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8	Aquino ⁴ , Teresa Campos ⁴ , Viral Shah ⁵ , Lyatt Jaeglé ⁵ Joel A. Thornton ⁵ , Joshua P.
9	DiGangi ⁶ , Glenn M. Wolfe ^{7,8} Solomon Bililign ¹ , Steven S. Brown ^{2,9}
10	
11	¹ Department of Physics & Applied Sciences and Technology, North Carolina A&T State
12	University, Greensboro NC, 27411
13	² NOAA Chemical Sciences Laboratory, Boulder, CO, 80305
14	³ Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder,
15	CO, 80309
16	National Center of Atmospheric Research, Boulder, CO, 8030/
1/	⁶ NASA Longley Descereb Center, Hermiten VA, 22681
18	⁷ Atmospheric Chamistry and Dynamics Lab. NASA Coddard Space Elight Center, Creenholt
20 19	Atmospheric Chemistry and Dynamics Lab, NASA Goddard Space Flight Center, Greenbell, MD 20771
20 21	⁸ Joint Center for Earth Systems Technology University of Maryland Baltimore County
21 22	Baltimore MD 21228
23	⁹ Department of Chemistry University of Colorado Boulder CO 80309
24	+Current address California Air Resources Board, Sacramento, CA, USA
25	[†] Current address Department of Energy, Environmental, and Chemical Engineering,
26	Washington University in St. Louis, St. Louis, MO, USA
27	
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31	Introduction

Data sets used in this study were obtained from the WINTER C-130 1s Merged Data set with the 32 exception of the reference GEOS-Chem and improved GEOS-Chem data sets which were 33 obtained from Shah et al. [2018]. Both the reference and improved data sets from Shah et al. 34 [2018] were merged and expanded to match the 1s time steps in the WINTER C-130 data set. 35 For example, for a chemical species data from the GEOS-Chem simulation was listed at a given 36 37 [HCHO] at one data point for the time interval start time of 10:59:00 UTC to the end time of 11:00:00 UTC. Within the time interval 60 data points were created listing the same [HCHO] 38 for each data point, completing the expansion of into 1s time steps within time domain of 39 10:59:00 - 11:00:00 UTC. This allowed for the GEOS-Chem data to match the 1s WINTER C-40 130 data sets. Data sets were temporally aligned by first selecting a primary instrument, using the 41 NASA ISAF instrument for example, and synchronizing the other data sets to the primary 42 instrument. For instance, in RF 03 the CO measurements from the Picarro CRDS were shifted 43 by -8 seconds to match the primary instrument. 44

45

46 Table S1

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	HCHO/CO by plume (percentage)												
RF	Avg	1	2	3	4	5	6	7	8	9	10	11	12
02	0.36	0.41	0.56	0.47	0.43	0.85	0.43	0.32	0.28	8.6			
03	0.57	0.59	0.56	0.57	0.68								
04	0.55	1.4	1.6	1.2	2.7	1.6	2.7						
10	0.97	1.8	1.2	2.0	1.5	0.66	0.29	0.35	0.34	1.9	2.6	0.75	0.0

Summary of HCHO/CO- Note. The plumes listed above in the table come from distinct plumes identified.
The average was taken for the total amount of data points collected in the region covered by the flight.



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Figure S1. Progression of enhancement ratios of HCHO (black circles), toluene (blue squares), and benzene (red triangles) in the Cincinnati/Columbus plume encountered on RF02. Enhancement ratios are calculated for each plume relative to CO (a dilution marker) as described in the main text. Lagrangian time is from a trajectory analysis and is relative to the first plume intercept, ~0.4h downwind of Cincinnati





text are where evidence of mixing was found. Locations indicated in black text indicate where stable atmospheric conditions allowed for the formation of the nocturnal and residual layers.



Figure S3: Research Flight 10 (RF10) on March 07, 2015, over the city of Atlanta, GA and moving south and toward the coast, before ending 100 km west of Savanna, GA. Missed approaches (MA) are marked by tags in red boxes, the numbers in purple and black mark the urban plumes. Background shading indicates terrain elevation. The color scale shows the HCHO mixing ratio measured along the flight track.



Figure S4: Vertical profiles for RF 10 over Atlanta. (Left) taken from MA# 1, (Right) taken from MA#4. Colors in the legend indicate HCHO, CO, and potential temperature.



Figure S5: Research Flight 04 (RF04) on February 11, 2015. Tags with the yellow lines indicate names of power stations associated with plume intercepts indicated by the numbered tags with dashed lines. The violet to white dots represent the location of co-generating and coal fired electric power generation stations, sized by NO₂ emissions from the CEMS database on a

Log10 scale, as shown in the legend in the upper right. Other features are described in Figure 4.



Figure S6: Research Flight 07 (RF07) on February 24, 2015. Tags with the yellow lines indicate names of power stations associated with plume intercepts indicated by the numbered tag with dashed line. The violet to white dots represent the location of co-generating and coal fired electric power generation stations, sized by NO_2 emissions from the CEMS database on a Log10 scale, as shown in the legend in the lower left. Other features are described in Figure 4.

