

Now what have we gotten into? (A brief look at data from the Southeast and a closer look at CTR)

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SOAS/SENEX/NOMMADS Science Coordination Meeting
Washington, DC
March 13, 2012



Outline

- Variability of PM and Trace Gases over Space and Time
- A Closer look at CTR Data (what might we expect in June-July 2013?)
 - Trace Gases
 - PM
 - Meteorology

The Southeastern Aerosol Research and Characterization (SEARCH) study: Temporal trends in gas and PM concentrations and composition 1999–2010

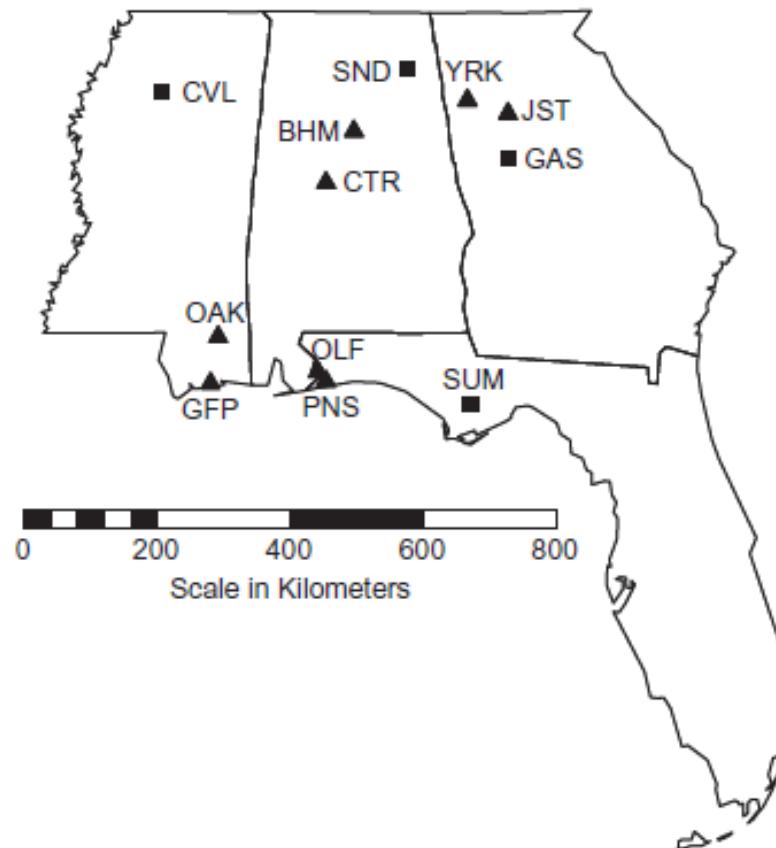
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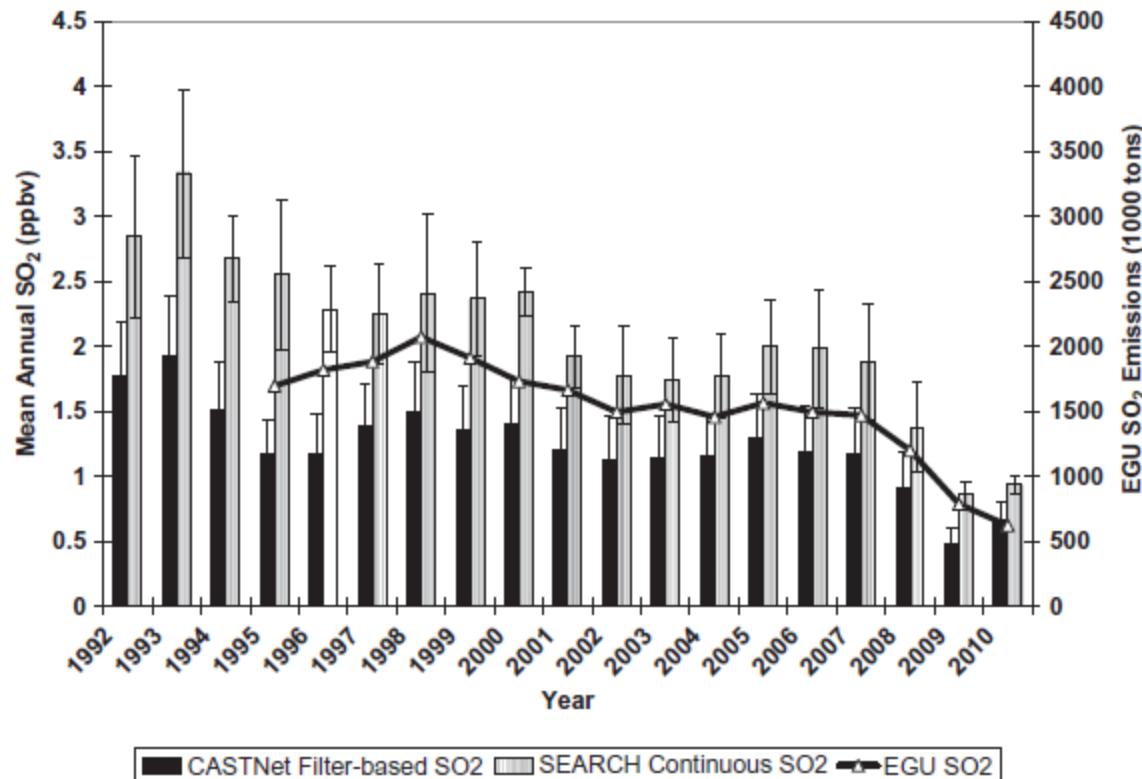
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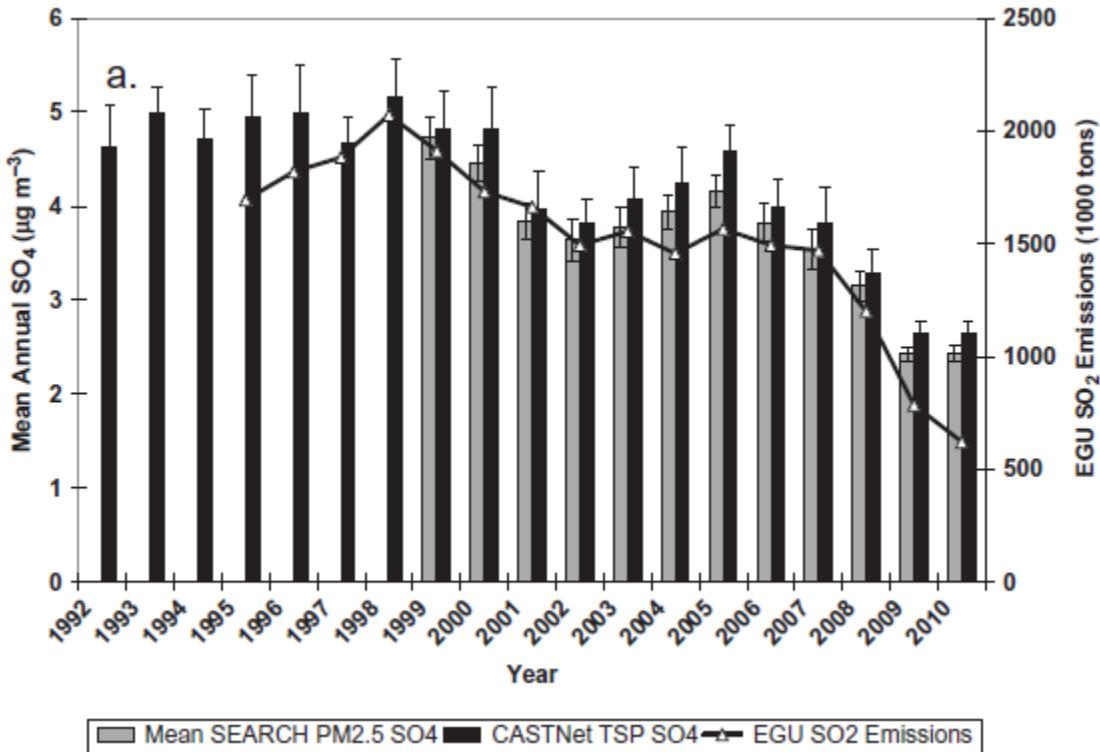
Emissions Trends

Year	CO	NH ₃	NO _x	PM ₁₀	PM _{2.5}	SO ₂	VOC	Mobile VOC ^a	On-road NO _x ^a	Nonroad NO _x ^a	EGU NO _x ^b	EGU SO ₂ ^b	EGU CO ₂ ^b
1990	18.22	0.34	2.62			2.80	2.59	1.69	1.25	0.40		2.17	
1991								1.62	1.23	0.41			
1992								1.56	1.21	0.42			
1993								1.49	1.19	0.43			
1994								1.42	1.17	0.43			
1995								1.35	1.15	0.44	0.72	1.70	274.6
1996	14.03	0.37	2.63	2.03	0.59	2.39	2.22	1.29	1.13	0.45	0.73	1.82	284.8
1997	13.72	0.37	2.66	2.12	0.61	2.46	2.20	1.23	1.14	0.45	0.73	1.88	298.8
1998	13.65	0.39	2.69	2.14	0.63	2.64	2.17	1.20	1.12	0.44	0.77	2.07	313.7
1999	15.99	0.36	2.70	2.66	1.03	2.46	2.42	1.17	1.09	0.44	0.73	1.91	316.1
2000	14.47	0.36	2.68	2.40	0.86	2.28	2.17	1.12	1.09	0.45	0.72	1.73	326.1
2001	15.16	0.32	2.62	2.39	0.88	2.23	2.19	1.06	1.01	0.44	0.68	1.66	318.8
2002	13.99	0.32	2.44	2.04	0.60	2.04	2.74	1.12	1.02	0.48	0.61	1.49	328.5
2003								1.07	0.96	0.50	0.56	1.56	335.8
2004								1.03	0.90	0.51	0.51	1.46	333.7
2005	12.48	0.33	2.25	2.12	0.68	2.11	2.66	0.98	0.84	0.52	0.50	1.57	349.8
2006								0.94	0.79	0.49	0.48	1.49	350.3
2007								0.90	0.74	0.45	0.46	1.47	361.0
2008 ^c	9.22	0.30	1.75	1.75	0.43	1.46	1.54	0.86	0.69	0.42	0.42	1.20	339.0
2009								0.75	0.62	0.36	0.23	0.79	296.2
2010			1.33 ^d			0.88 ^e		0.63	0.56	0.31	0.24	0.62	338.6

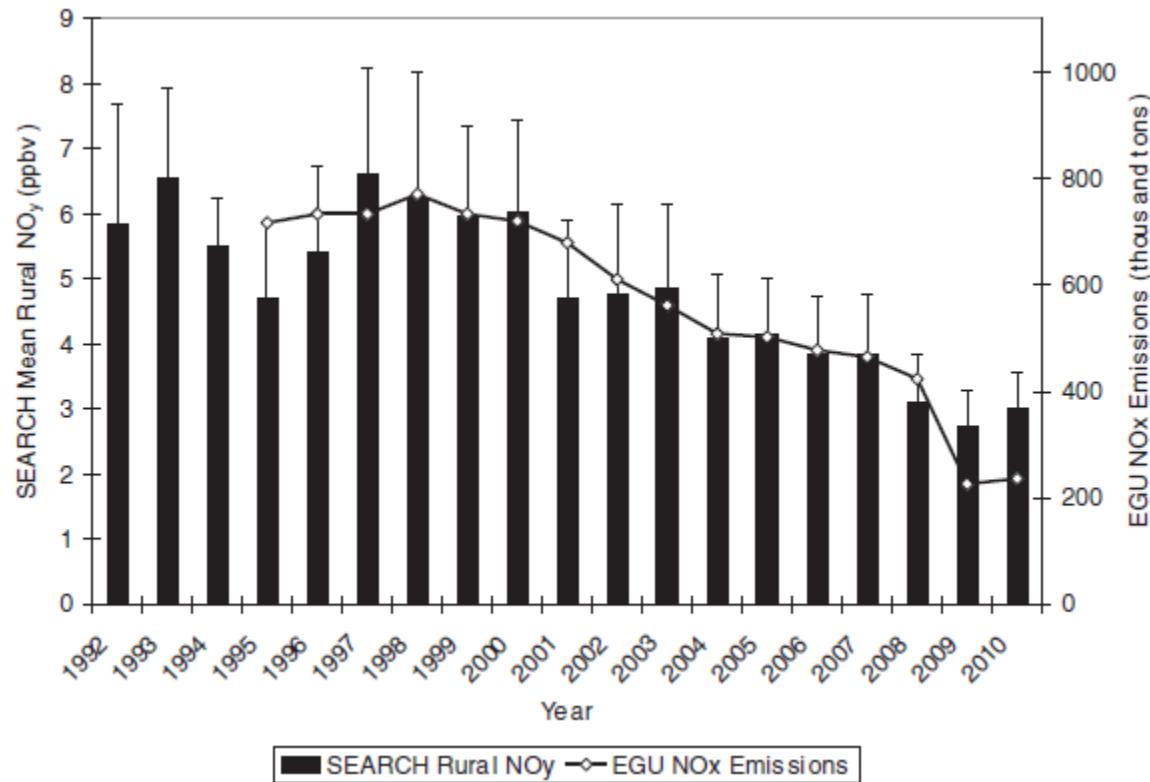
SO₂ Emissions and Concentrations Trends



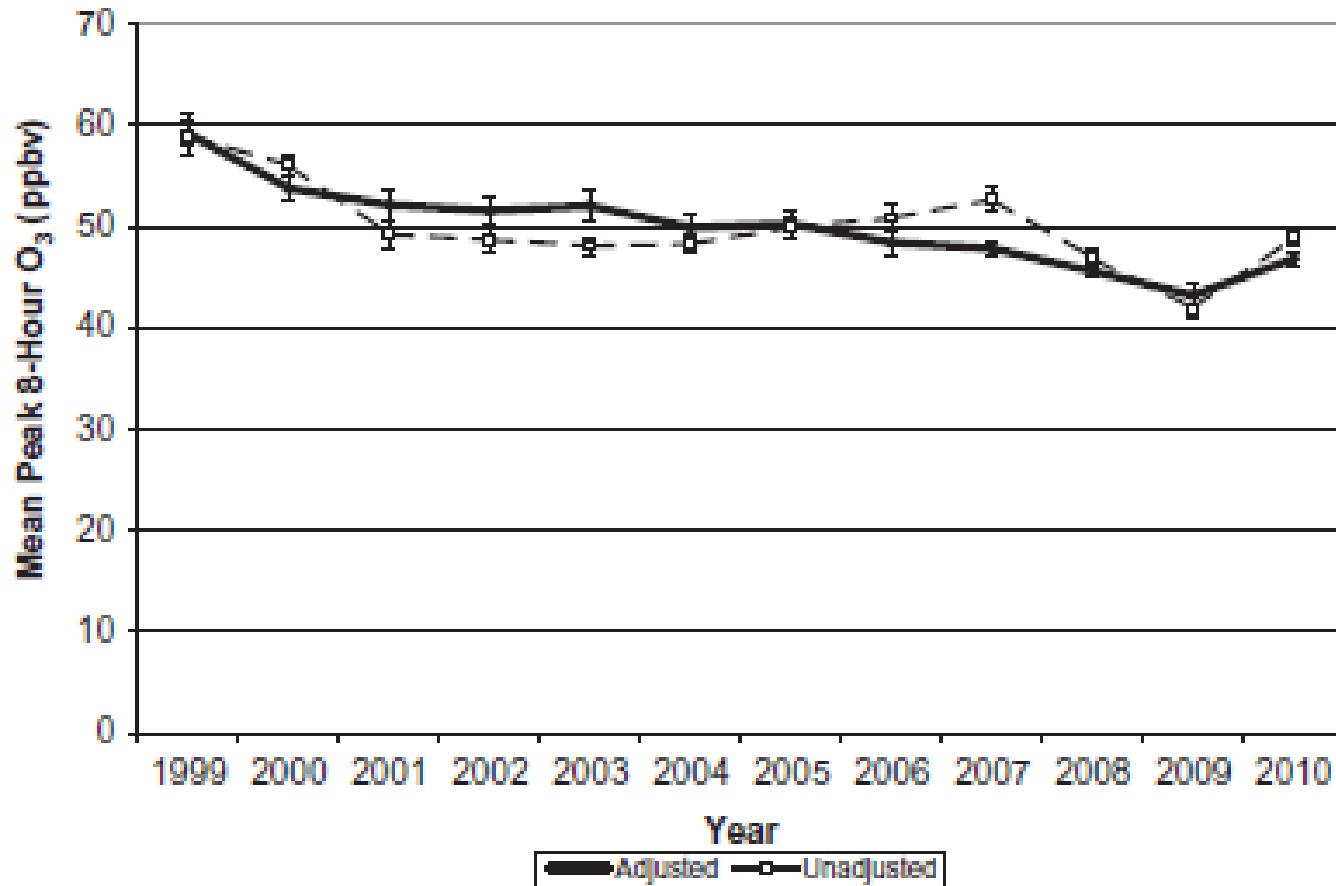
SO₄ Concentrations and SO₂ Trends



NO_X and NO_y Trends



Meteorologically adjusted Ozone

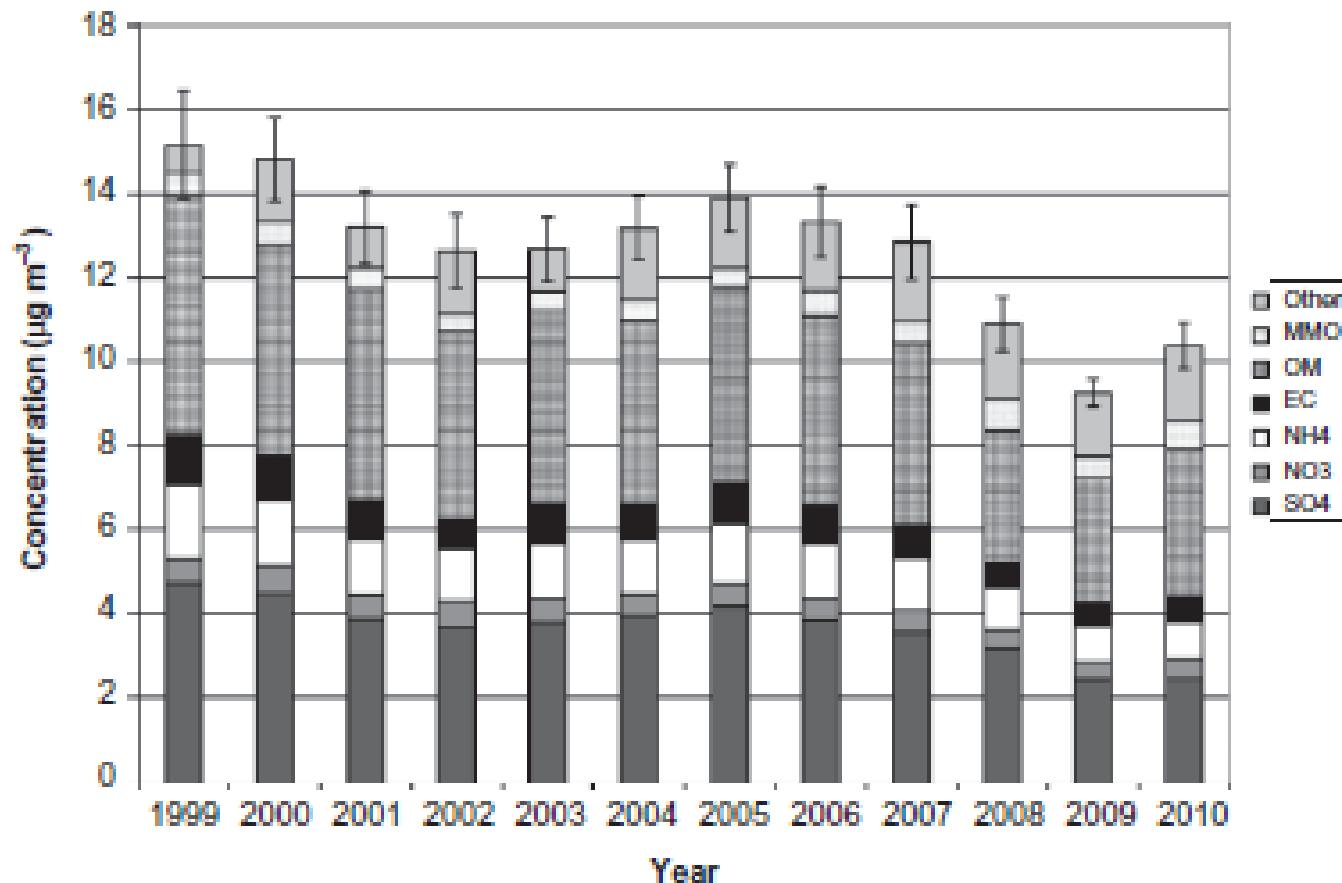


Annual Rate of Change for SEARCH Gases and PM_{2.5} (%/yr)

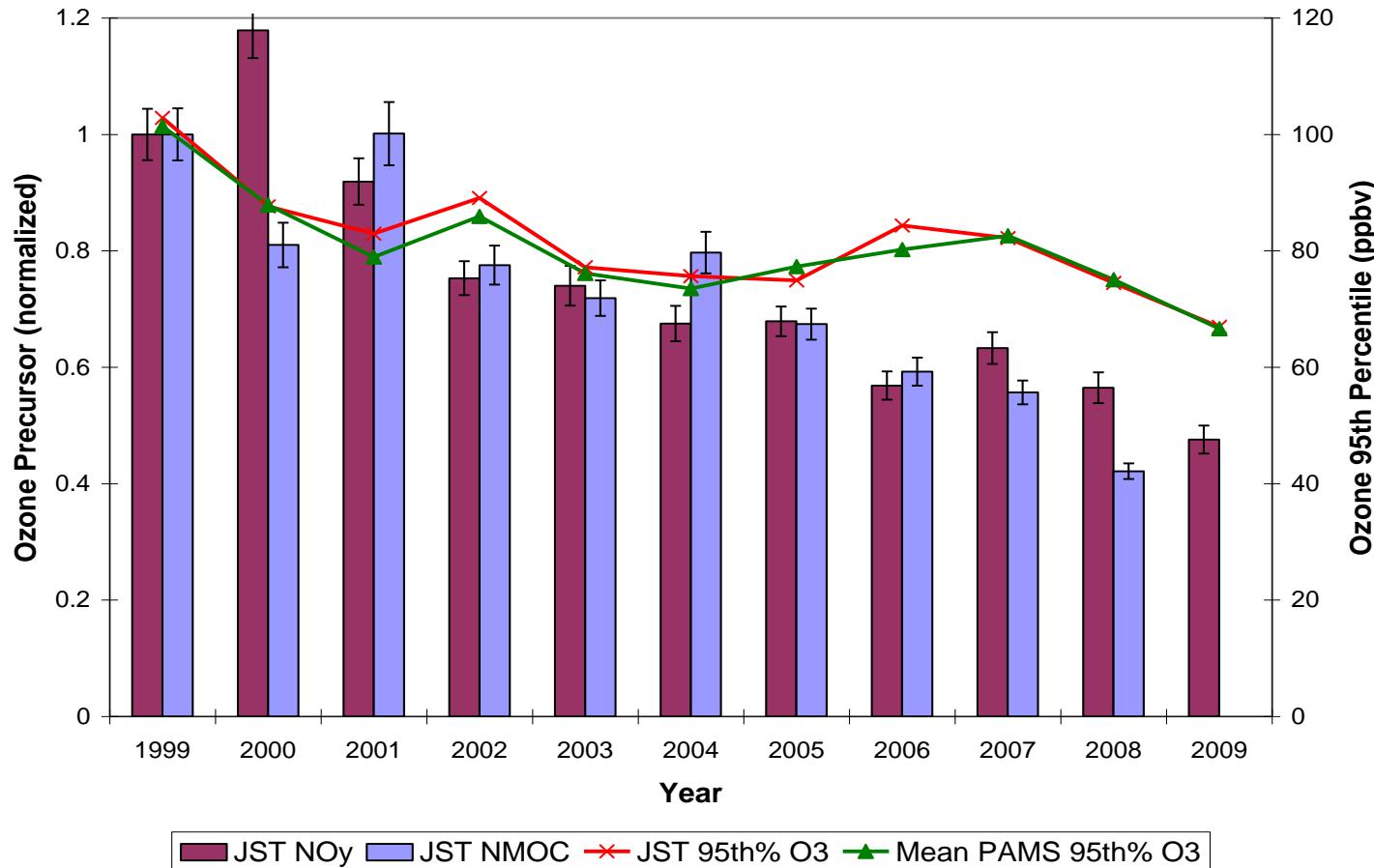
Site	SO ₂	SO ₄	NO _y	NO ₃	O ₃	CO	EC	OM
BHM	-5.1 ± 2.0	-5.2 ± 1.0	-6.6 ± 0.8	-5.8 ± 0.9	-1.9 ± 0.9	-6.0 ± 0.6	-6.6 ± 1.8	-6.1 ± 1.3
CTR	-6.8 ± 1.4	-6.2 ± 1.1	-6.1 ± 0.8	-3.3 ± 1.3	-3.1 ± 0.8	-1.6 ± 1.0	-1.3 ± 1.9	-3.3 ± 0.8
GFP	-7.1 ± 1.4	-4.6 ± 0.8	-7.4 ± 1.1	-0.6 ± 1.2	-1.5 ± 0.6	-5.0 ± 0.8	-5.5 ± 1.6	-4.1 ± 0.9
JST	-9.1 ± 1.7	-5.5 ± 1.1	-7.3 ± 0.8	-4.7 ± 0.6	-1.8 ± 0.7	-7.2 ± 0.4	-6.8 ± 1.3	-5.0 ± 1.0
OAK	-8.0 ± 1.5	-4.2 ± 0.8	-6.0 ± 0.9	-0.9 ± 1.2	-1.9 ± 0.4	-2.2 ± 0.7	-4.0 ± 1.6	-4.9 ± 1.2
OLF	-9.7 ± 1.8	-4.4 ± 0.9	-7.8 ± 0.7	-2.3 ± 1.1	-2.0 ± 0.5	-1.2 ± 0.4	-4.9 ± 1.3	-4.5 ± 1.2
PNS	-4.1 ± 8.4	-3.7 ± 1.1	-9.0 ± 1.3	-1.7 ± 0.8	-2.2 ± 0.6	-6.5 ± 0.5	-7.8 ± 0.7	-6.5 ± 0.3
YRK	-7.6 ± 2.3	-5.3 ± 1.3	-7.7 ± 0.7	-4.0 ± 0.9	-2.8 ± 0.5	-1.6 ± 0.7	-6.0 ± 1.8	-4.0 ± 1.3

BLANCHARD, ET AL., 2012. THE SOUTHEASTERN AEROSOL RESEARCH AND CHARACTERIZATION (SEARCH) STUDY: TEMPORAL TRENDS IN GAS AND PM CONCENTRATIONS AND COMPOSITION, 1999-2010. SUBMITTED.

SEARCH-Average PM_{2.5} Mass and Composition

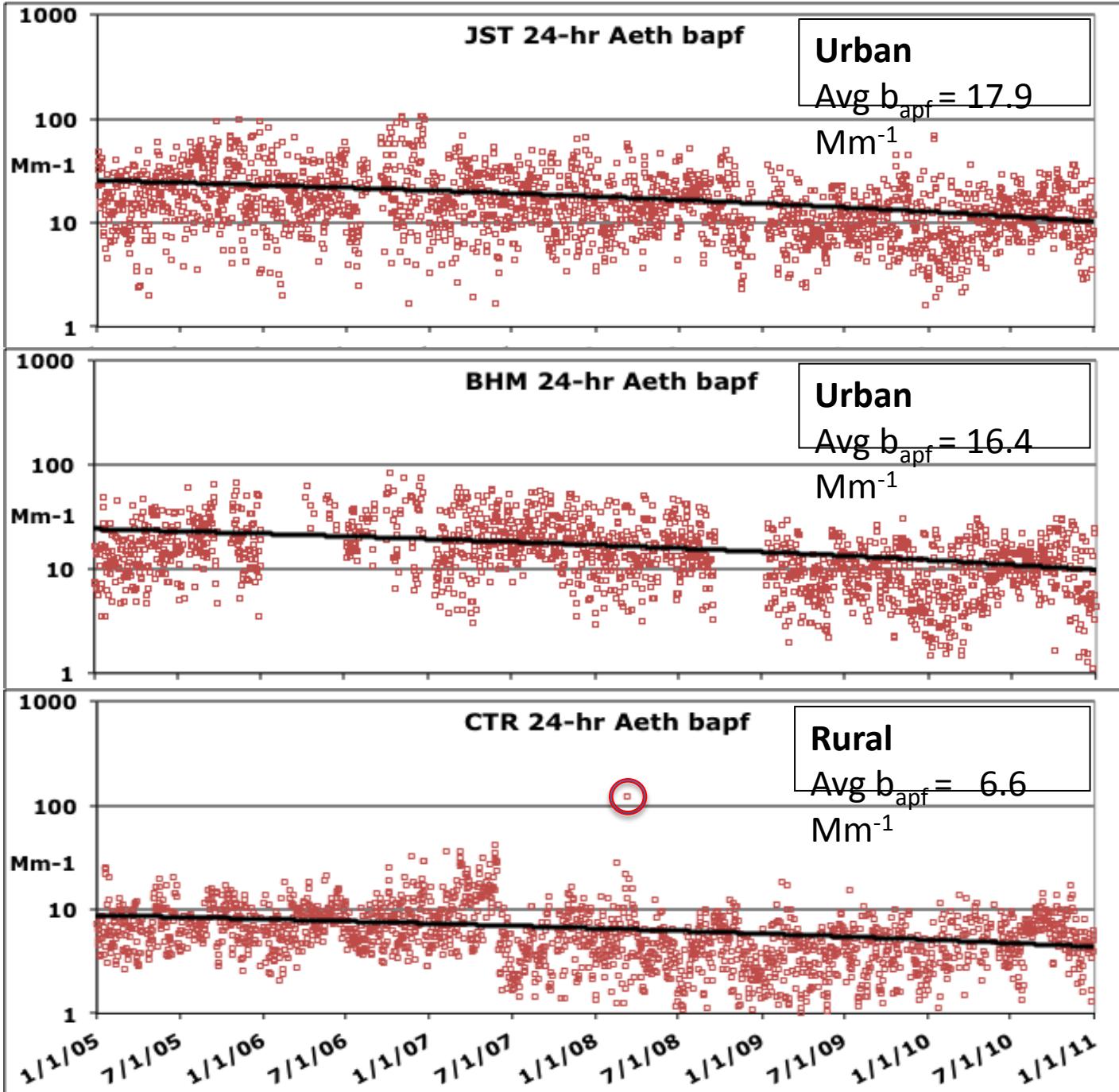


VOCs (JST only) and O₃ also decreasing



Dry b_{apf} Trends 2005- 2010

Note: log scale

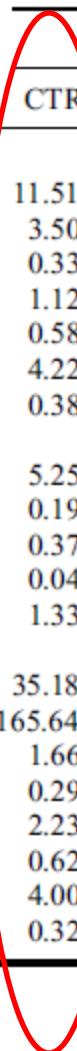


In 6 years, urban b_{apf} fell by ~45% and rural (CTR) by ~25%.

Anomaly on 3/12/08

Mean Gas and PM Concentrations

Species	Coastal sites				Inland sites			
	Nonurban		Urban		Nonurban		Urban	
	OAK ^a	OLF	GFP ^a	PNS ^b	CTR	YRK	BHM ^c	JST
PM_{2.5} (µg m⁻³)								
Mass	11.06	10.69	10.93	12.28	11.51	12.89	16.53	15.46
SO ₄	3.26	3.22	3.28	3.42	3.50	4.11	4.31	4.27
NO ₃	0.32	0.36	0.42	0.42	0.33	0.76	0.83	0.87
NH ₄	0.98	1.04	1.06	1.12	1.12	1.62	1.55	1.67
EC	0.50	0.60	0.62	0.78	0.58	0.57	1.76	1.34
OM	3.71	3.41	3.35	3.96	4.22	4.31	6.17	5.88
MMO	0.49	0.44	0.50	0.59	0.38	0.32	1.02	0.51
PM_{10-2.5} (µg m⁻³)								
Mass	6.10	6.59	8.08	9.06	5.25	5.23	14.38	8.28
SO ₄	0.21	0.26	0.30	0.29	0.19	0.21	0.34	0.24
NO ₃	0.45	0.56	0.72	0.64	0.37	0.37	0.48	0.41
NH ₄	0.03	0.03	0.03	0.03	0.04	0.05	0.02	0.03
MMO	1.58	1.41	2.02	2.39	1.33	1.34	4.91	3.03
Gases (ppbv)								
O ₃	34.81	32.95	32.54	29.12	35.18	37.68	24.66	24.47
CO	160.39	181.95	218.21	262.11	165.64	178.73	397.67	417.25
SO ₂	1.29	1.76	1.62	2.30	1.66	2.31	4.20	4.33
NO	0.15	0.86	2.24	3.59	0.29	0.46	13.52	21.21
NO ₂	1.36	4.22	5.68	7.29	2.23	3.8	15.34	18.84
HNO ₃	0.39	0.32	0.31	0.23	0.62	0.86	0.54	1.03
NO _y	2.64	6.49	9.76	11.72	4.00	6.12	30.66	42.75
NH ₃ ^d	0.31	0.47	0.76	0.82	0.32	2.67	2.53	1.40



Source Contributions to Atmospheric Gases and Particulate Matter in the Southeastern United States

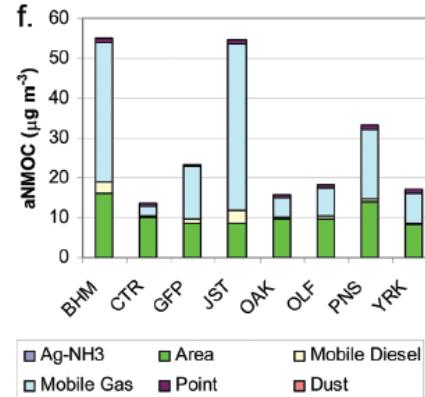
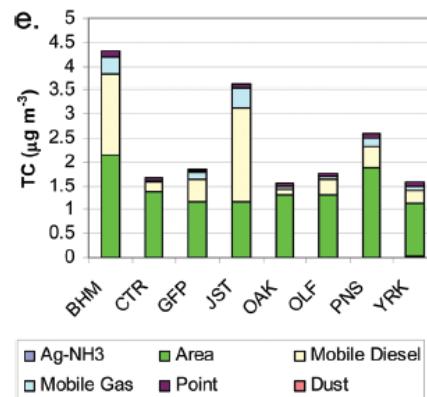
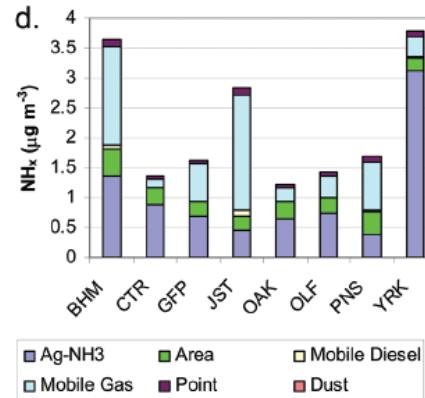
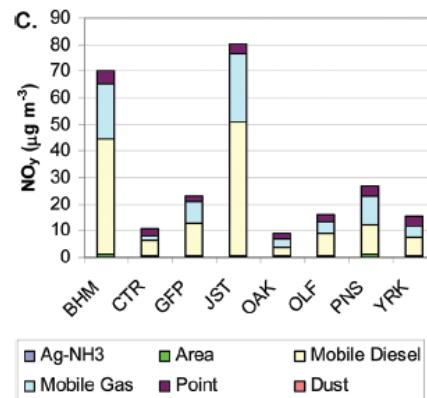
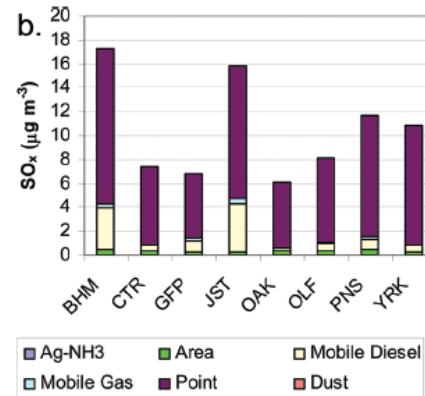
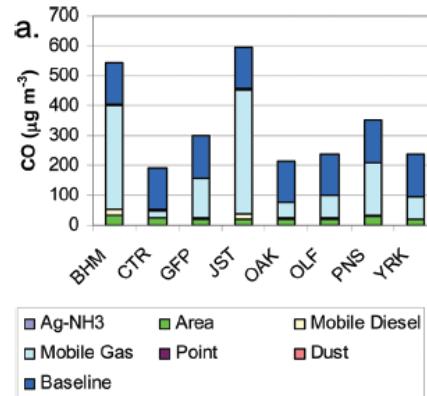
Charles L. Blanchard,* Shelley Tanenbaum, and George M. Hidy

- Chemical Mass Balance Approach

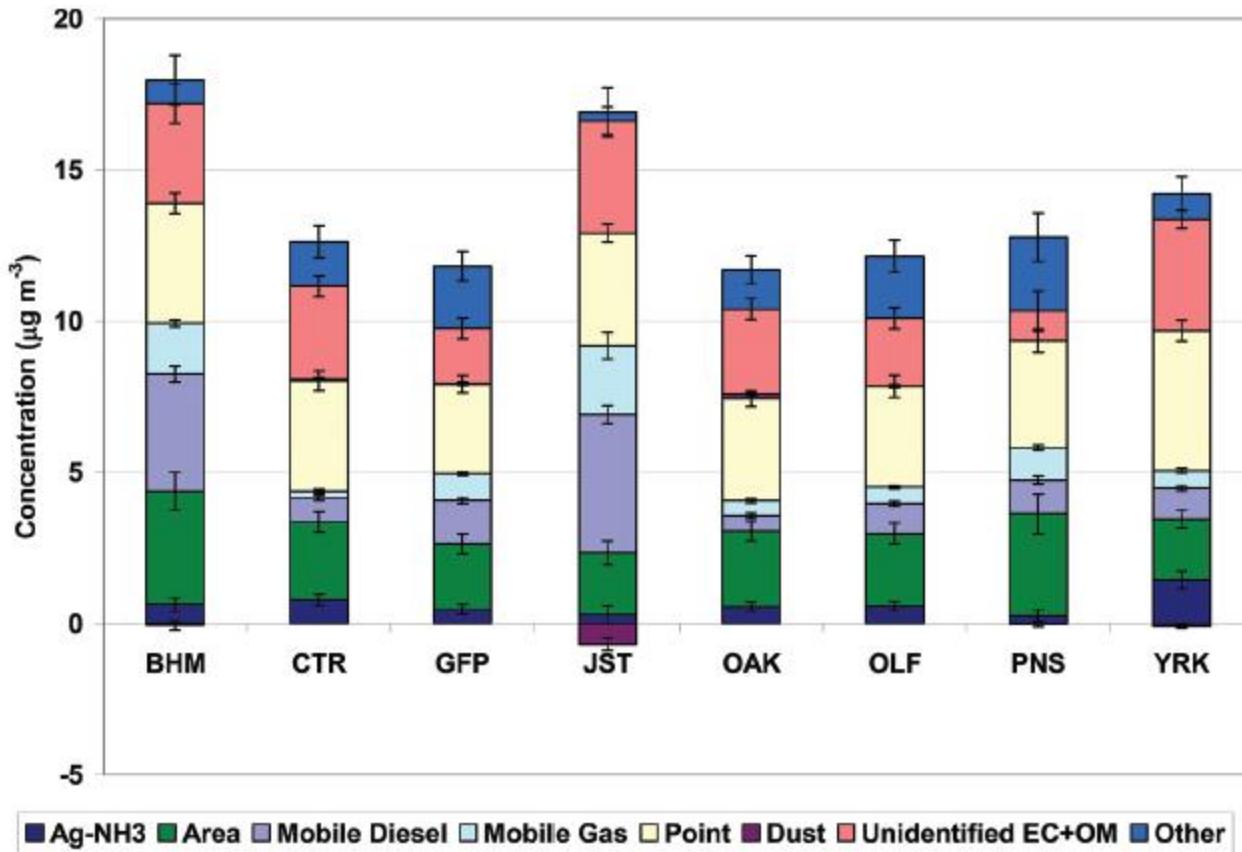
$$\begin{aligned} [\text{PM}_{2.5}]_i = & [\text{SO}_x]_i (\text{SO}_4/\text{SO}_x) + [\text{NO}_y]_i (\text{NO}_3/\text{NO}_y) \\ & + [\text{NH}_x]_i (\text{NH}_4/\text{NH}_x) + [\text{MMOx}]_i + [\text{K}_2\text{O}]_i \\ & + [\text{EC}]_i + 1.6 * [\text{OC}]_i \end{aligned}$$

- 2004-2007 SEARCH Data
- 2005 NEI

Source Contributions to Mean Concentrations



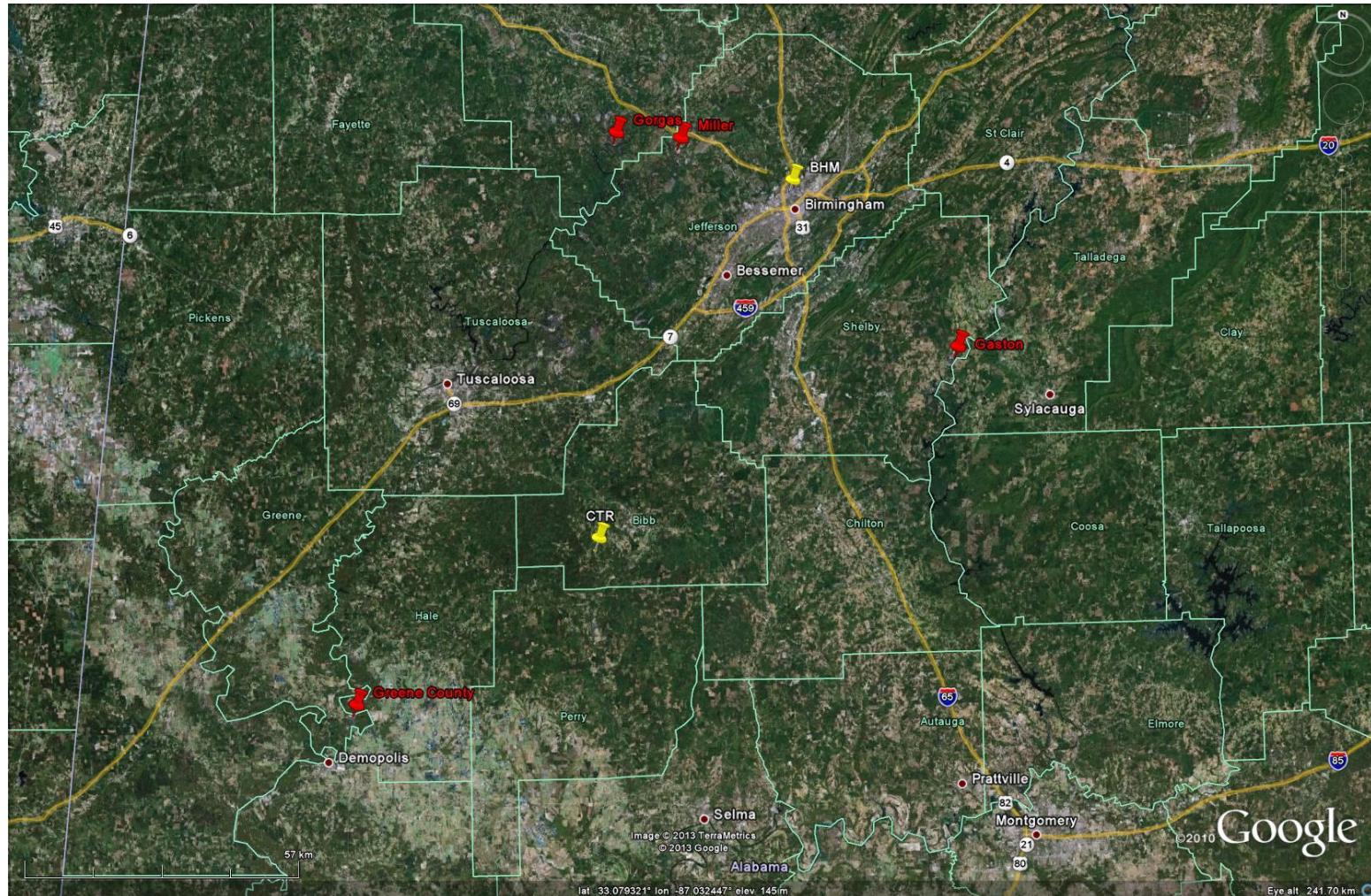
Source Contributions to PM_{2.5}



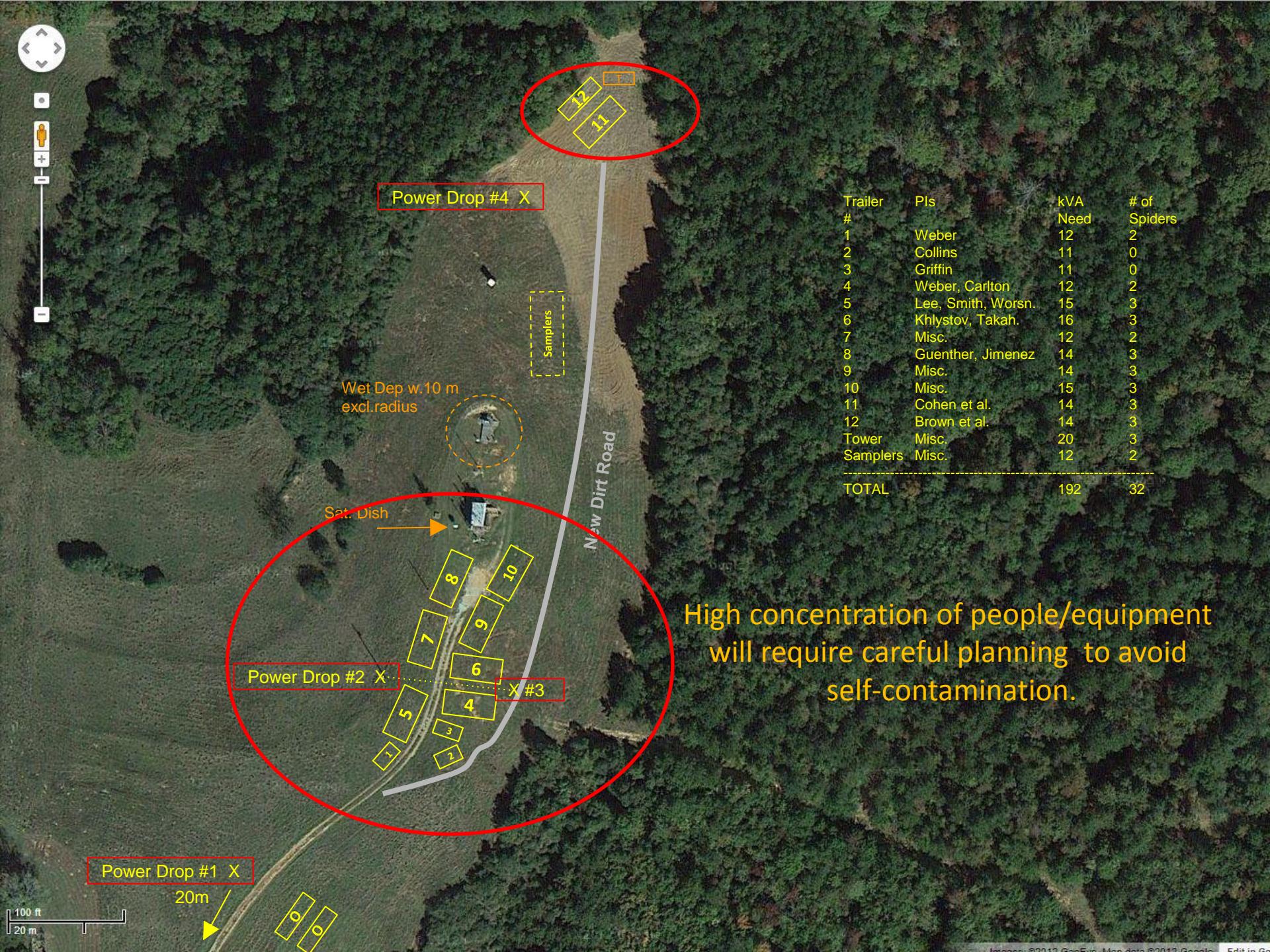
Multiple sources contribute to PM_{2.5} mass at all SEARCH sites.
Large unidentified EC+OM component likely dominated by SOA.

Recent Observations at CTR

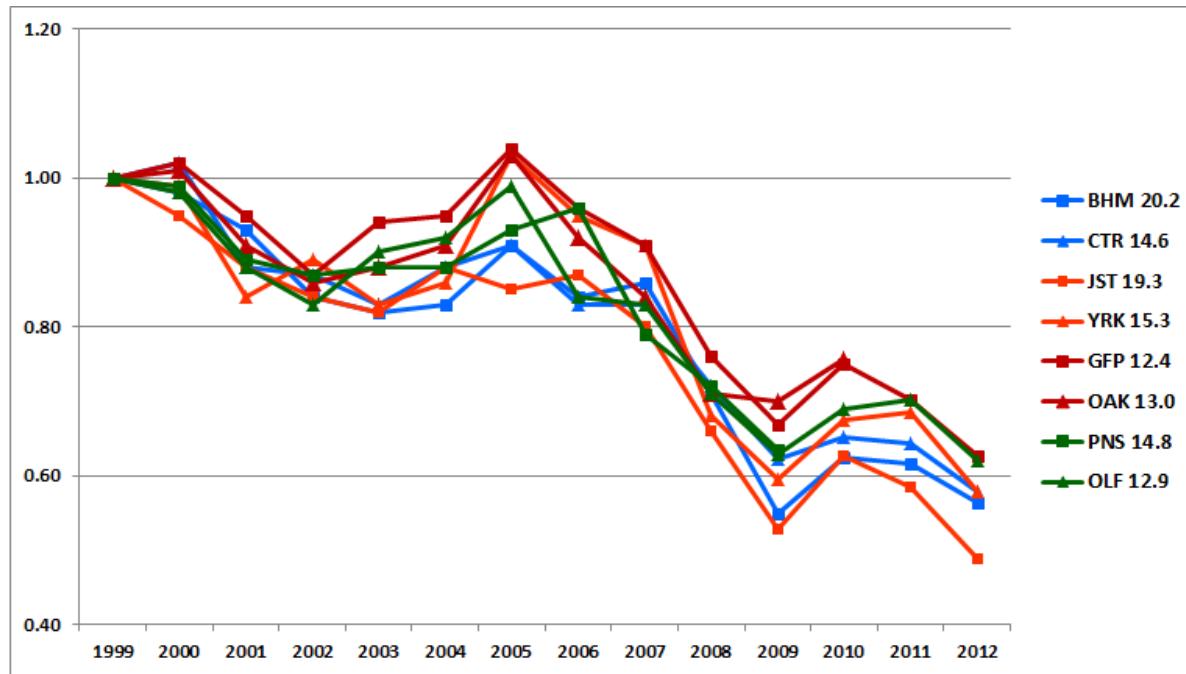
General Location of Centreville SEARCH Site



EGU	Size (MW)	D (km)	City	Population	D (km)	POI	D (km)
Green County	1,220	60	Tuscaloosa	222,000	45	Starbucks	51
Gaston	1,880	81	Birmingham	1,128,000	80	Fresh Market	68
Miller	2,640	82	Montgomery	375,000	105	Whole Foods	82
Gorgas	1,221	83					



The downward march of PM_{2.5} has continued since Blanchard et al. analysis



Daily PM_{2.5} Time Series at CTR 2005 (above) and 2012 (below)

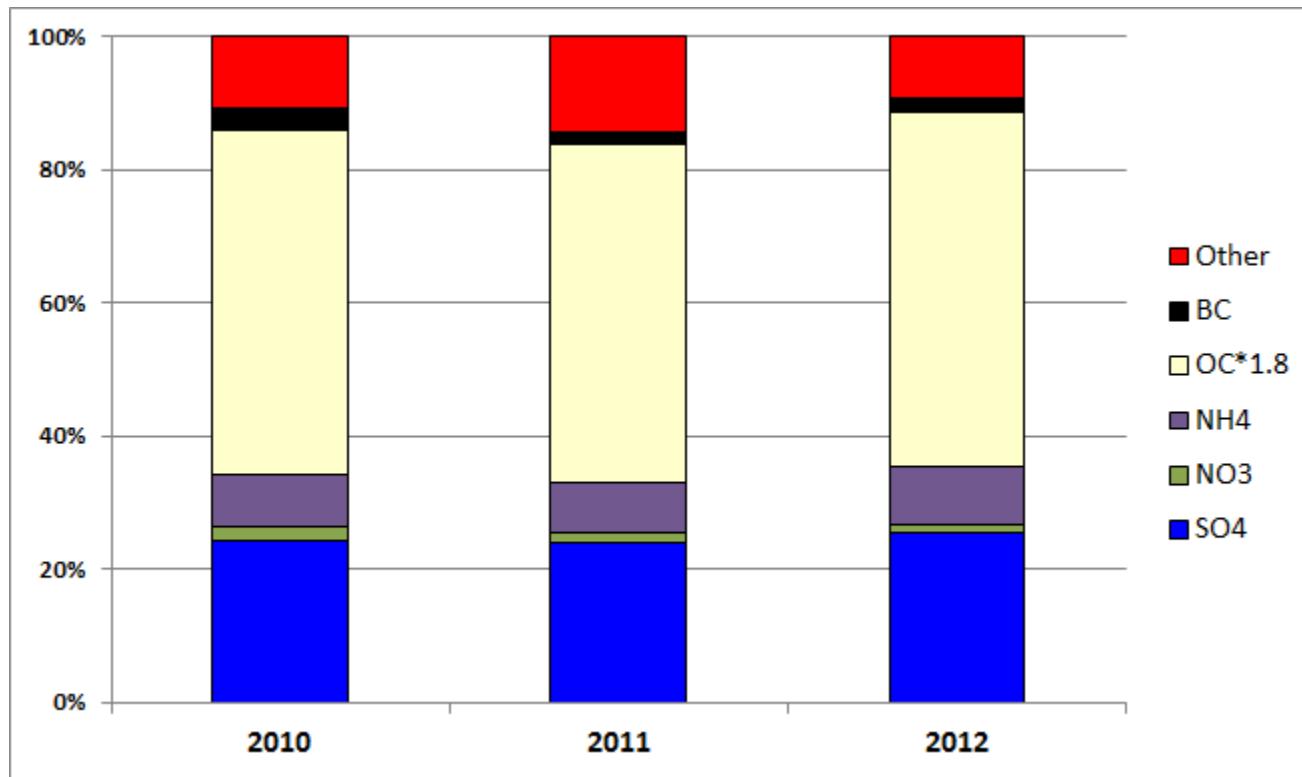


Major summertime excursions of PM_{2.5} mass have largely disappeared.

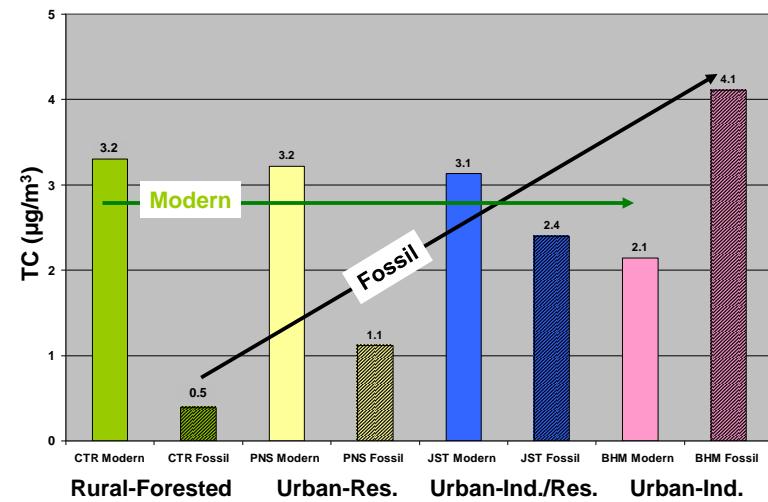
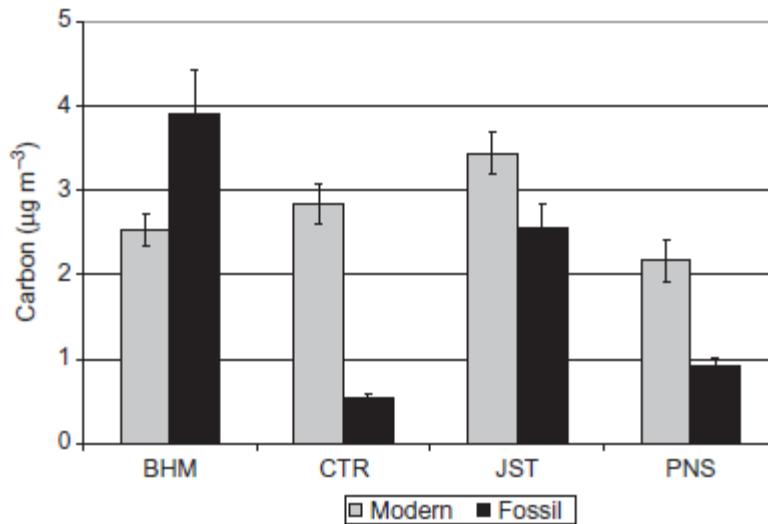
PM2.5 Mass and Components

June-July 2010-2012

Year	PM _{2.5} [ug/m ³]			SO ₄ [ug/m ³]			NO ₃ [ug/m ³]			NH ₄ [ug/m ³]			OC[ug/m ³]			BC [ug/m ³]		
	Max.	Mean	24-Hr.	Max.	Mean	24-Hr.	Max.	Mean	24-Hr.	Max.	Mean	24-Hr.	Max.	Mean	24-Hr.	Max.	Mean	
2010	10.79	18.2	34.3	2.60	4.8	12.7	0.23	0.5	1.6	0.87	2.4	4.9	3.09	5.4	5.8	0.38	0.6	1.6
2011	13.29	33.5	68.1	3.17	7.8	11.2	0.22	0.8	2.0	0.99	2.4	3.4	3.76	11.6	36.0	0.24	1.0	3.1
2012	9.28	23.9	31.1	2.44	6.1	9.8	0.14	0.4	1.0	0.82	2.6	3.8	2.84	6.9	10.8	0.22	0.4	0.9



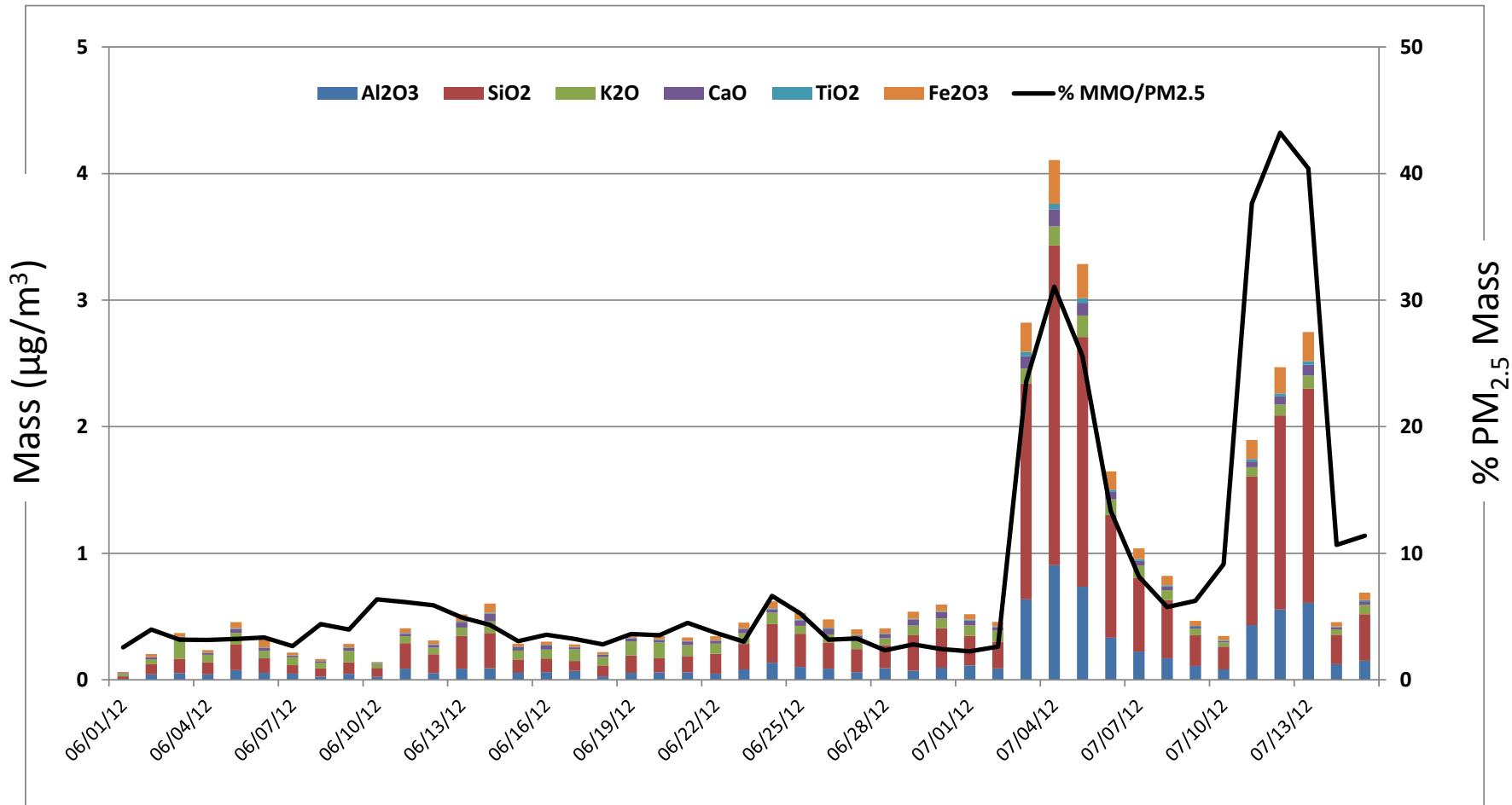
^{14}C Measurements: 2003-2005



Qtr	n	C-weighted		Modern	Fossil
		Fmodern	Fmodern	TC	TC
1	19	0.83	0.90	3.94	0.41
2	27	0.87	0.87	2.59	0.38
3	18	0.83	0.88	3.95	0.55
4	19	0.83	0.82	2.91	0.62
All	83	0.84	0.86	3.24	0.53

Regional TC is about 85% F_{modern} implying roughly $3 \mu\text{g}/\text{m}^3$ modern carbon at CTR and other sites.

North African Dust Excursions



N. African (mineral) dust events normally occur in August, but sometimes in July as in 2012.

Selected Trace Gases

June-July 2010-2012

Year	O3[ppb]			CO[ppb]			SO2[ppb]			NH3[ppb]			H2O [%]	
	Mean	Max	1-Hr	Max.	Mean	Max	1-Hr	Max. 5-min	Mean	Max	1-Hr	Max. 5-min	Mean	1-Hr
2010	30.6	76.7	79.2	141	248	336	0.62	9.7	18.6	n.a.	n.a.	n.a.	3.0	3.6
2011	32.6	91.0	92.9	167	886	1266	0.61	11.7	17.6	n.a.	n.a.	n.a.	2.8	3.7
2012	34.0	84.4	87.0	162	268	397	0.47	31.7	41.6	0.41	1.59	1.71	2.6	3.6

Average concentrations for O₃, CO, SO₂ and NH₃ are relatively low, but short-term maxima indicate interesting excursions.

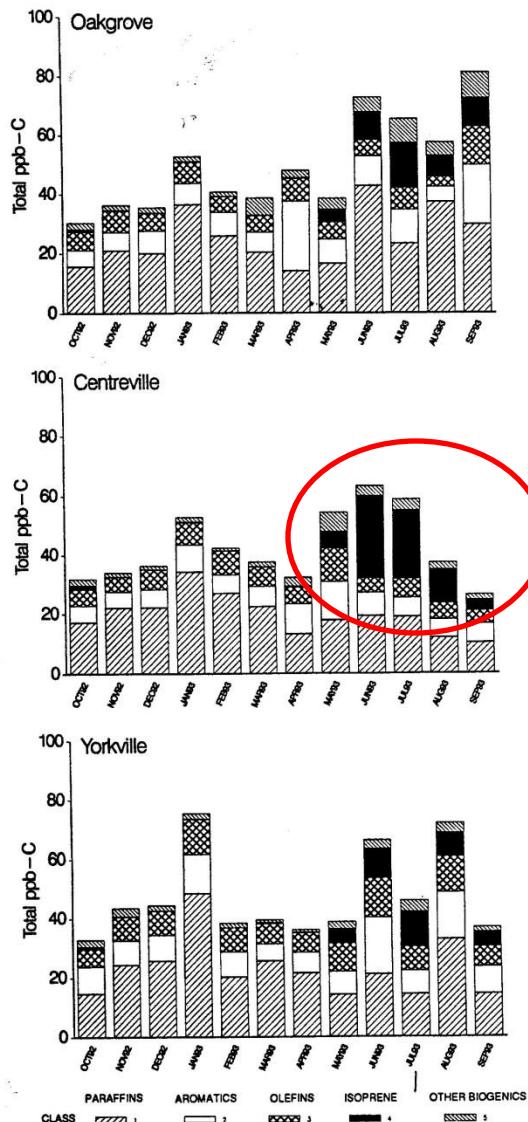
Water vapor, both a friend (reactant and medium) and enemy (condensation), is abundant.
Be prepared!

Dry(blue) and ambient (red) light scattering at 530 nm



Scattering data show lots of water associated with particles, even in winter.

VOC data at CTR are very limited, but indicate significant mid-afternoon isoprene in June and July



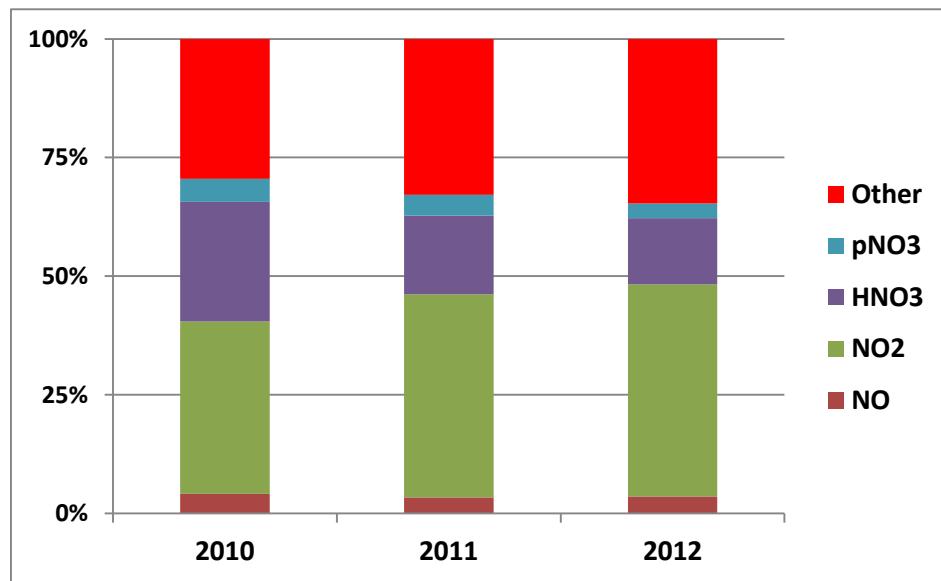
SOS-SCION data
October 1992-September 1993

Figure 5-1. Monthly Mean Hydrocarbon Concentration, by Class, for Three SCION Sites

NOy, NOx and NOz

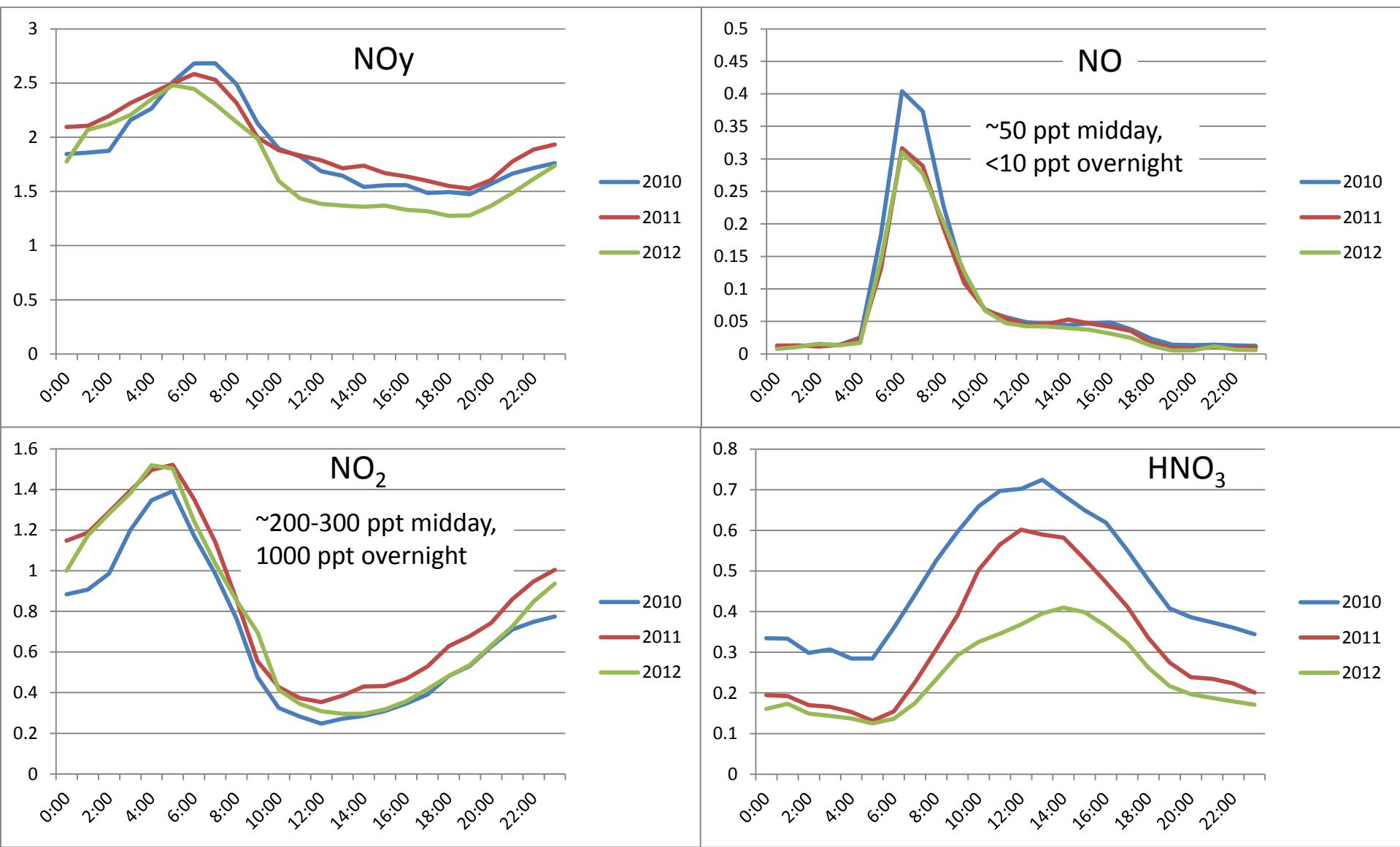
June-July 2010-2012

Year	NOy[ppb]			NO[ppb]			NO2[ppb]			HNO3[ppb]			pNO3[ppb]		
	Mean	Max	1-Hr	Max.	Mean	1-Hr	Max.	5-min	Mean	1-Hr	Max.	5-min	Mean	1-Hr	Max.
2010	1.89	8.9	11.8	0.08	2.2	3.0	0.69	6.2	7.3	0.48	1.8	3.0	0.09	0.7	1.0
2011	1.97	10.3	11.4	0.07	1.5	4.2	0.84	6.9	7.7	0.33	1.4	1.5	0.09	0.8	1.2
2012	1.74	16.8	23.5	0.06	1.8	3.1	0.78	9.8	15.4	0.24	1.3	1.4	0.05	0.4	0.8



Lots of highly oxidized NOy, in addition to HNO₃.

Diurnal behavior of NO_y, NO, NO₂ and HNO₃



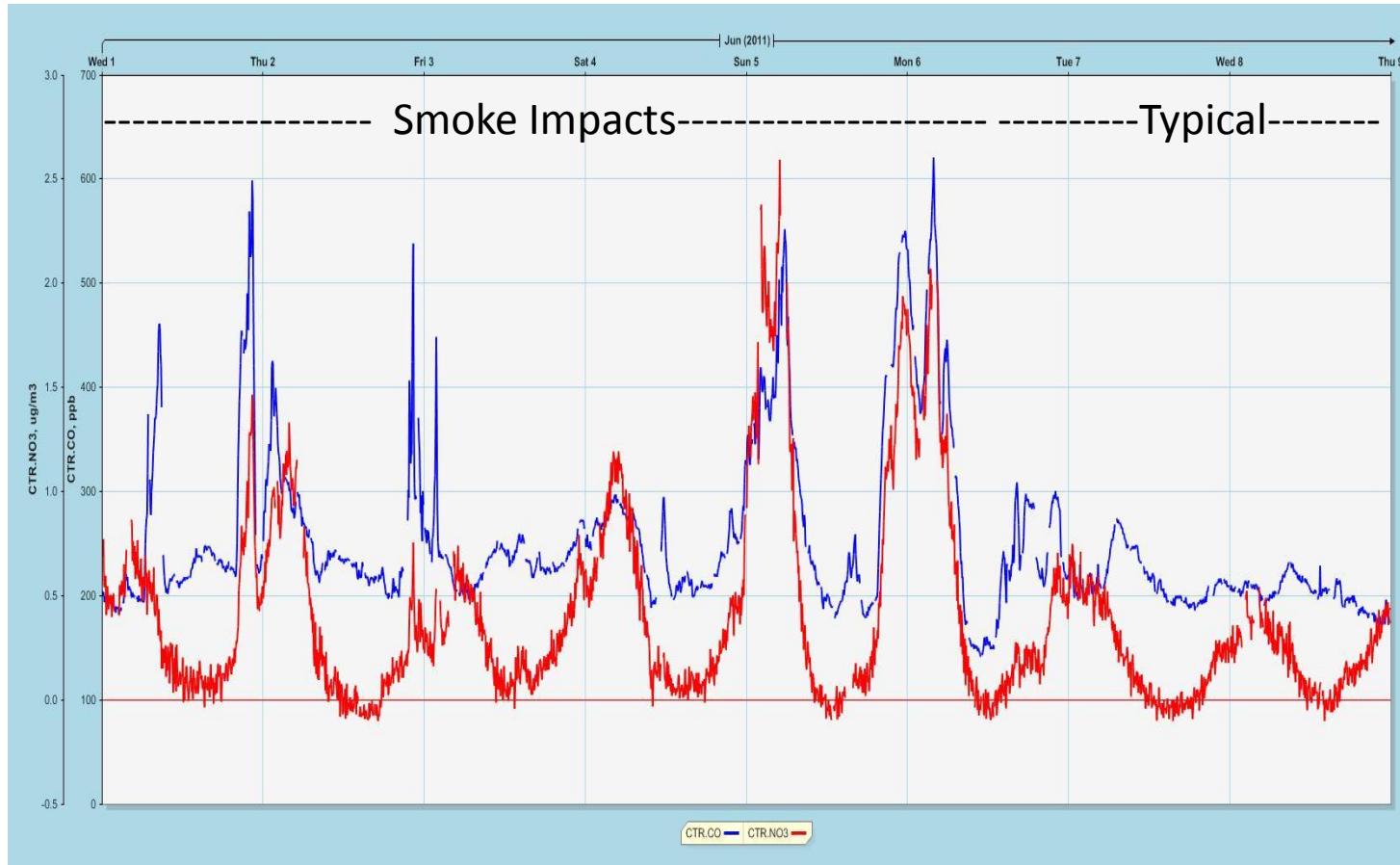
Typical HNO₃ Behavior



Strong daytime production, rapid removal by precipitation (green trace),
interesting overnight excursions.

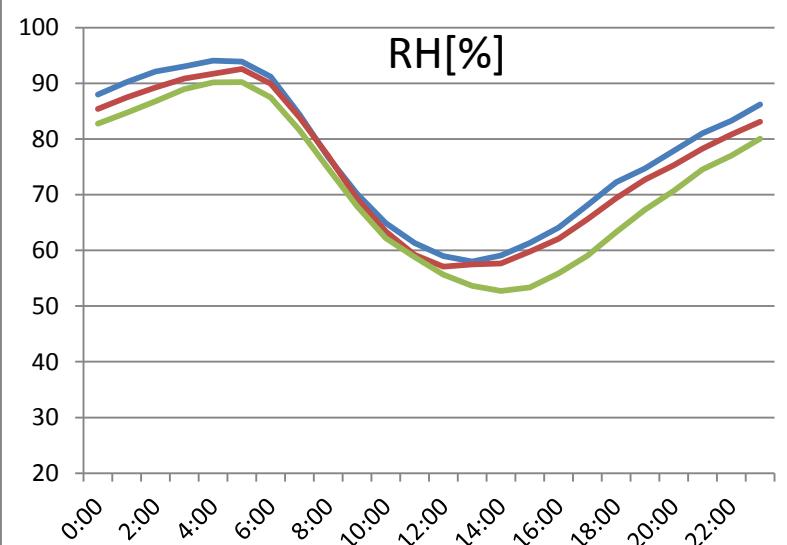
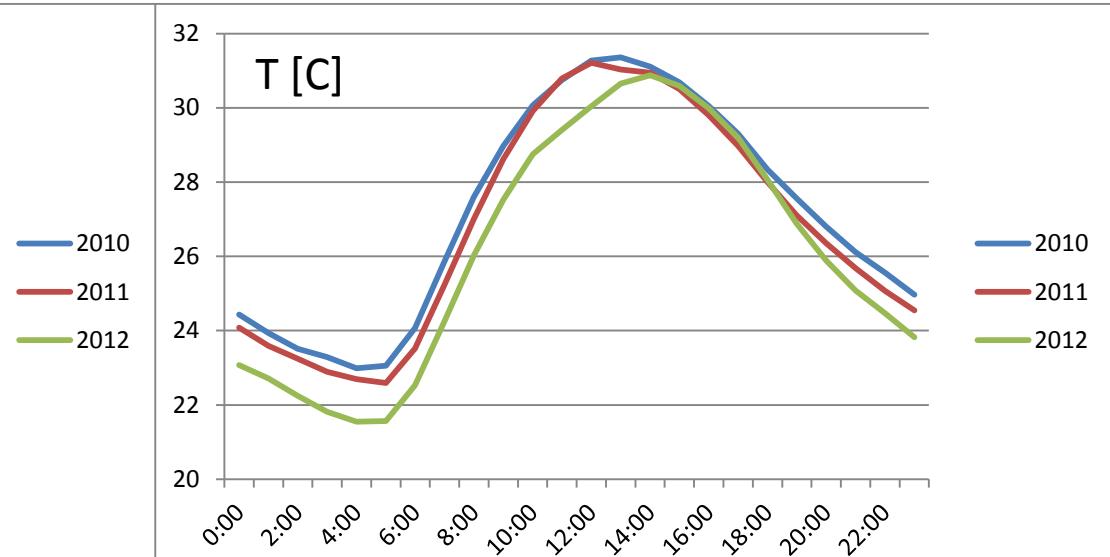
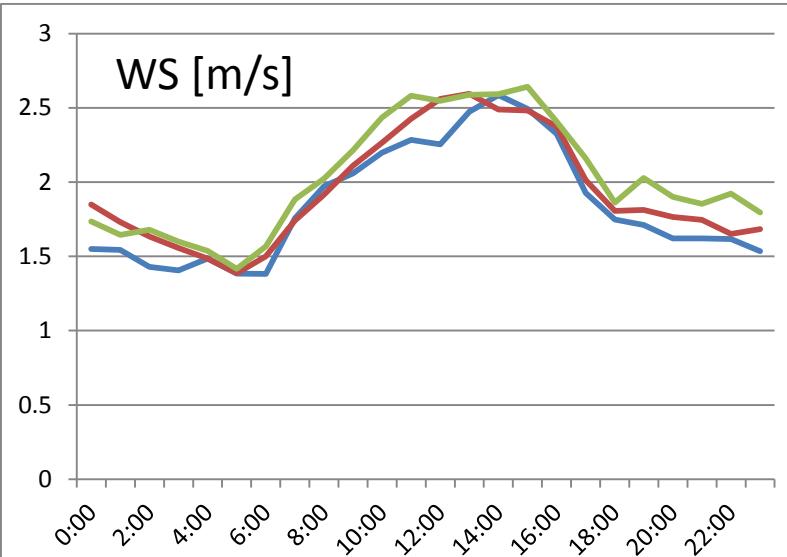
CO (blue) and pNO₃⁻ (red)

June 1-7, 2011



Particulate-nitrate (red) a minor component of NO_y, but always forms overnight.
Significant excursions occur, even in summer, when smoke impacted.

Meteorological Data for CTR



2012 was windier, cooler and drier than 2010, with 2011 in between.

Overnight winds (10m agl) rarely calm.

Bulk Formula for Fine OC



- Oxygen is the big player
- Multiple groups, both real-time and filter-based
- ARA to estimate org.-S and org.-O on 24-hour filters
- Contribute to understanding OM/OC (mass closure)
- Contribute to understanding f(RH) for OC (Bscat closure)

* Filterable water-soluble organic nitrogen in Fine particles over the southeastern USA during summer

Neeraj Rastogi, Xiaolu Zhang, Eric S. Edgerton, Ellery Ingall, Rodney J. Weber

Contribution of organosulfur compounds to organic aerosol mass

Michael P. Tolocka, and Barbara J. Turpin