Auxiliary Material

Table S1 Description of five emission categories of NEI.

Emission Category	Descriptions	Examples
Point	emission sources individually inventoried and usually located at a fixed location	industrial facilities and power plants
Nonpoint	individual emission sources too small in magnitude or too numerous to inventory as point sources	residential heating and consumer solvent use
Onroad	estimates from MOBILE6[a] model for highway vehicles, aircraft, locomotives, marine vessels, etc.	highway vehicles
Nonroad	estimate from NONROAD[b] model for internal combustion engine emissions	railroad equipment, construction equipment
Event	random sources of air pollution emissions	wildfires and prescribed burns

Information from NEI description
(http://www.epa.gov/ttnchie1/net/2008inventory.html)
[a]MOBILE6 Vehicle Emission Modeling Software, an emission factor model to predict gram per mile emissions of VOCs, CO, NO_x, CO_2, PM and toxics from cars, trucks, and motorcycles under various conditions. (Model website http://www.epa.gov/otaq/m6.htm)
[b]NON-ROAD model, estimate the emission inventories for all nonroad equipment with different fuel types, including VOCs, CO, NO_x, PM, SO_2, and CO_2. (Model website http://www.epa.gov/otaq/nonrdmdl.htm#model)

Table S2 List of Instruments on the Cessna 402B. (Information adapted from http://www.atmos.umd.edu/~RAMMPP/Instruments.html)

Parameter	Temporal Resolution	Detection Limit	Technique/Instrument
Position	10 s	15 m	Garmin GPS90
Pressure	10 s	0.2 hPa	Vaisala PTU 300
Temperature	10 s	0.2 °C	Vaisala PTU 300
RH	10 s	1.00%	Vaisala PTU 300
O_3	10 s	1 ppbv	UV O_3 Analyzer (TEI 49C)
CO	10 s	40 ppbv	Modified NDIR/GFC CO Analyzer (TEI 48S)
SO_2	10 s	0.3 ppbv	Modified UV Fluorescence SO_2 Analyzer (TEI 43C)
NO_2	10 s	60 ppt	Cavity Ringdown Spectroscopy (LGR[a] RMT-200)
Aerosol	2 min	0.1 μg/m^3	7 Wavelength[b] Aethalomter (Magee Scientific AE31)

Absorption			
Aerosol Scattering	30 s	0.17-0.44 Mm^{-1}	3 Wavelength[c] Integrating Nephelometer (TSI 3563)
Particle Counts	1 s	0.01 um	Condensation Particle Counter (TSI 3007)
Aerosol Size	10 s	N/A	Laser based optical (MetOne 9012)

[a]LGR: Los Gatos Research, Mountain View, CA

[b]7 wavelengths: 380, 470, 520, 590, 660, 880, and 950 nm

[c]Three wavelengths: 450, 550, and 700 nm

Table S3 List of some major point sources in Figure 2c. (Point sources with annual NO_x emissions > 1×10^3 tons/year, and NO_x/CO_2 ratio > 3.0×10^{-4})

State	Facility Name	Latitude	Longitude
OH	Avon Lake Power Plant	41.50	-82.05
OH	Kyger Creek	38.92	-82.13
OH	Muskingum River	39.59	-81.68
OH	Niles	41.17	-80.75
OH	Richard Gorsuch	39.37	-81.52
OH	Walter C Beckjord Station	38.99	-84.30
PA	Armstrong Power Station	40.93	-79.47
PA	Homer City	40.51	-79.20
PA	New Castle	40.94	-80.37
PA	Portland	40.91	-75.08
PA	Shawville	41.07	-78.37
PA	Sunbury	40.84	-76.83
PA	Titus	40.31	-75.91
WV	Albright Power Station	39.49	-79.64

Table S4 Meteorological fields used for the HYSPLIT model runs.

Year	Meteorology	Resolution	Layer
1997-2004	Eta Data Assimilation System (EDAS)	80 km	26
2004-2006	Eta Data Assimilation System (EDAS)	40 km	26
2007-2011	North America Mesoscale (NAM)	12 km	26

Figure S1. Statistics of RAMMPP research spirals in the Baltimore/Washington airshed.

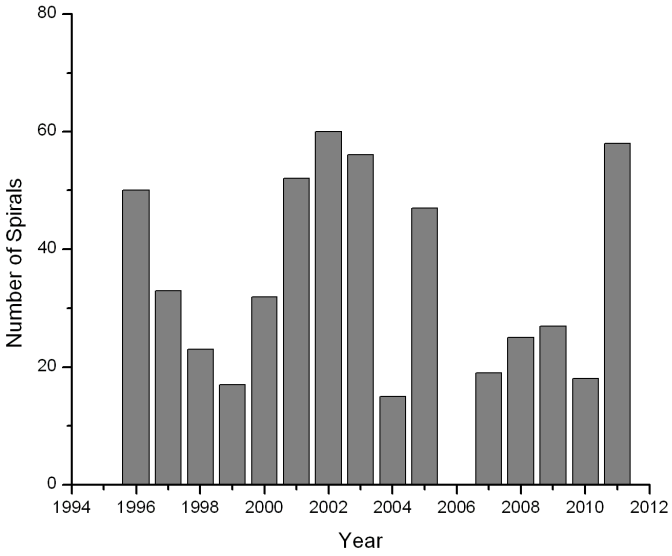


Figure S2. Trends of NO_x and CO emissions of the U.S. and in the research domain. Annual U.S. emissions are scaled (4% and 5% for NO_x and CO respectively) for better demonstration.

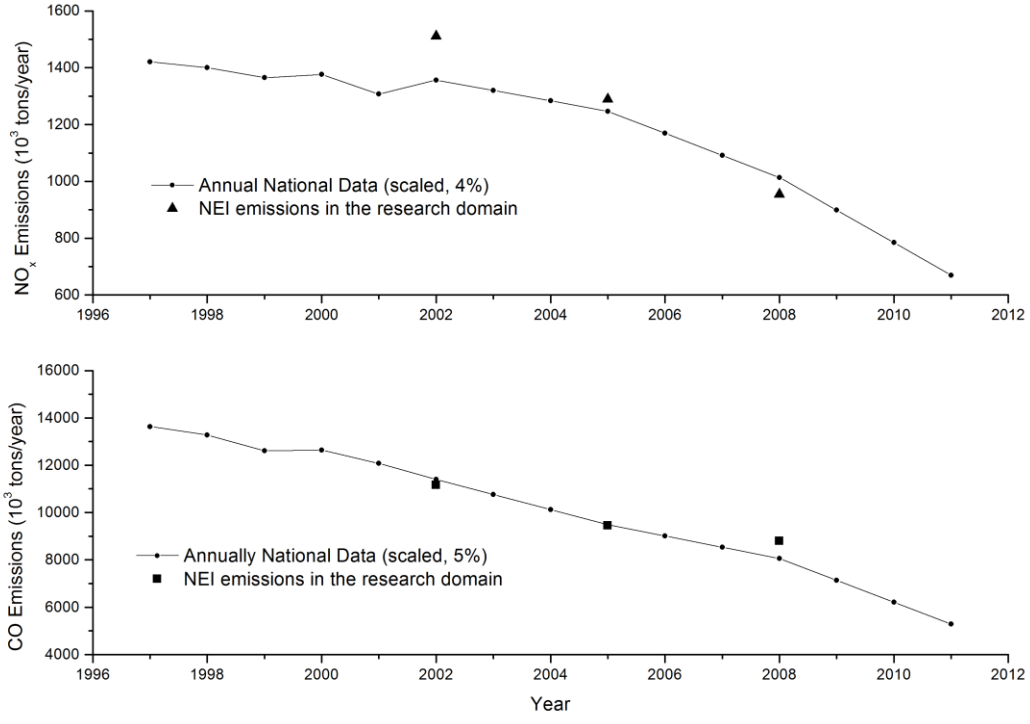


Figure S3. Long-term trends of CEMS CO_2 emissions in the research domain.

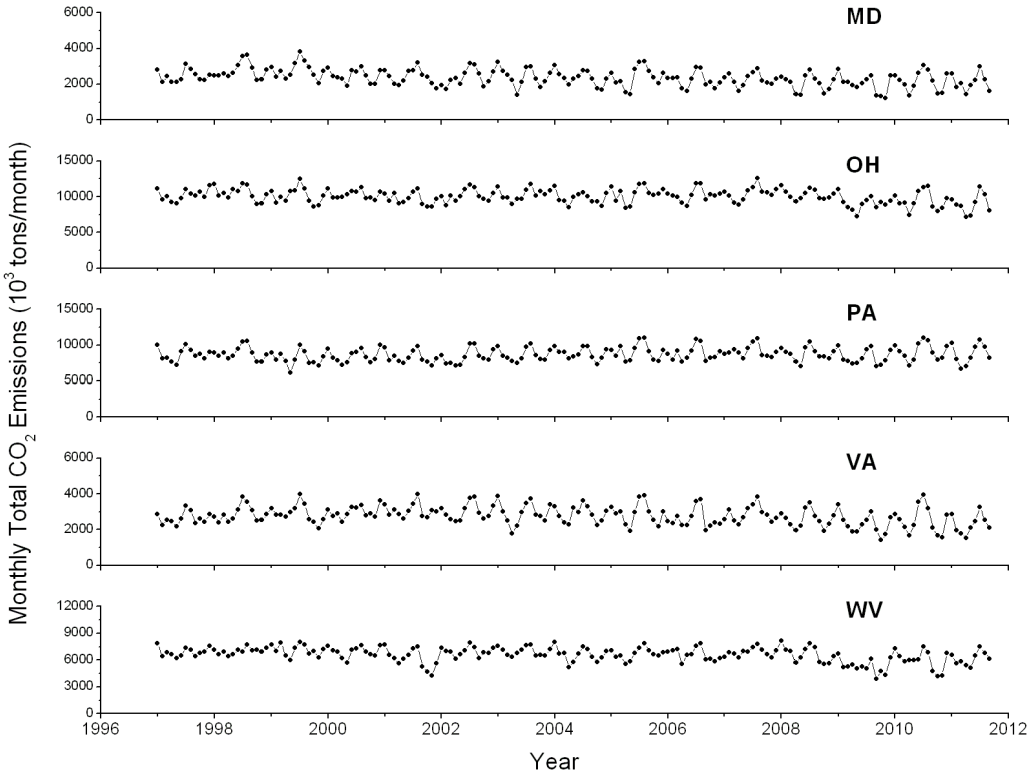


Figure S4. Locations of EPA AQS sites of ground-level O_3, CO, and NO_x measurements (valid sites in the last 15 years)

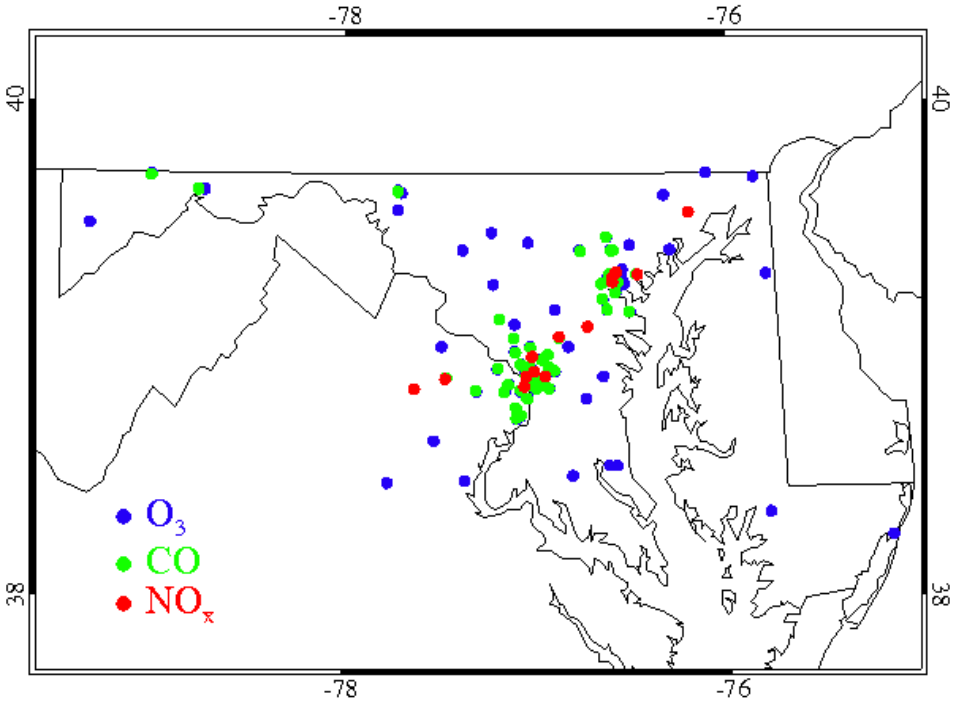
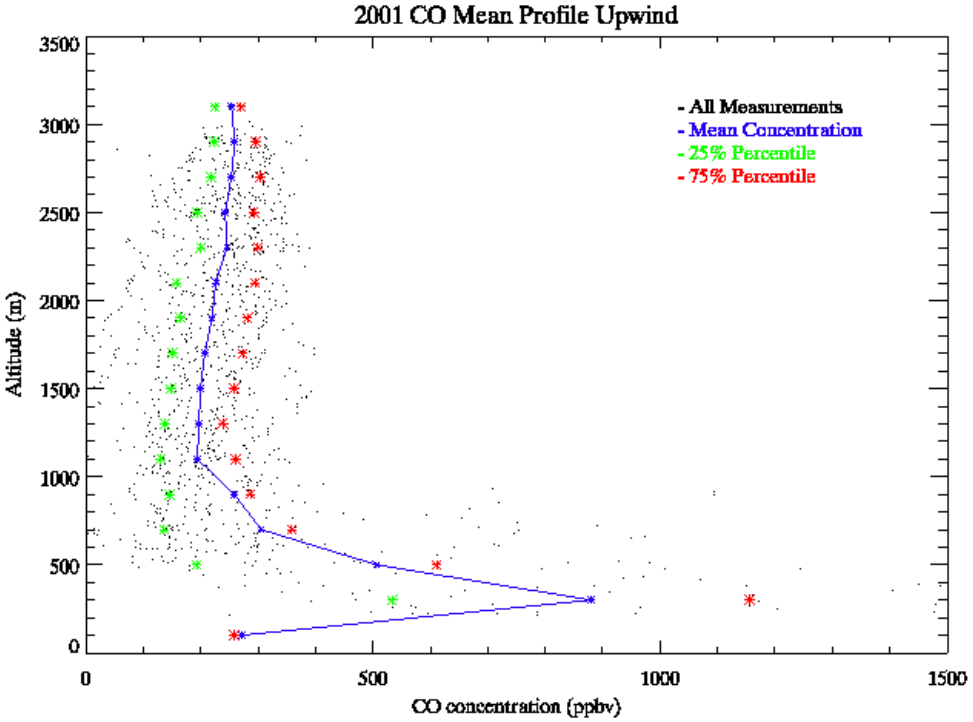


Figure S5 Vertical distributions of tropospheric CO over the Baltimore/Washington region in 2001. (a: only the upwind spirals; b: only the downwind spirals).

a)



b)

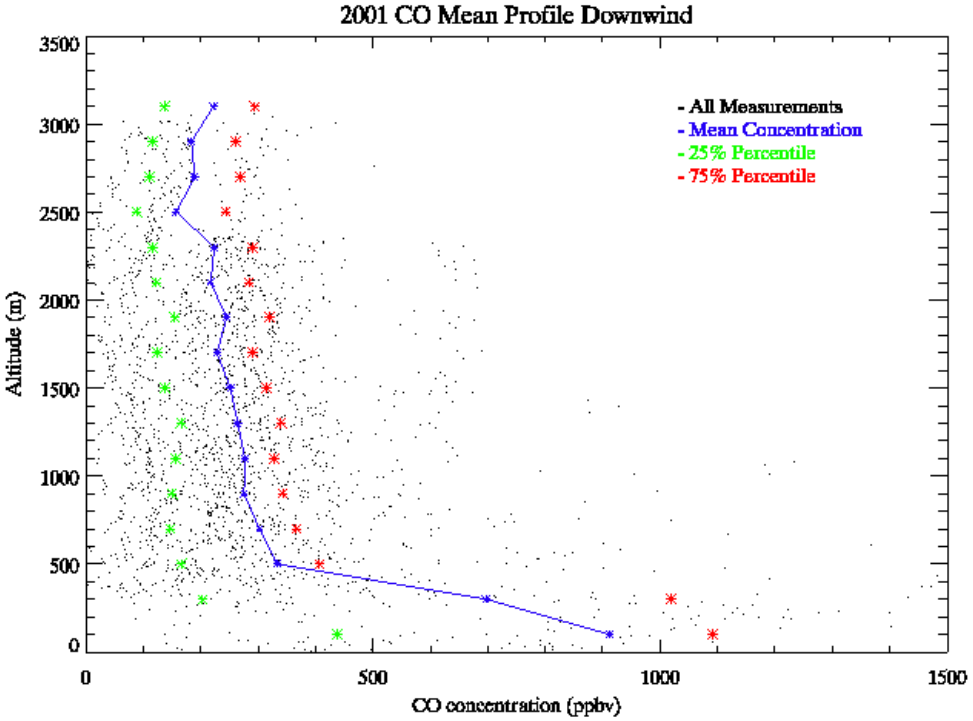


Figure S6. Number of day with daily mean temperature (T_{mean}) high than 28 °C Temperature measurements are collected in the Baltimore Washington International Airport.

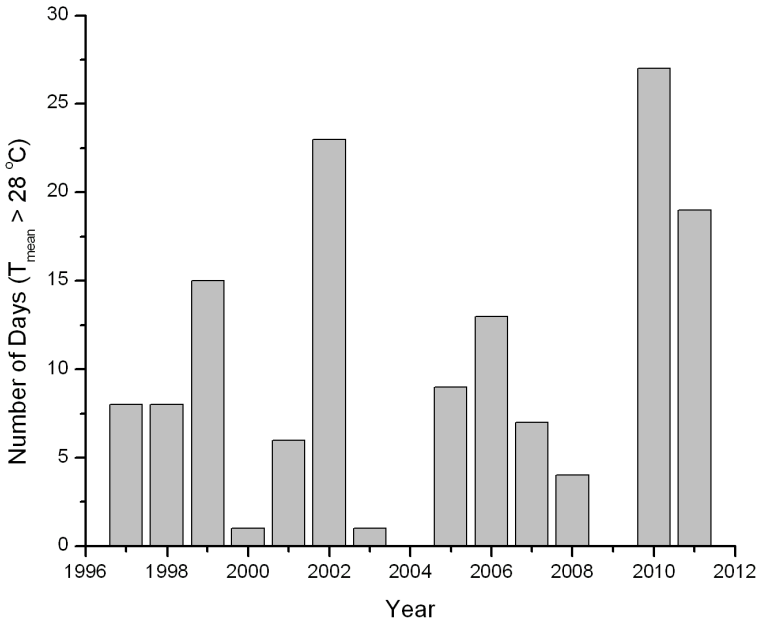
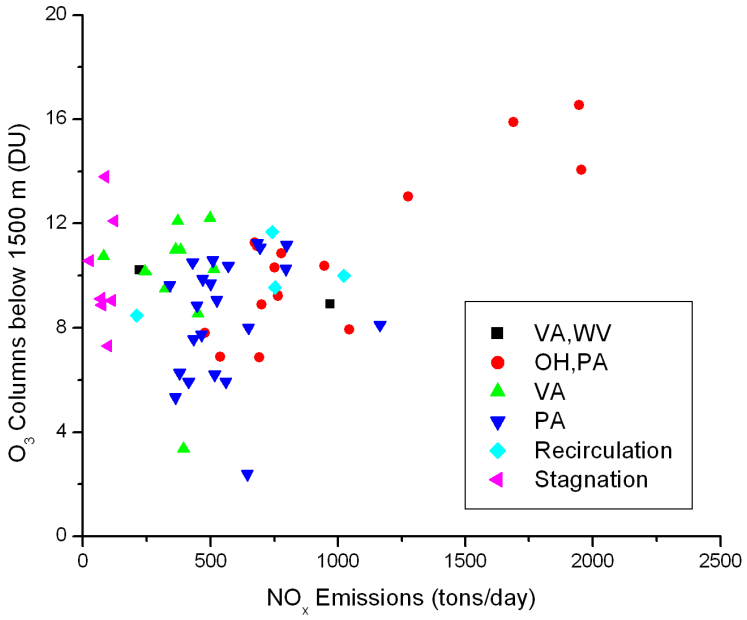
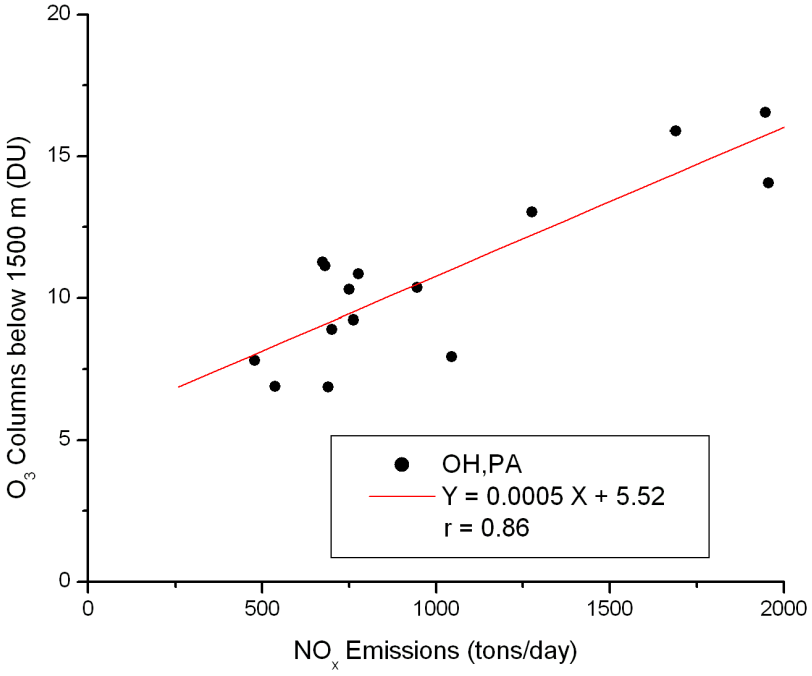


Figure S7. Similar plots as Figure 13 except in a) and b) all coefficients in Table 1 are doubled, e.g., Equation 1 is expressed in $Emission_{total}=Emission_{MD,i}+Emission_{PA,i-1}+0.5Emission_{OH,i-1}$; c) and d) all coefficients in Table 1 are divided by two, e.g., Equation 1 is expressed in $Emission_{total}=Emission_{MD,i}+0.25Emission_{PA,i-1}+0.125Emission_{OH,i-1}$

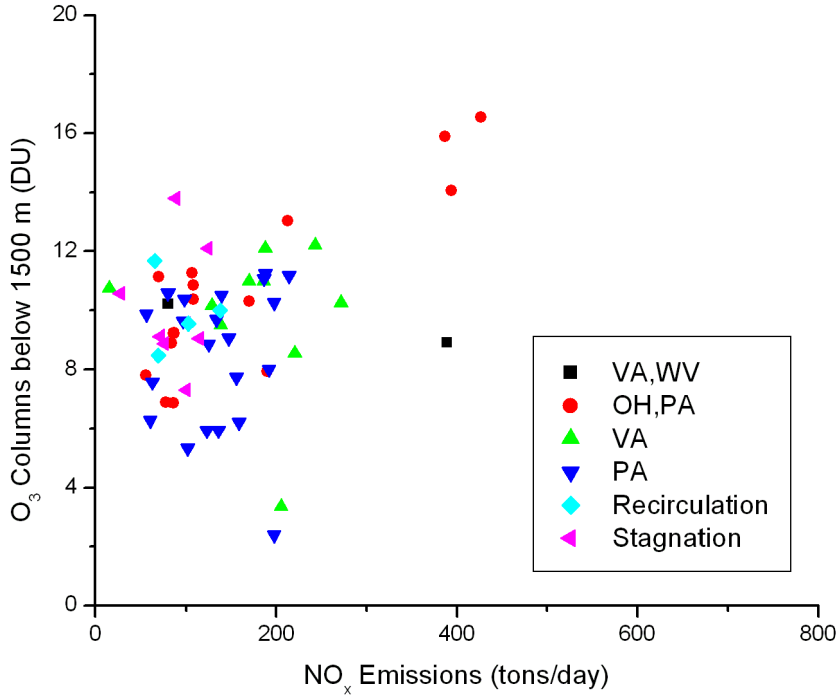
a)



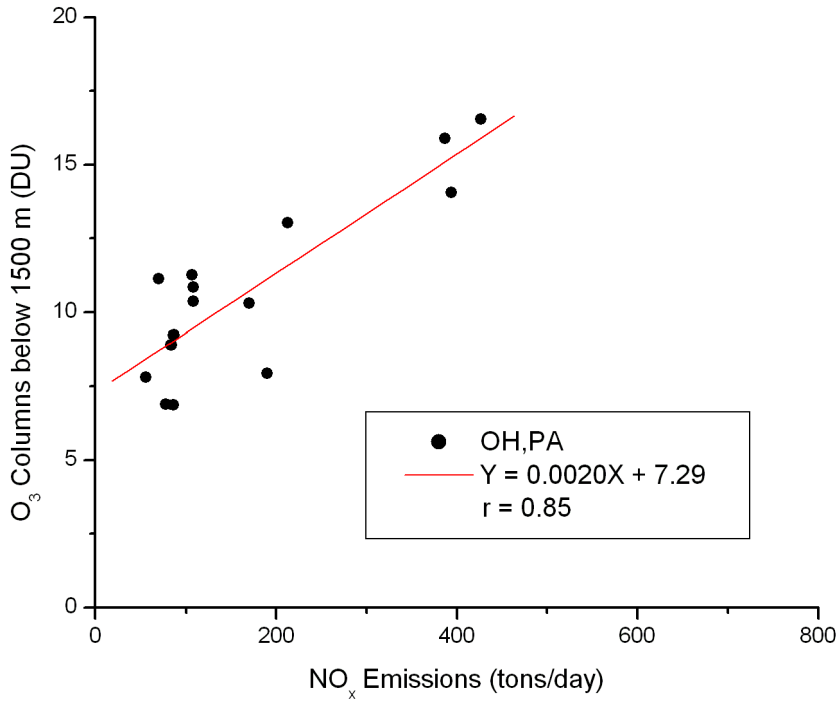
b)


20
15
10
5
0
0
500
1000
1500
2000
O3 Columns below 1500 m (DU)
NOx Emissions (tons/day)
OH,PA
Y = 0.0005 X + 5.52
r = 0.86

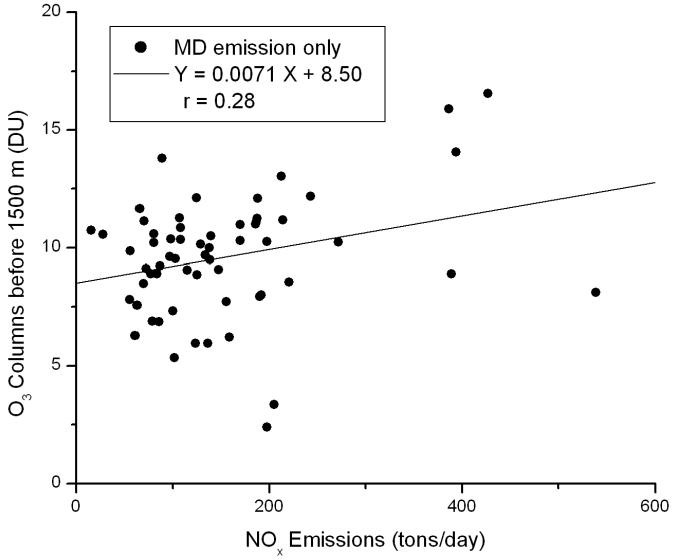
c)


20
16
12
8
4
0
0
200
400
600
800
O3 Columns below 1500 m (DU)
NOx Emissions (tons/day)
VA,WV
OH,PA
VA
PA
Recirculation
Stagnation

69 d)



70
71
72
73 Figure S8. RAMMPP ozone column contents versus CEMS NO_x emissions in MD



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