

Supplementary Material

Table S1. Number of observation data samples (N) and arithmetic means of the observed (Obs) and modeled (Mod) values.

		2002			2005				2006			
		N	Obs	Mod ¹	N	Obs	Mod ²	Mod ³	N	Obs	Mod ²	Mod ³
Concentrations (CASTNET) ⁴												
SO ₂	Native	4074	1.8	2.2	4114	1.9	2.1	2.8	4183	1.6	2.1	2.8
	Annual	82	1.8	2.2	84	1.9	2.0	2.8	85	1.6	2.1	2.8
SO ₄	Native	4075	0.99	0.82	4114	1.1	0.76	1.0	4183	0.90	0.73	0.99
	Annual	82	1.0	0.82	84	1.0	0.76	1.0	85	0.88	0.73	0.99
HNO ₃	Native	4075	0.32	0.36	4114	0.30	0.25	0.28	4183	0.26	0.25	0.27
	Annual	82	0.32	0.36	84	0.30	0.24	0.27	85	0.26	0.24	0.27
NO ₃	Native	4075	0.20	0.24	4114	0.19	0.28	0.21	4183	0.17	0.24	0.18
	Annual	82	0.20	0.25	84	0.19	0.28	0.21	85	0.17	0.25	0.19
Dry deposition (CASTNET) ⁴												
SO ₂	Native	3828	0.036	0.058	3715	0.038	0.076	0.066	3814	0.032	0.070	0.063
	Annual	70	2.0	3.1	64	1.9	3.9	3.3	69	1.6	3.4	3.1
SO ₄	Native	3829	0.0072	0.0025	3715	0.0076	0.013	0.010	3814	0.0065	0.013	0.0097
	Annual	70	0.38	0.13	64	0.38	0.71	0.52	69	0.33	0.64	0.49
HNO ₃	Native	3829	0.026	0.040	3715	0.024	0.034	0.046	3814	0.021	0.031	0.044
	Annual	70	1.4	2.1	64	1.2	1.7	2.4	69	1.1	1.6	2.2
NO ₃	Native	3829	0.0013	0.0004	3715	0.0012	0.0027	0.0018	3814	0.0011	0.0025	0.0015
	Annual	70	0.071	0.024	64	0.061	0.14	0.091	69	0.058	0.13	0.081
NH ₄	Native	3824	0.0058	0.0016	3715	0.0061	0.0068	0.0064	3814	0.0050	0.0064	0.0059
	Annual	69	0.30	0.086	64	0.29	0.35	0.32	69	0.25	0.33	0.30
Wet deposition (NADP) ⁴												
Total SO ₄	Native	9683	0.068	0.074	8343	0.085	0.091	0.075	8087	0.088	0.096	0.080
	Annual	242	3.1	3.7	252	3.3	3.9	3.2	249	3.3	3.9	3.2
Total NO ₃	Native	9682	0.041	0.030	8343	0.045	0.035	0.020	8087	0.046	0.032	0.019
	Annual	242	1.9	1.5	252	1.7	1.5	0.86	249	1.7	1.3	0.78
Total NH ₄	Native	9433	0.038	0.029	8343	0.045	0.038	0.024	8087	0.047	0.041	0.025
	Annual	242	1.7	1.4	252	1.7	1.6	1.0	249	1.8	1.7	1.0
Concentrations (AQS) ⁴												
NO _y	Native	97143	9.0	8.1	14833 8	7.0	-	5.7	171622	6.1	5.1	5.2
	Annual	20	7.9	8.7	32	5.8	-	6.1	32	5.8	5.7	5.7
Concentrations (SEARCH) ⁴												
NO _y	Native	57362	9.2	6.0	64456	8.3	-	5.0	65054	7.9	4.9	4.8
	Annual	8	9.1	5.9	8	8.2	-	5.0	8	7.8	4.9	4.8

¹ VISTAS CMAQ modeling; ² UBAQS CMAQ modeling; ³ General CAMx modeling; ⁴ Units for the mean values:
 Concentrations—µg-S/m³ (for sulfur compound) or µg-N/m³ (for nitrogen compound).
 Deposition (native)—kg-S/ha/week (for sulfur compound) or kg-N/ha/week (for nitrogen compound).
 Deposition (annual)—kg-S/ha/year (for sulfur compound) or kg-N/ha/year (for nitrogen compound).

Table S2. Mean bias (MB) and mean error (ME) ¹.

		VISTAS 2002		UBAQS 2005		UBAQS 2006		General 2005		General 2006	
		MB	ME	MB	ME	MB	ME	MB	ME	MB	ME
Concentrations (CASTNET)											
SO ₂	Native	0.39	0.71	0.14	0.76	0.56	0.82	0.84	1.0	1.2	1.3
	Annual	0.42	0.55	0.10	0.40	0.57	0.65	0.82	0.87	1.2	1.2
SO ₄	Native	−0.17	0.24	−0.30	0.34	−0.16	0.26	−0.025	0.35	0.098	0.31
	Annual	−0.18	0.18	−0.27	0.28	−0.15	0.17	0.0088	0.14	0.11	0.20
HNO ₃	Native	0.035	0.14	−0.058	0.13	−0.015	0.11	−0.029	0.11	0.0058	0.10
	Annual	0.034	0.087	−0.057	0.089	−0.016	0.068	−0.028	0.064	0.0063	0.059
NO ₃	Native	0.037	0.17	0.092	0.18	0.074	0.16	0.025	0.15	0.016	0.13
	Annual	0.042	0.13	0.088	0.13	0.082	0.12	0.022	0.10	0.022	0.089
Dry deposition (CASTNET)											
SO ₂	Native	0.022	0.032	0.038	0.043	0.037	0.042	0.028	0.035	0.031	0.036
	Annual	1.1	1.3	2.0	2.0	1.8	1.8	1.5	1.6	1.5	1.6
SO ₄	Native	−0.0047	0.0052	0.0058	0.0084	0.0061	0.0081	0.0025	0.0047	0.0031	0.0046
	Annual	−0.25	0.26	0.33	0.35	0.32	0.34	0.14	0.17	0.16	0.19
HNO ₃	Native	0.014	0.019	0.0095	0.016	0.010	0.015	0.022	0.025	0.022	0.025
	Annual	0.71	0.79	0.53	0.60	0.54	0.63	1.2	1.2	1.2	1.2
NO ₃	Native	−0.0009	0.0011	0.0015	0.0022	0.0014	0.0021	0.0006	0.0014	0.0004	0.0012
	Annual	−0.046	0.050	0.077	0.087	0.074	0.083	0.031	0.051	0.023	0.043
NH ₄	Native	−0.0042	0.0044	0.0007	0.0042	0.0014	0.0036	0.0003	0.0032	0.0009	0.0027
	Annual	−0.22	0.22	0.057	0.12	0.081	0.12	0.030	0.086	0.052	0.084
Wet deposition (NADP)											
Total SO ₄	Native	0.0059	0.049	0.0059	0.062	0.0081	0.061	−0.011	0.064	−0.008	0.066
	Annual	0.59	1.2	0.64	1.1	0.64	1.0	−0.11	0.97	−0.048	0.92
Total NO ₃	Native	−0.011	0.026	−0.010	0.028	−0.013	0.029	−0.024	0.029	−0.026	0.030
	Annual	−0.39	0.62	−0.23	0.43	−0.37	0.47	−0.87	0.91	−0.93	0.94
Total NH ₄	Native	−0.0085	0.026	−0.0068	0.032	−0.0063	0.033	−0.021	0.032	−0.022	0.033
	Annual	−0.25	0.66	−0.10	0.53	−0.054	0.47	−0.73	0.79	−0.74	0.80
Concentrations (AQS)											
NO _y	Native	−0.89	5.4	-	-	−1.0	3.8	−1.3	4.2	−0.96	3.7
	Annual	0.79	3.4	-	-	−0.13	2.1	0.27	2.6	−0.12	2.2
Concentrations (SEARCH)											
NO _y	Native	−3.2	5.8	-	-	−3.0	4.9	−3.3	5.3	−3.1	5.1
	Annual	−3.2	4.1	-	-	−2.9	3.6	−3.2	4.1	−3.0	3.9

$$^1 \text{MB} = \frac{1}{N} \sum (\text{Model} - \text{Observation}) \quad \text{ME} = \frac{1}{N} \sum |\text{Model} - \text{Observation}|$$

where N is the number of pairs of modeled and observed quantities matched in time and space. The unit of these statistics follows that of sample data (concentrations or deposition—see footnote of Table S1).

Table S3. Fractional bias (FB) and fractional error (FE) ¹.

		VISTAS 2002		UBAQS 2005		UBAQS 2006		General 2005		General 2006	
		FB	FE	FB	FE	FB	FE	FB	FE	FB	FE
Concentrations (CASTNET)											
SO ₂	Native	14	43	3.1	44	19	49	32	49	45	59
	Annual	17	32	2.3	26	19	35	28	39	42	51
SO ₄	Native	−25	34	−33	40	−25	37	−3.1	39	5.1	38
	Annual	−26	27	−37	37	−28	29	−6.6	20	1.9	25
HNO ₃	Native	8.9	44	−27	53	−15	48	−9.6	43	0.53	41
	Annual	11	28	−19	33	−8.2	29	−9.0	26	0.7	25
NO ₃	Native	−48	113	0.86	100	0.98	98	−28	99	−23	96
	Annual	−21	92	46	63	44	63	14	60	12	58
Dry deposition (CASTNET)											
SO ₂	Native	49	69	61	76	64	79	68	78	70	82
	Annual	53	59	67	69	65	72	68	71	67	75
SO ₄	Native	−84	101	48	79	52	79	37	62	42	64
	Annual	−93	98	54	62	57	66	26	40	30	47
HNO ₃	Native	42	61	26	62	31	63	64	74	69	79
	Annual	39	46	36	44	38	47	67	67	69	72
NO ₃	Native	−123	141	11	119	9.9	119	−0.19	102	−2.7	103
	Annual	−105	113	85	97	80	91	45	79	35	74
NH ₄	Native	−104	113	12	70	19	69	7.3	59	16	58
	Annual	−115	115	16	42	23	47	2.0	34	9.0	36
Wet deposition (NADP)											
Total SO ₄	Native	11	85	4.2	81	−1.2	79	−37	96	−46	97
	Annual	4.1	37	5.2	37	6.9	38	−14	40	−13	41
Total NO ₃	Native	−12	88	−27	79	−35	81	−79	98	−87	103
	Annual	−31	45	−22	36	−29	41	−71	76	−77	81
Total NH ₄	Native	−9.2	93	0.93	86	−2.7	85	−56	102	−64	104
	Annual	−34	57	−12	41	−8.4	39	−57	65	−58	67
Concentrations (AQS)											
NO _y	Native	23	57	-	-	12	57	17	57	12	56
	Annual	25	42	-	-	11	39	21	41	11	38
Concentrations (SEARCH)											
NO _y	Native	0.82	59	-	-	3.5	58	−1.6	61	2.1	62
	Annual	−24	50	-	-	−19	47	−22	52	−18	52

$$^1 \text{FB (\%)} = \frac{2}{N} \sum \frac{\text{Model}-\text{Observation}}{\text{Model}+\text{Observation}} \times 100 \quad \text{FE (\%)} = \frac{2}{N} \sum \frac{|\text{Model}-\text{Observation}|}{\text{Model}+\text{Observation}} \times 100$$

where N is the number of pairs of modeled and observed quantities matched in time and space.

Figure S1. Scatter plots of hourly average NO_Y concentrations ($\mu\text{g-N}/\text{m}^3$) between the VISTAS 2002 CMAQ simulation and SEARCH measurements at each of the monitoring sites: BHM, GFP, JST and PNS are urban sites; CTR, OAK and YRK are rural sites; OLF is a suburban site).

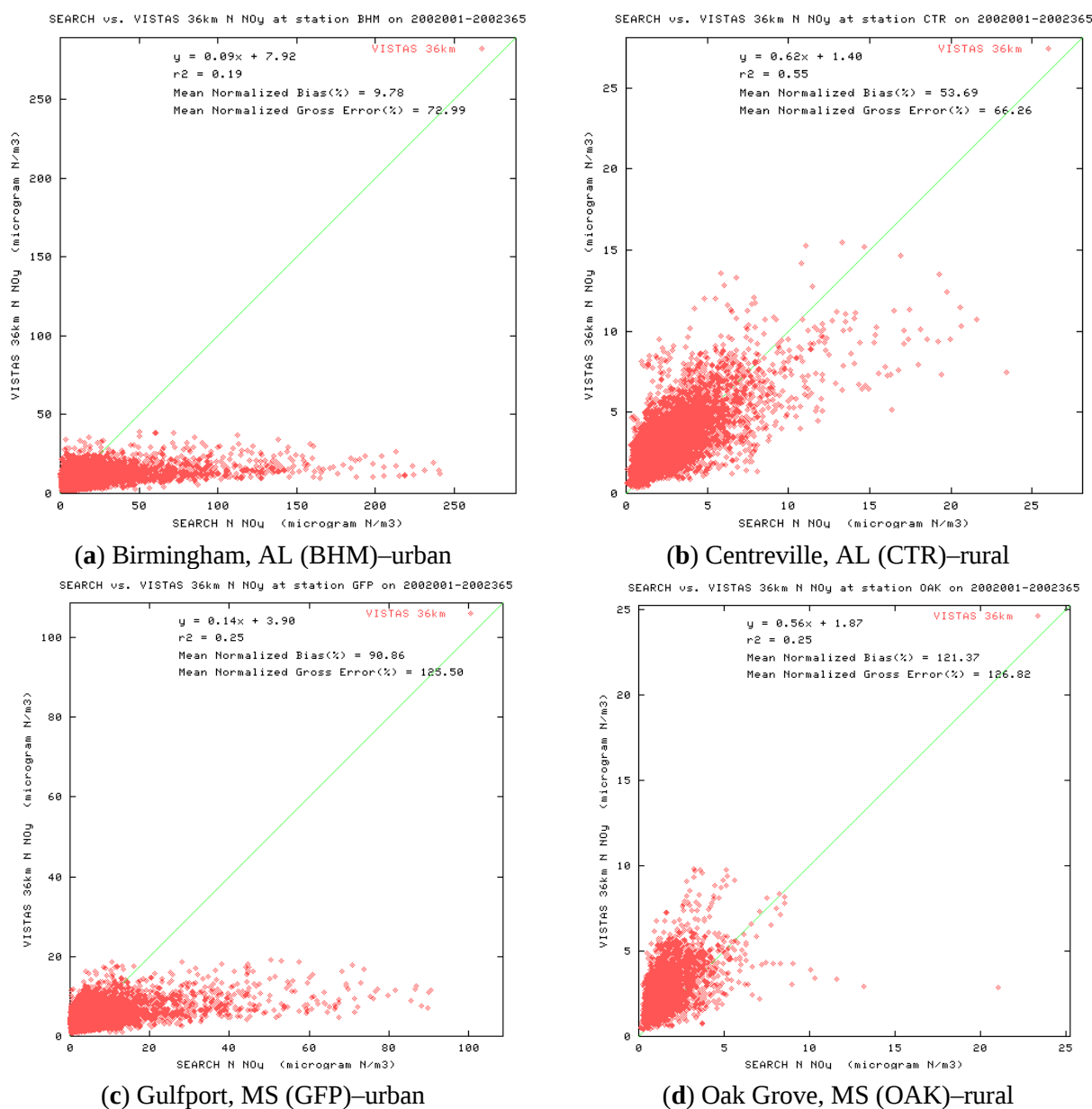
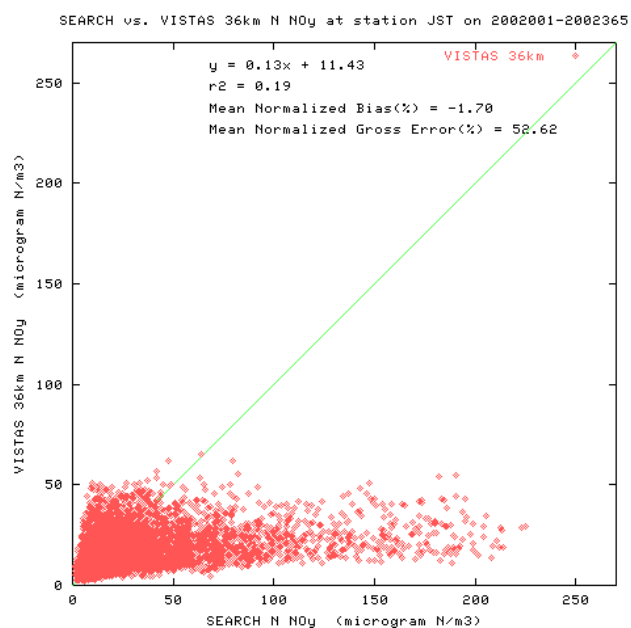
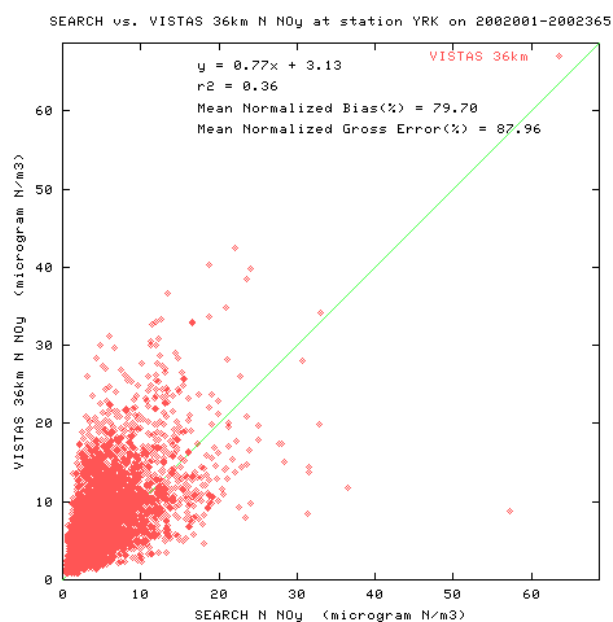


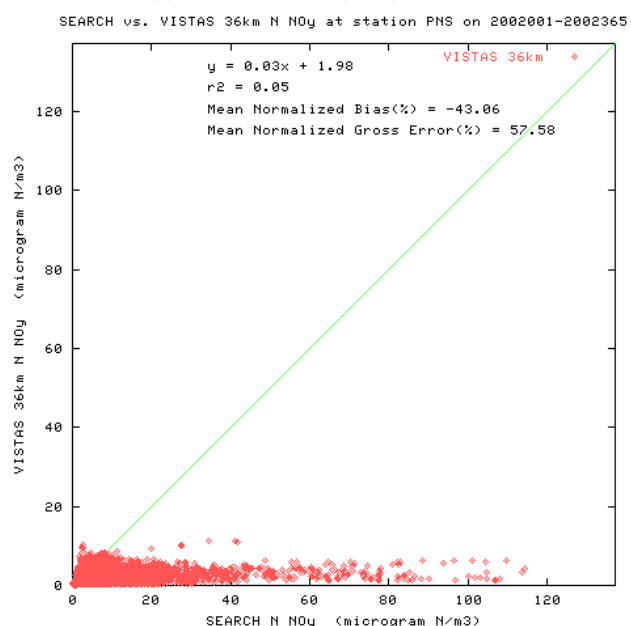
Figure S1. Cont.



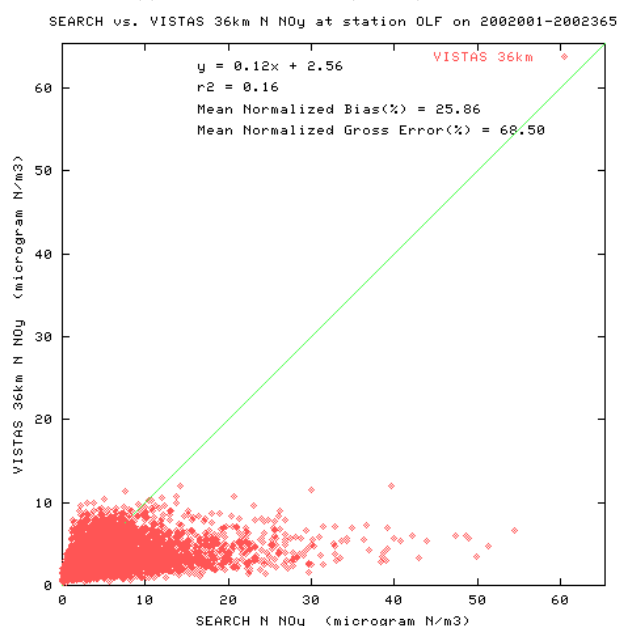
(e) Atlanta, GA (JST)—urban



(f) Yorkville, GA (YRK)—rural



(g) Pensacola, FL (PNS)—urban



(h) Outlying Landing Field, FL (OLF)—suburban