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Supporting Information for

**CAMx Ozone Source Attribution in the Eastern United States using Guidance from
Observations during DISCOVER-AQ Maryland**

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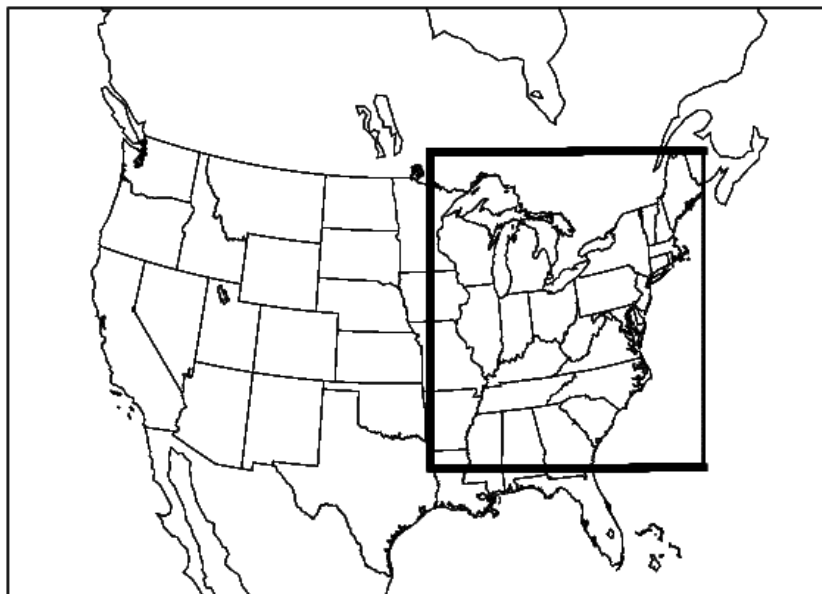


Figure S1. CAMx v6.10 model domain used in our study. Values of ozone and other chemical compounds at the boundary (rectangle) are obtained from GEOS-Chem v8-03-02. Within the modeling domain, calculations are conducted at 12-km horizontal resolution.

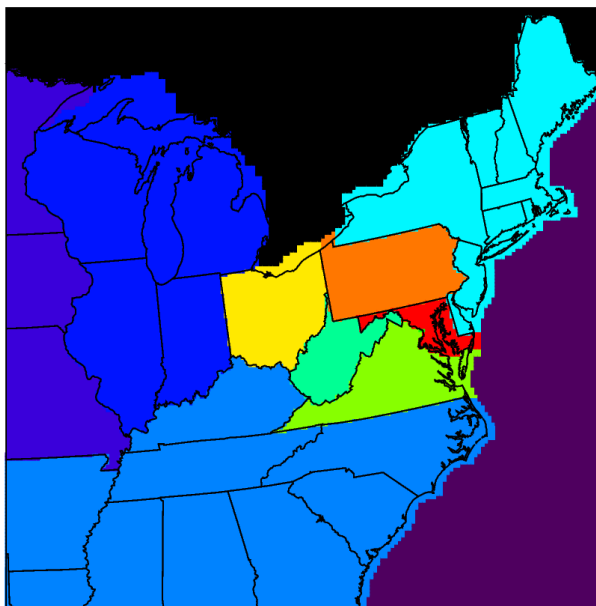


Figure S2. The twelve source regions used for tagging the attribution of ozone using the CAMx APCA tool. The various regions are denoted by a uniform color, which match the colors used in Figure 3. The Atlantic Ocean (purple) represents offshore marine emissions and the region of Canada within the modeling domain (black) is also considered. The twelfth source region is ozone attributed to outside the model domain.

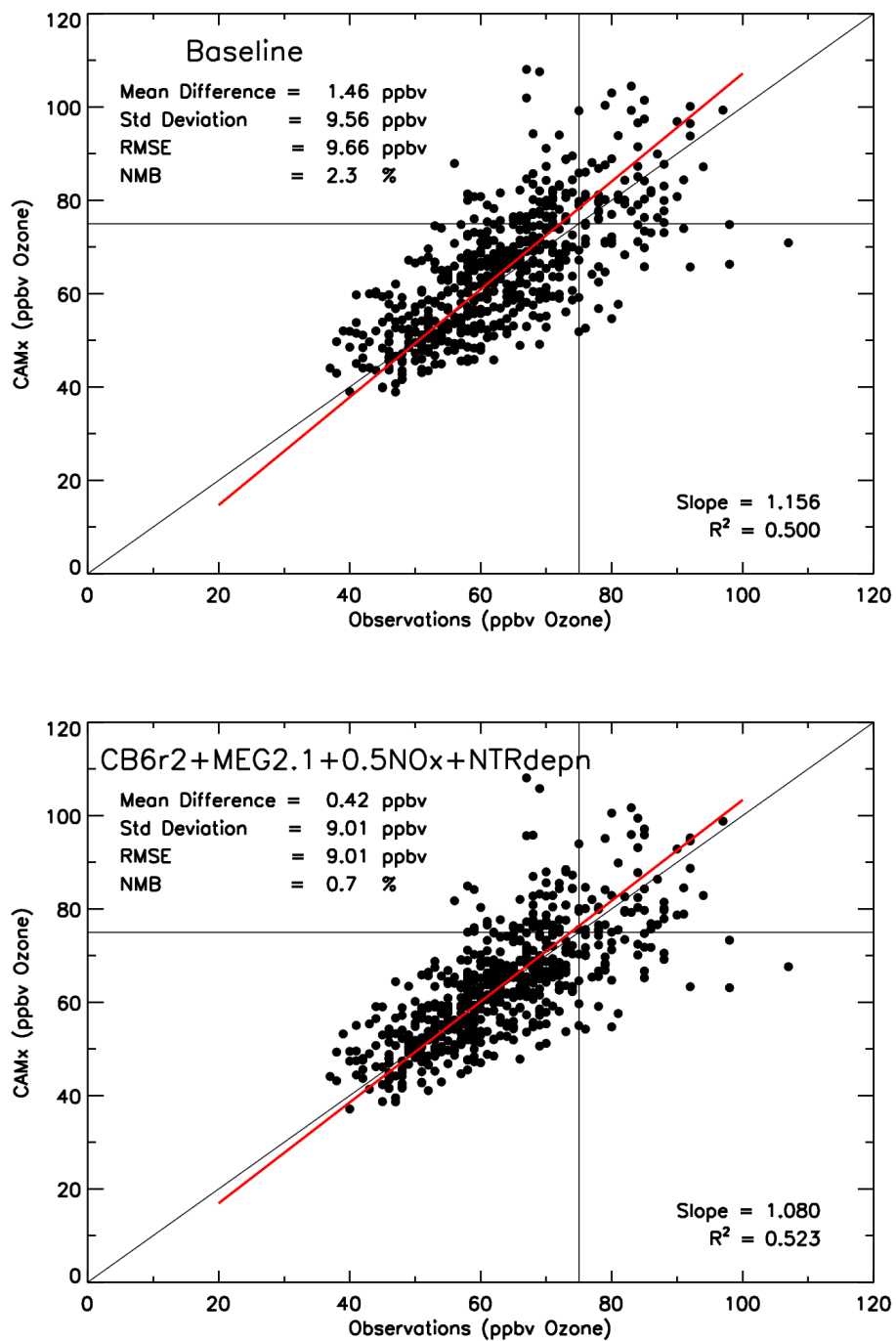


Figure S3. CAMx v6.10 simulated 8-hour maximum ozone mixing ratios compared to observations of the same quantity, matched spatially to observations from nineteen monitoring sites in Maryland, for July 2011. The top panel shows results from the baseline simulation and the bottom panel shows results from the updated “Beta” simulation. Various statistical measures of the two quantities are shown. The 1:1 line (black), linear least-squares best fit (red), as well as the NAAQS for 8-hour surface ozone in effect at the time of the observations (75 ppbv) is also indicated.

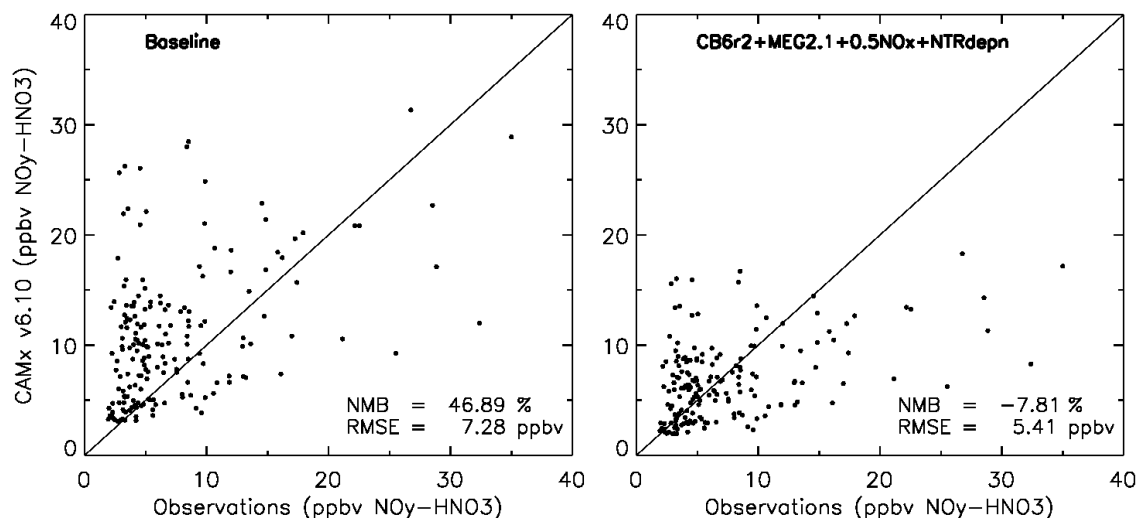


Figure S4. Sixty-minute averaged observations of NO_y – HNO₃ from the Edgewood, MD ground site during DISCOVER-AQ Maryland in July 2011 compared to modeled NO_y – HNO₃ from CAMx v6.10 at the nearest model grid point and closest hourly interval. Left panel shows a comparison between observations and the baseline simulation, while right panel shows a comparison between the same observations and the updated “Beta” simulation. Observations are acquired by NO/NO_y chemiluminescence using an external molybdenum converter heated to 325°C; a converter at this temperature can measure all NO_y species, except a portion of HNO₃ [Fehsenfeld et al., 1987; Williams et al., 1998]. If there was full conversion of HNO₃, it would push the model further from observations. The large spread in observations may be related to the relatively large size of the grid box (12 km).

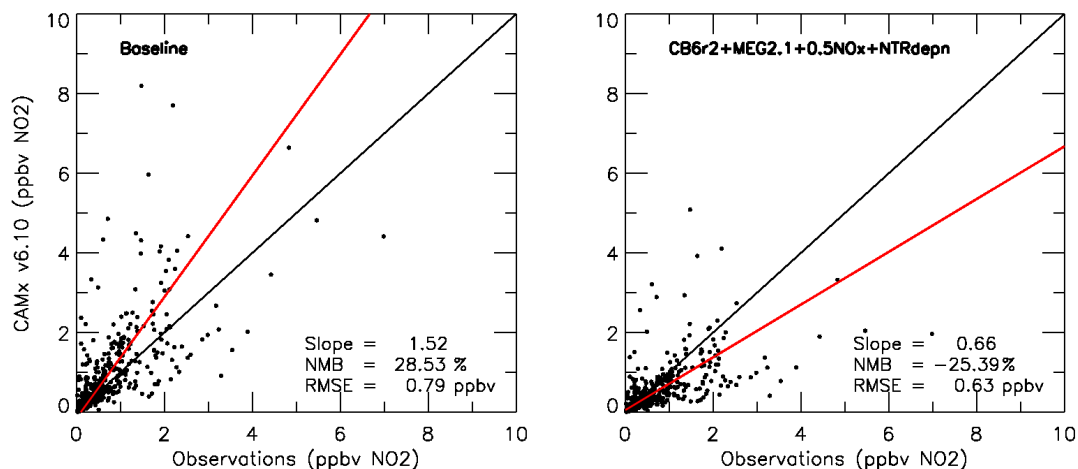


Figure S5. Ten-minute averaged data from the P3-B aircraft during DISCOVER-AQ Maryland in July 2011 compared to model data from CAMx v6.10 at the nearest model grid point and closest hourly interval. The closest hourly model output is matched to each one-minute averaged P3-B observation; both quantities are then averaged over the same ten-minute interval. Left panel shows a comparison between NO_2 observations and the baseline simulation, while right panel shows a comparison between the same observations and the updated “Beta” simulation.

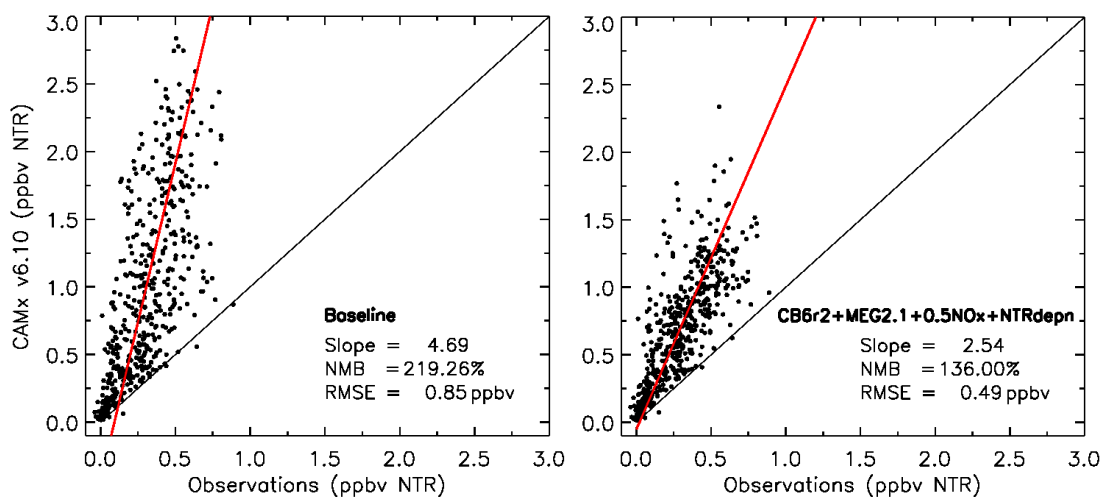


Figure S6. Same as Figure S5, except for alkyl nitrates (NTR).

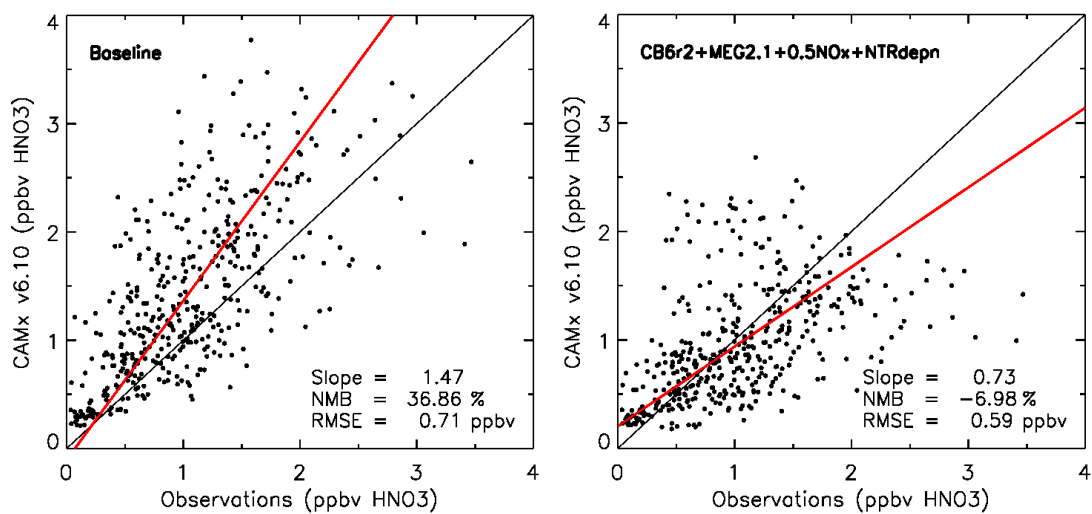


Figure S7. Same as Figure S5, except for nitric acid (HNO_3).

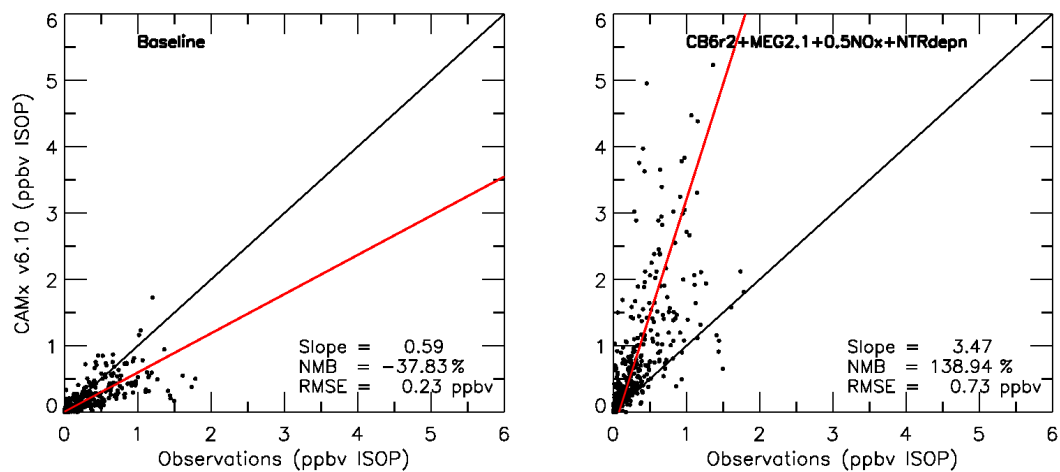


Figure S8. Same as Figure S5, except for isoprene (ISOP).

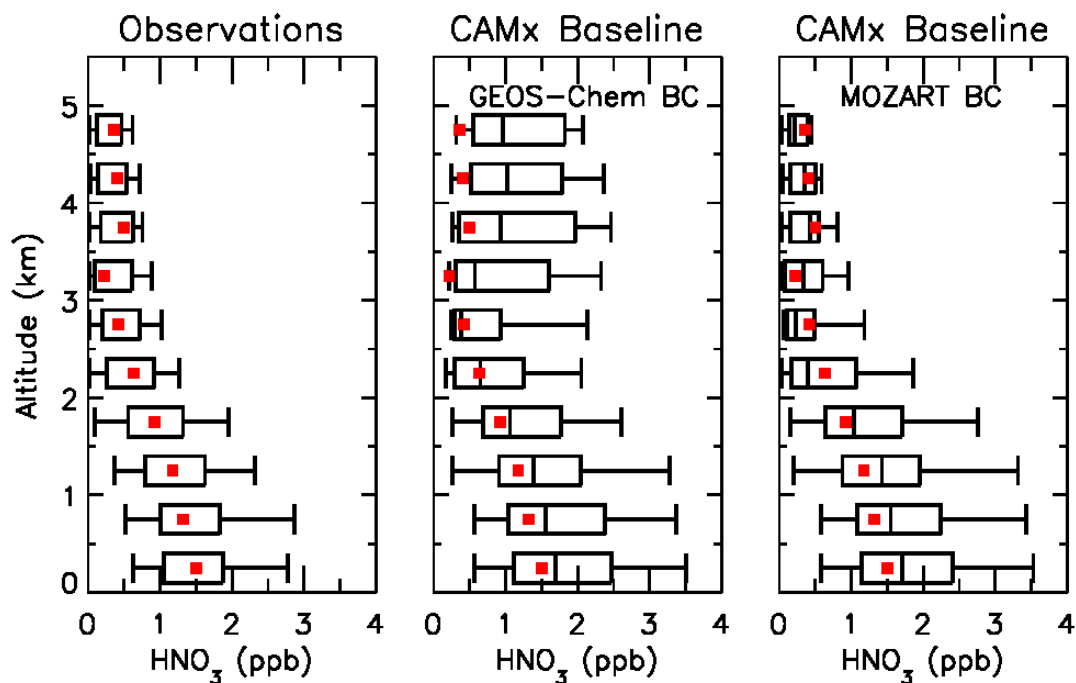


Figure S9. Left panel shows one-minute averaged HNO_3 observations acquired by the P3-B aircraft binned by altitude. Center panel shows the CAMx baseline simulation with GEOS-Chem v8-03-02 boundary conditions binned by altitude, and the right panel shows the CAMx baseline simulation with MOZART v4 boundary conditions binned by altitude. Model output from CAMx v6.10 is matched spatially and temporally. Red squares indicate the median values of the observations, which are shown on all panels to facilitate visual comparison.

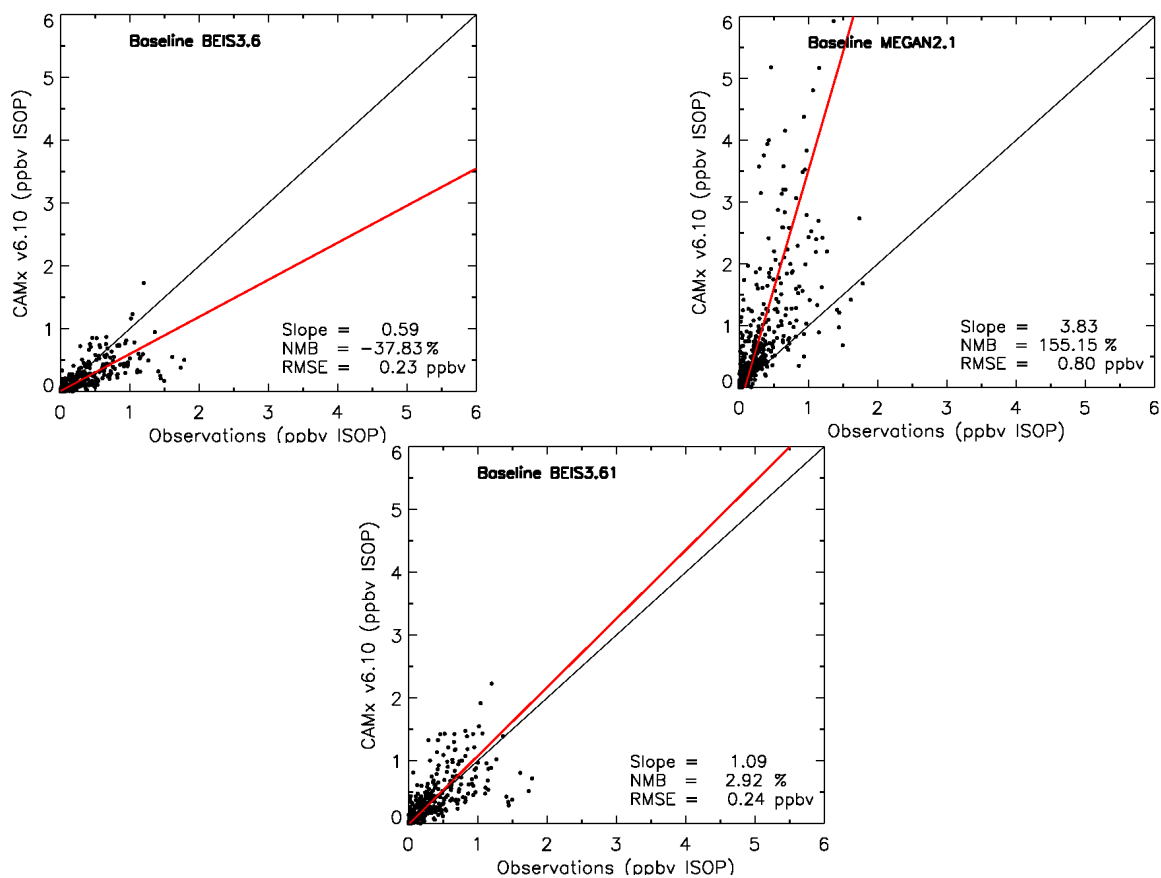


Figure S10. Same as Figure S8, except now showing the baseline simulation (top left panel), a simulation using biogenic emissions from MEGAN v2.1 (top right panel), and a recently completed simulation using biogenic emissions from BEIS v3.61, which became available during final revisions to this manuscript (bottom center panel).

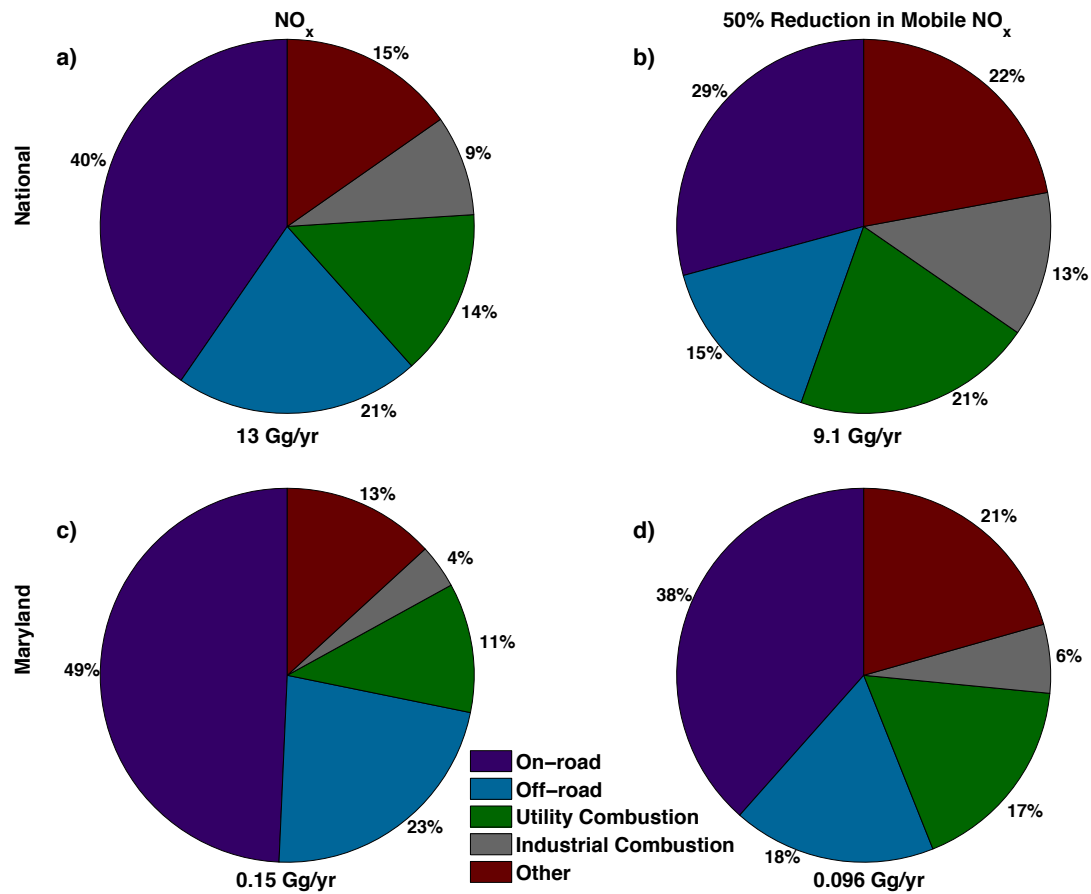


Figure S11. NO_x emissions sorted by sector for (left) the 2011 National Emissions Inventory and (right) a scenario with a 50% reduction in mobile (on-road and off-road) sources. Top row shows percentages for the national inventory. Bottom row shows percentages for the Maryland inventory.

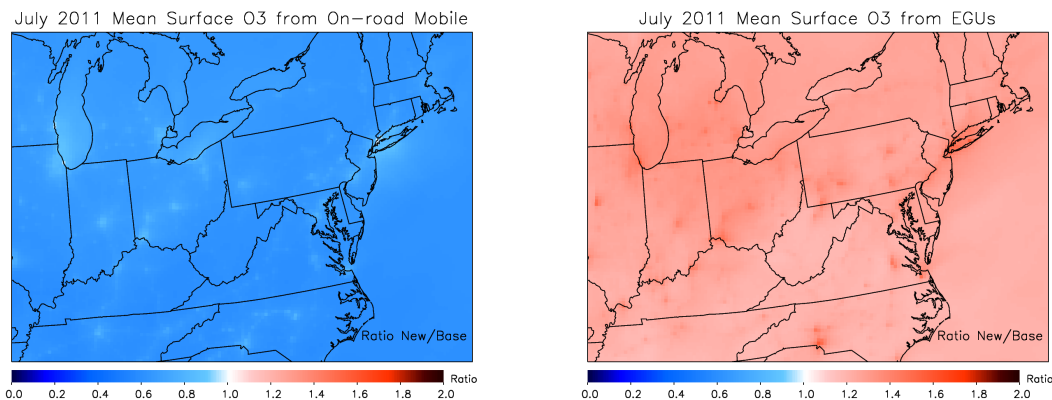


Figure S12. Ratio of ozone source apportionment due to (left) on-road mobile sources and (right) electric generating units (EGUs) for two simulations: the updated “Beta” simulation divided by the same source apportionment value from the baseline version of CAMx. Results are for July 2011 averaged over daytime (8 AM – 8 PM local time) for the entire month.

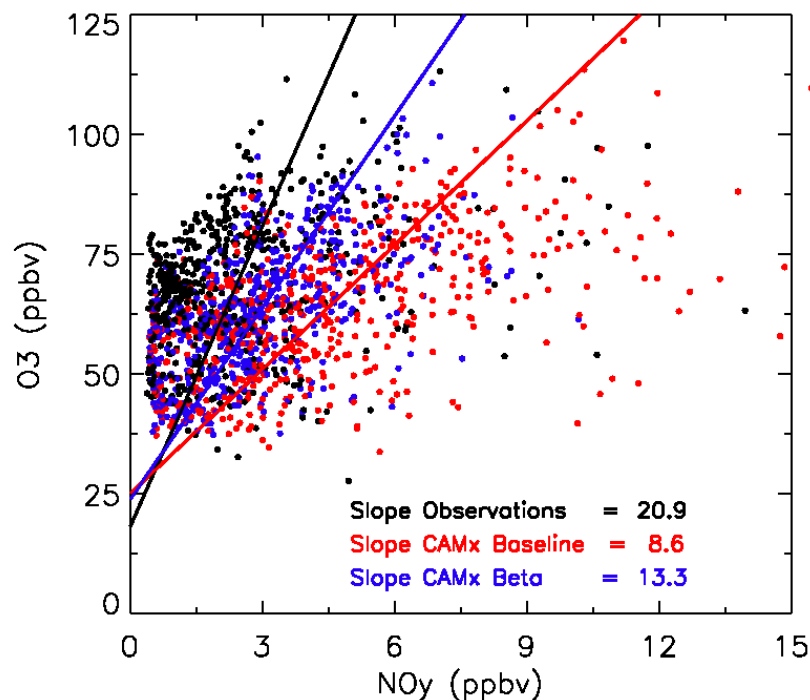


Figure S13. (Black dots) Ten-minute averaged O_3 data from the P3-B aircraft during DISCOVER-AQ Maryland in July 2011 compared to NO_y data acquired from the P3-B aircraft at the same spatial and temporal location. (Red dots) Same as black dots, except now using the CAMx Baseline simulation matched spatially and temporally to the location of the P3-B aircraft. (Blue dots) Same as red dots, except now using the CAMx Beta simulation. Each line represents the linear best fit of the data.

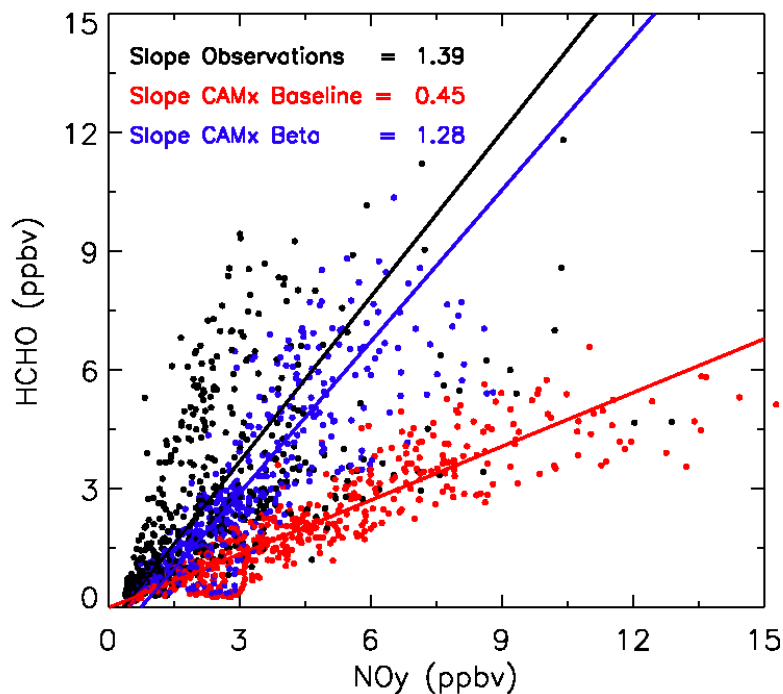


Figure S14. Same as Figure S13, but now plotting HCHO on the y-axis.

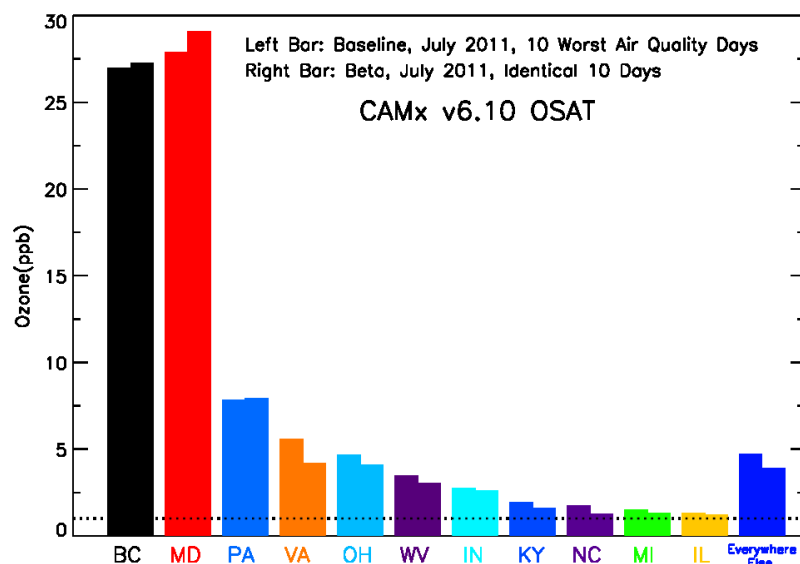


Figure S15. Ozone attributed to individual states during the ten worst air quality days in July 2011 at 2 PM local time at the Edgewood, MD monitoring site for the (left bar) baseline simulation and (right bar) Beta simulation. Only states with greater than 1 ppbv (shown as the dotted line) contribution to the Edgewood monitor are individually denoted; all other states within the modeling domain are grouped into “everywhere else”. The ozone attributed to outside the modeling domain (BC) is shown in black.

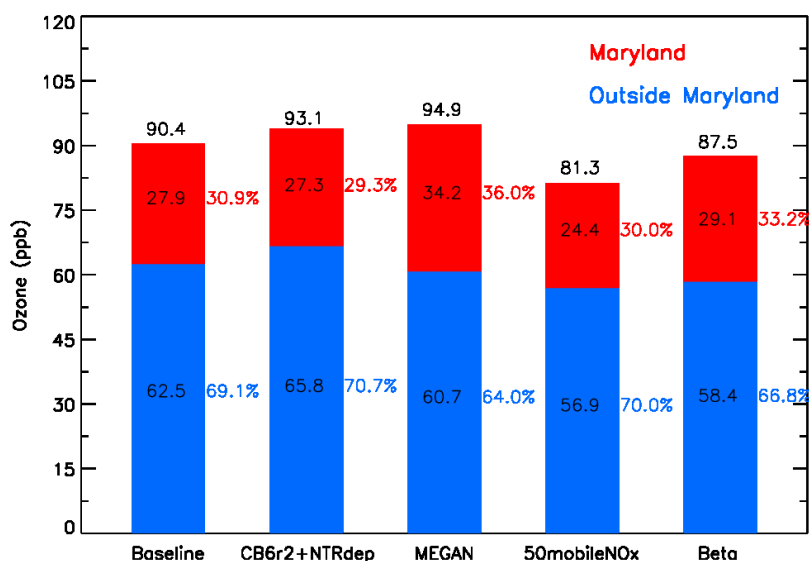


Figure S16. Ozone attributed to Maryland and to sources outside of Maryland during the ten worst air quality days in July 2011 at 2 PM local time at the Edgewood, MD monitoring site for the Baseline simulation, modified chemistry and alkyl nitrate dry deposition only simulation (CB6r2+NTRdep), modified biogenic emissions only simulation (MEGAN), modified mobile source emissions only simulation (50mobileNOx), and “Beta” simulation with all of the modifications.

Table S1. Source sectors tagged in the APCA simulation

Biogenic
On-road mobile (Mobile)
Non-road mobile (Nonroad)
C3Marine (Ship)
Electric Generating Units (EGU)
Other Point sources (Other Point)
Area sources (Area)

Table S2. CAMx version 6.10 model options used for baseline simulation

Horizontal resolution	12 km
Vertical Layers	35
Start Date	June 25, 2011
Days of Spin-up	6
Photolysis rate calculation	TUV, Discrete-ordinates method
Dry deposition model	Zhang et al. [2003]
Advection solver	Piecewise Parabolic Method
Vertical eddy diffusion	K _z -theory
Chemistry solver	Euler-backward iterative (EBI)
Gas-phase chemistry	CB05 [Yarwood et al., 2005]
Aerosol mechanism	Coarse-fine (CF)
CAMx Emissions	U.S.: 2011 NEI Version 2, Canada: 2010 NPRI
GEOS-Chem emissions	EDGAR with NEI in the U.S. [Henderson et al. 2014]

Table S3. Normalized mean bias of seven trace gases (O₃, NO_y, NO₂, NTR, HNO₃, HCHO, ISOP) in the six separate model simulations presented in this study (Baseline, CB6r2 modification-only (CB6r2), CB6r2 and increased alkyl nitrate deposition modification-only (CB6r2+NTRdepn), MEGANv2.1 biogenic emissions modification-only, (MEG2.1), 50% NO_x emissions from on-road and off-road mobile sources modification-only (50NO_x), and Beta with all four changes) when compared to observations acquired by the P3-B aircraft.

Model simulation	O₃	NO_y	NO₂	NTR	HNO₃	HCHO	ISOP
Baseline	−6.9%	+86.2%	+28.5%	+219%	+36.9%	−28.3%	−37.8%
CB6r2	−2.3%	+72.3%	+28.9%	+141%	+65.6%	−32.8%	−52.4%
CB6r2+NTRdepn	−1.8%	+65.3%	+29.0%	+102%	+60.5%	−32.0%	−50.9%
MEG2.1	−4.7%	+99.0%	+18.4%	+382%	+2.3%	+15.0%	+155%
50NO _x	−11.9%	+44.3%	−14.7%	+197%	−1.2%	−29.3%	−14.7%
CB6r2+50NO _x + MEG2.1+NTRdepn	−7.8%	+22.4%	−25.4%	+136%	−7.0%	−0.4%	+140%

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