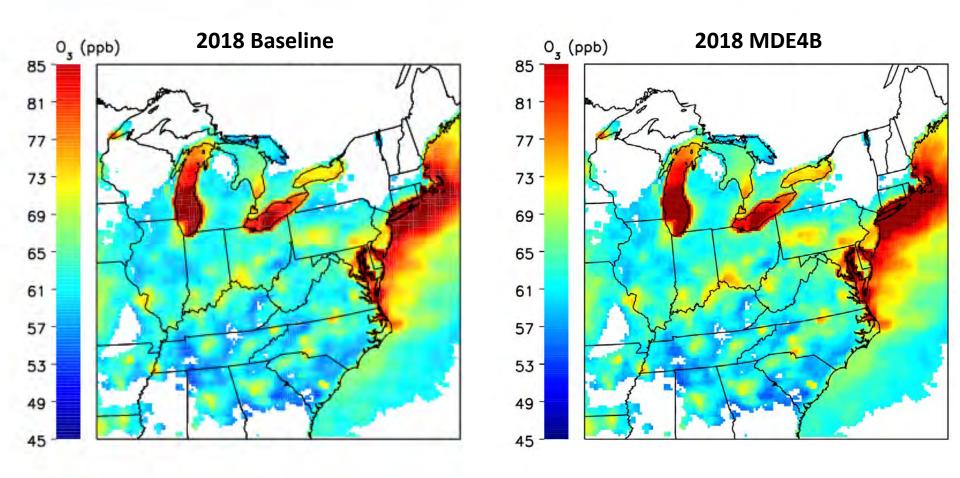
- 2011 Baseline
- MDE4A 2018 w/Tier 3, On the books/On the way reductions, Optimized EGU's
- MDE4B EGUs at worst rates
- MDE4C EGUs at real rates seen in 2011 or 2012
- MDE4D SCR reductions at remaining post-2017/2018 uncontrolled EGUs

Following slides show CMAQ output

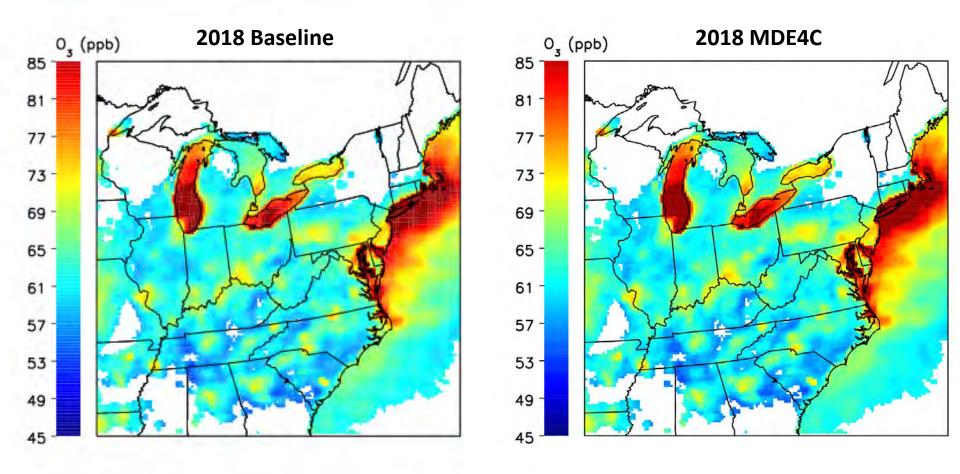
2018 w/Tier 3 OTB/OTW, Opt. EGU



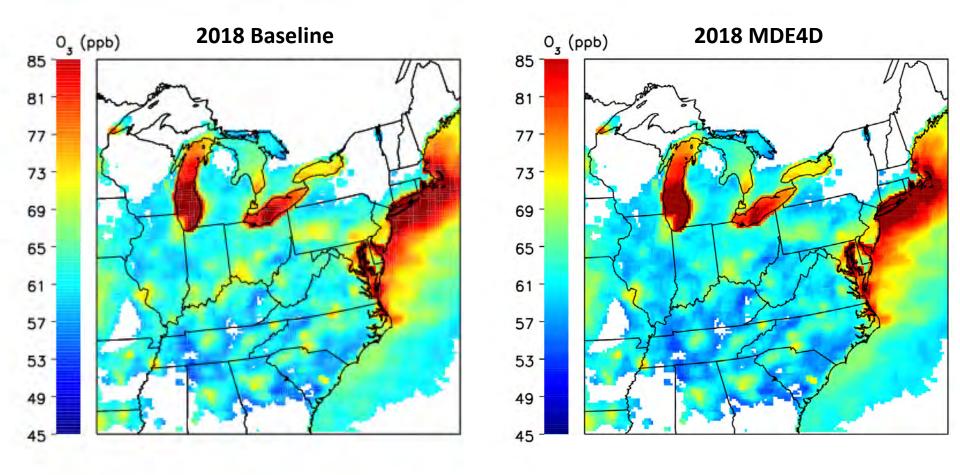
EGUs at worst rates



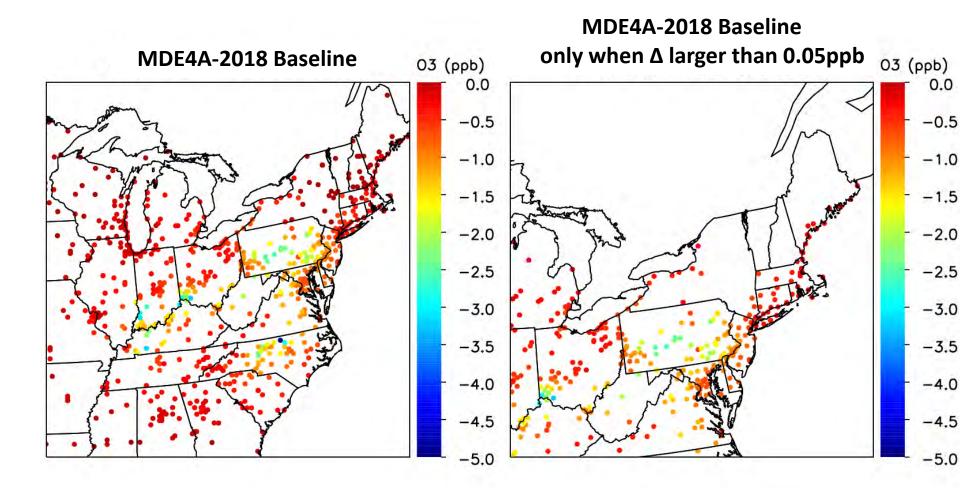
EGUs at real rates seen in 2011 or 2012



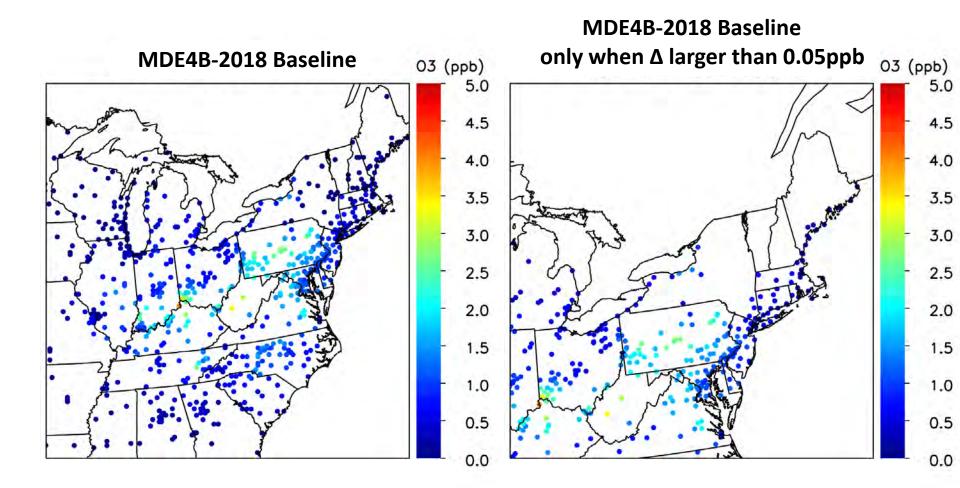
SCR reductions at remaining uncontrolled EGUs



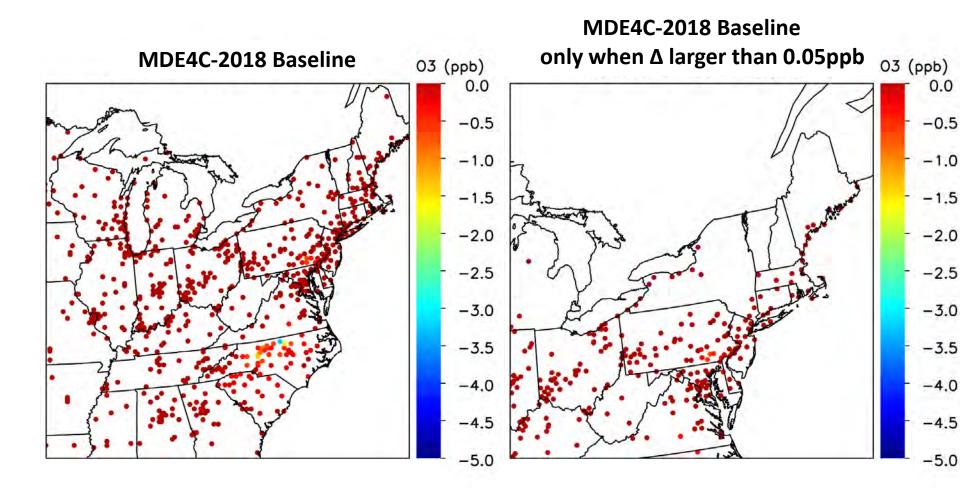
2018 w/Tier 3 OTB/OTW, Opt. EGU

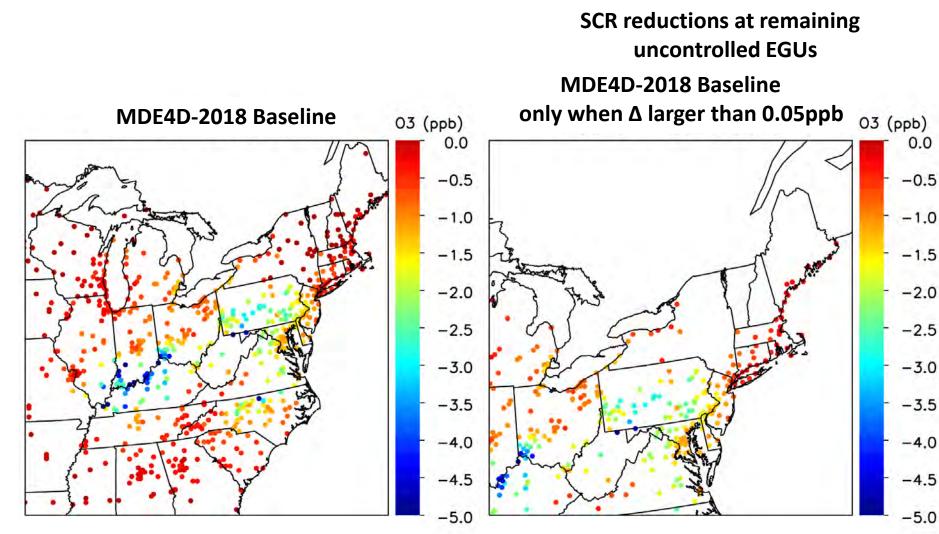


EGUs at worst rates



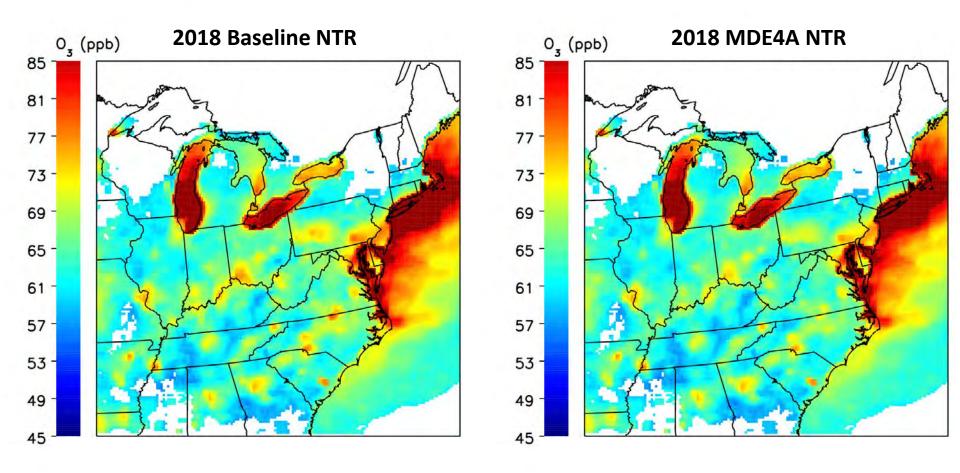
EGUs at real rates seen in 2011 or 2012





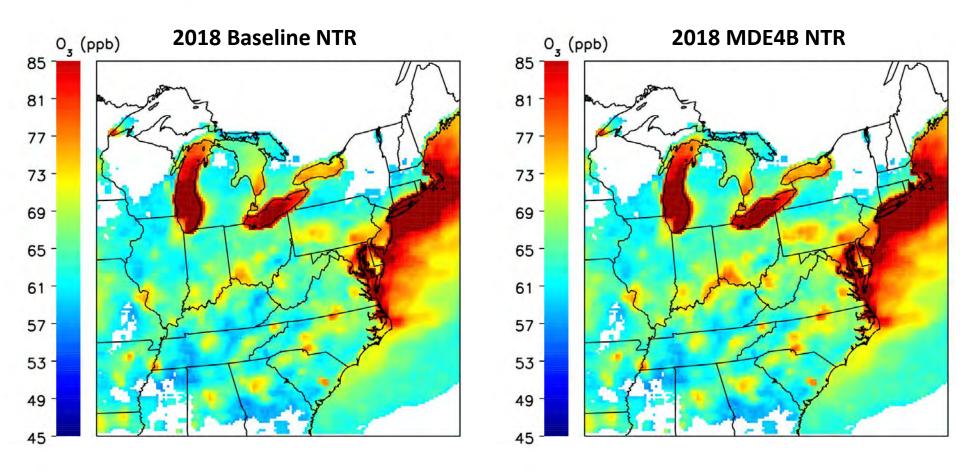
2018 w/Tier 3 OTB/OTW, Opt. EGU

NTR = Decrease in lifetime of Alkyl Nitrates



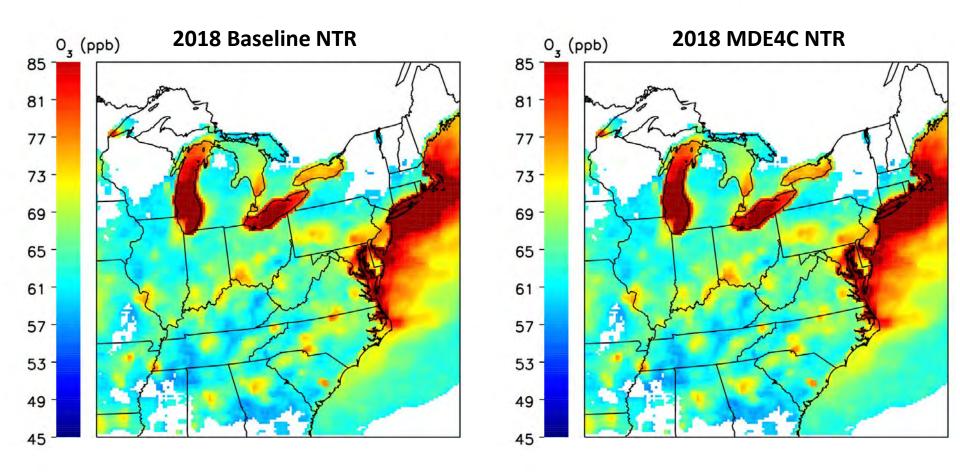
EGUs at worst rates

NTR = Decrease in lifetime of Alkyl Nitrates



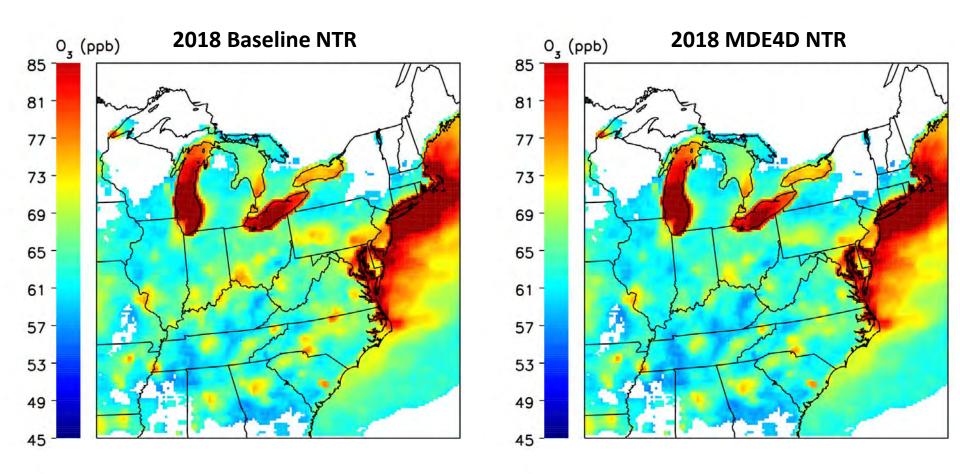
EGUs at real rates seen in 2011 or 2012

NTR = Decrease in lifetime of Alkyl Nitrates



SCR reductions at remaining

uncontrolled EGUs NTR = Decrease in lifetime of Alkyl Nitrates



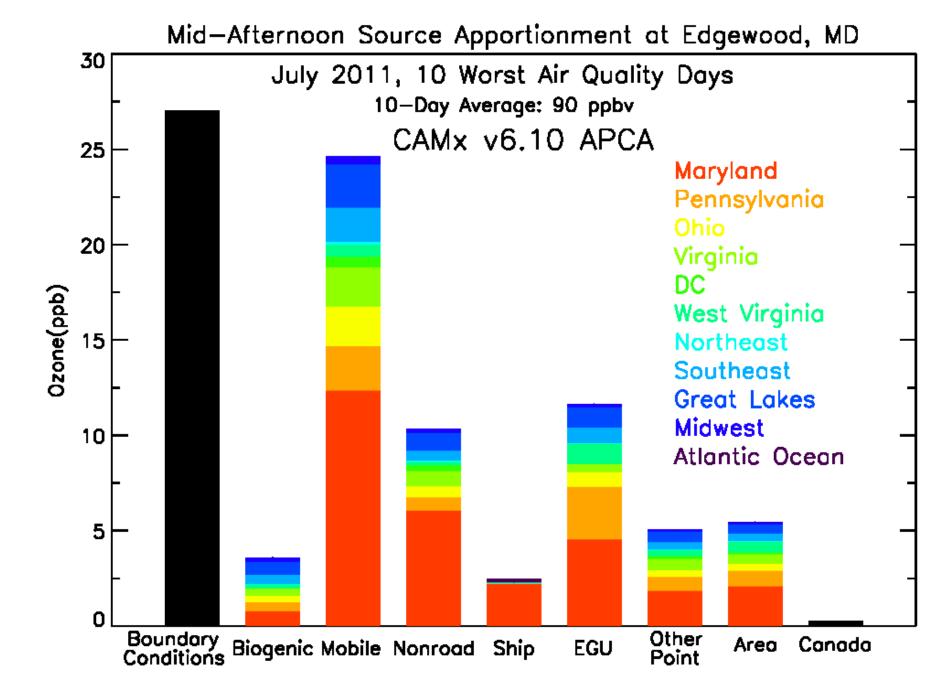
Design Values for Standard Model and Model w/modified NTR chemistry

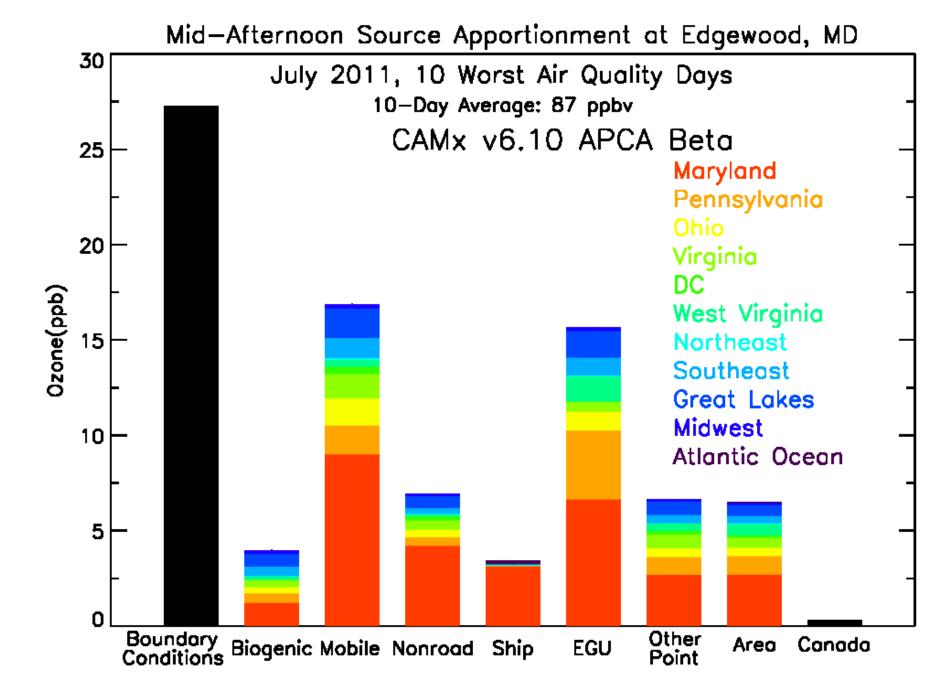
County	Site	DV 2011	DV 2018	2018 NTR	MDE4A	MDE4A NTR	MDE4B	MDE4B NTR	MDE4 C	MDE4C NTR	MDE4 D	MDE4 D NTR
Anne Arundel	Davidsonville	83	72.3	73.4	71.7	72.8	73.2	74.2	72.4	73.5	71.3	72.3
Baltimore	Padonia	79	70.8	71.5	69.9	70.5	71.9	72.6	70.9	71.6	69.3	67.0
Baltimore	Essex	80.7	74.3	74.5	73.8	74.0	75.0	75.2	74.4	74.6	73.5	73.7
Calvert	Calvert	79.7	72.3	73.0	71.6	72.3	73.1	73.8	72.3	73.0	71.3	72.1
Carroll	South Carroll	76.3	68.3	68.8	67.1	67.7	69.9	70.4	68.5	69.0	66.4	67.0
Cecil	Fair Hill	83	74.6	75.3	73.5	74.2	75.7	76.1	74.5	75.1	73.0	73.7
Calvert	S.Maryland	79	70.4	71.3	69.5	70.5	71.6	72.4	70.6	71.4	69.0	70.0
Cambridge	Blackwater	75	67.3	68.2	66.7	67.7	68.2	69.`	67.5	68.3	66.3	67.3
Frederick	Frederick Airport	76.3	68.1	69.0	66.7	67.6	69.9	70.6	68.3	69.2	65.9	66.9
Garrett	Piney Run	72	61.7	62.5	60.2	61.1	63.6	64.3	61.8	62.6	57.3	58.3
Harford	Edgewood	90	82.1	82.7	81.5	82.1	82.9	83.5	82.2	82.8	81.2	81.8
Harford	Aldino	79.3	70.7	71.5	69.9	70.8	71.6	72.4	70.7	71.6	69.5	70.4
Kent	Millington	78.7	70.5	71.3	69.6	70.5	71.5	72.2	70.5	71.2	69.1	70.0
Montgomery	Rockville	75.7	66.5	67.2	65.7	66.4	67.6	68.2	66.7	67.3	65.2	65.8
PG	HU-Beltsville	79	68.4	69.3	67.7	68.7	69.4	70.2	68.6	69.4	67.3	68.2
PG	PG Equest.	82.3	71.8	72.9	71.1	72.1	72.8	73.8	72.0	73.0	70.7	71.7
PG	Beltsville	80	69.6	70.3	69.0	69.7	70.4	71.1	69.7	70.34	68.5	69.3
Washington	Hagerstown	72.7	64.3	65.1	63.2	64.0	65.8	66.5	64.5	65.3	62.3	63.2
Baltimore City	Furley	73.7	67.5	67.8	67.0	67.3	68.2	68.4	67.7	67.9	66.8	67.1

CAMx SIP Scenarios: 2011 Emissions

- We are showing two figures for surface ozone at Edgewood, Maryland during the 10 worst air quality days during July 2011; Goldberg et al., in preparation.
 - 1. Baseline simulation (CB05, BEISv3.6, no changes to NTR)
 - Updated "Beta" simulation (on-road, off-road, AND non-road NO_x emissions reduced by 50%, in addition to changing to MEGAN v2.1 biogenics, CB6r2, and increased NTR deposition)

Following slides show CAMx output





Total ozone concentrations attributed to each source sector

Ozone (ppb) attributed to each Source Sector

	Biogenic	On-road Mobile	Non-road Mobile	Ships	EGUs	Other Point	Area
Baseline Simulation	3.6	24.6	10.3	2.5	11.6	5.1	5.6
Beta Simulation	4.0	16.9	6.9	3.4	15.7	6.7	6.6
Percentage change	10.9%	-31.4%	-33.0%	39.6%	34.6%	31.0%	18.7%

CAMx Design Values for 2018 using the Baseline and the Updated Version of the model

Maryland Monitoring Location	County	Observed 2011 DV (ppb)	CAMx 2018: CB05 and BEIS v3.6, Version 1 Emissions, July Only (ppb)	CAMx 2018: CB6r2, MEGANv2.1, and 50% mobile NOx, Version 1 Emissions, July Only (ppb)
Davidsonville	Anne Arundel	83.0	71.1	70.6
Padonia	Baltimore	79.0	70.6	70.1
Essex	Baltimore	80.7	71.6	70.6
Calvert	Calvert	79.7	69.5	69.2
South Carroll	Carroll	76.3	67.7	68.4
Fair Hill	Cecil	83.0	71.9	72.0
Southern Maryland	Charles	79.0	68.3	68.5
Frederick Airport	Frederick	76.3	68.0	68.4
Piney Run	Garrett	72.0	62.2	61.2
Edgewood	Harford	90.0	79.1	77.7
Aldino	Harford	79.3	68.6	67.9
Millington	Kent	78.7	67.8	67.9
Rockville	Montgomery	76.3	67.8	65.6
HU-Beltsville	Prince George's	79.0	68.2	66.7
PG Equestrian Center	Prince George's	82.3	70.4	69.4
Hagerstown	Washington	72.7	64.5	64.4
Furley	Baltimore City	73.7	66.2	64.5

The model with <u>all three changes</u> yields lower Design Values at most locations throughout the region. It is more responsive to NOx emission reductions. ⁶⁶