Auxiliary Material

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

Table S1 Description of five emission categories of NEI.

Information from NEI description

(http://www.epa.gov/ttnchie1/net/2008inventory.html)

^aMOBILE6 Vehicle Emission Modeling Software, an emission factor model to predict gram per mile emissions of VOCs, CO, NO_x, CO₂, PM and toxics from cars, trucks, and motorcycles under various conditions. (Model website http://www.epa.gov/otaq/m6.htm)

^bNON-ROAD model, estimate the emission inventories for all nonroad equipment with different fuel types, including VOCs, CO, NO_x, PM, SO₂, and CO₂. (Model website <u>http://www.epa.gov/otaq/nonrdmdl.htm#model</u>)

Table S2 List of Instruments on the Cessna 402B. (Information adapted from

	Temporal	Detection	
Parameter	Resolution	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)
СО	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)

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NO ₂	10 s	60 ppt	Cavity Ringdown Spectroscopy (LGR ^a RMT-200)
Aerosol			
Absorption	2 min	$0.1 \ \mu g/m^3$	7 Wavelength ^b Aethalomter (Magee Scientific AE31)
Aerosol		0.17-0.44	
Scattering	30 s	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)

^aLGR: Los Gatos Research, Mountain View, CA

^b7 wavelengths: 380, 470, 520, 590, 660, 880, and 950 nm

^cThree wavelengths: 450, 550, and 700 nm

Table S3 Major point sources of NO_x in Figure 2c. (Point sources with annual NO_x emissions $> 1 \times 10^3$ tons/year, and NO_x/CO₂ ratio $> 3.0 \times 10^{-4}$)

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. Number of RAMMPP research spirals conducted in the Baltimore/Washington airshed for various summers during the past 15 years.



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) to present the emissions in the research domain.



Figure S3. Long-term trends of CEMS CO₂ 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 past 15 years)



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



b)



Figure S6. Number of days with daily mean temperature (T_{mean}) higher than 28 °C, based on measurements collected at the Baltimore Washington International Airport.



Case studies of single back trajectories and 27-member ensembles from Figure S7. HYSPLIT. Nine cases of back trajectories are investigated, all driven by the Eta Data Assimilation System (EDAS) 40 km meteorological fields for years 2004 to 2005 (Table S4). We have examined ensemble trajectories for the dates of 06/21/2004, 06/21/2005, 06/25/2005, 06/26/2005, 07/10/2005, 07/11/2005, 08/02/2005, 08/13/2005, and 09/13/2005. In each plot, the left panel shows single back trajectories initiated at 500 m, 1000 m, and 1500 m AGL, respectively; the right panel shows the 27-member ensemble back trajectories initiated at 1000 m AGL. Each member of the trajectory ensemble is calculated by offsetting the initial point of the backwards trajectory by either -1, 0, or +1 (hereafter ± 1) grid point in the east/west direction, ± 1 grid point in the north/south direction, and ±0.01 sigma unit (about 250 m) in the vertical. The 27 member ensemble consists of 9 points in 3 vertical planes, representing all possible combinations of these perturbations plus the central point. Further details given at http://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive. Comparison of the single back trajectories initiated at 1000 m AGL (blue line in the left panel) to the ensemble back trajectories initiated at 1000 m AGL shows that the single back trajectories accurately represent the behavior of the ensemble over the past 24 to 48 hrs. Our study focuses on regional transport of pollution over a much smaller area and much shorter period of time than considered in Stohl et al. (2002; 2003). Hence, the divergence between various members of the ensemble is small. These results support the validity of our approach of using single back trajectories in the analysis of aircraft data.





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Figure S8. Case studies of HYSPLIT single back trajectories and the 24-hr backward concentration dispersion. Nine cases of back trajectories (for same dates as described in Figure S7) are investigated. In each plot, the left panel shows single back trajectories initiated at 500 m, 1000 m, and 1500 m AGL, respectively; the right panel shows 24-hr backward concentration contours averaged in the lowest 1500 m with unit of air pollutant released at 1000 m AGL. The HYSPLIT backward concentration dispersion demonstrates the possible origins of air pollutants, within the most recent 24-hr period of time, for air pollution observed by the RAMMPP aircraft. The comparison of single back trajectories (the left panel) and the 24-hr backward concentration dispersion (the right panel) shows that the single back trajectories approximately pass over the locations of upwind sources. The source apportionment footprint suggested by the 24-hr backward concentration (the right panel) is comparable to the area of a state in the eastern U.S., supporting the approach we have used to estimate emissions of power plants grouped by state.





























NOAA HYSPLIT MODEL Backward trajectories ending at 1900 UTC 13 Aug 05







Figure S9. Similar plots as Figure 13 except in a) and b) all coefficients in Table 1 are doubled, e.g., Equation 1 is expressed in $Emission_{total} = Emission_{MD,i} + Emission_{PA,i-1} + 0.5 Emission_{OH,i-1}$; c) and d) all coefficients in Table 1 are divided by two, e.g., Equation 1 is expressed in $Emission_{total} = Emission_{MD,i} + 0.25 Emission_{PA,i-1} + 0.125 Emission_{OH,i-1}$ a)





c)





Figure S10. RAMMPP ozone column abundances versus CEMS NO_x emissions in

