Supplement of

Air quality in the eastern United States and Eastern Canada for 1990-2015: 25 years of change in response to emission reductions of SO$_2$ and NOx in the region

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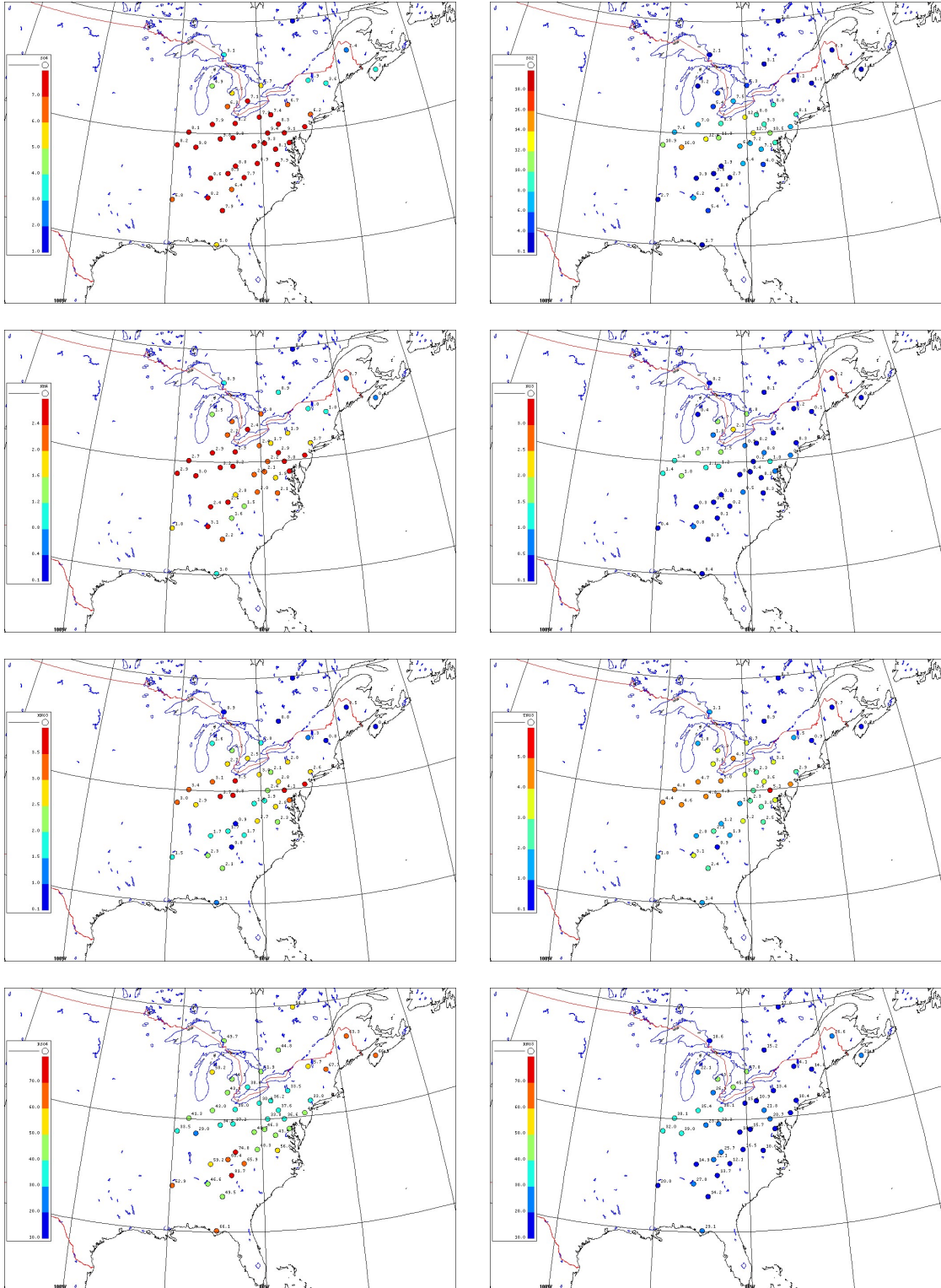
(a) Warm season

(b) Cold season

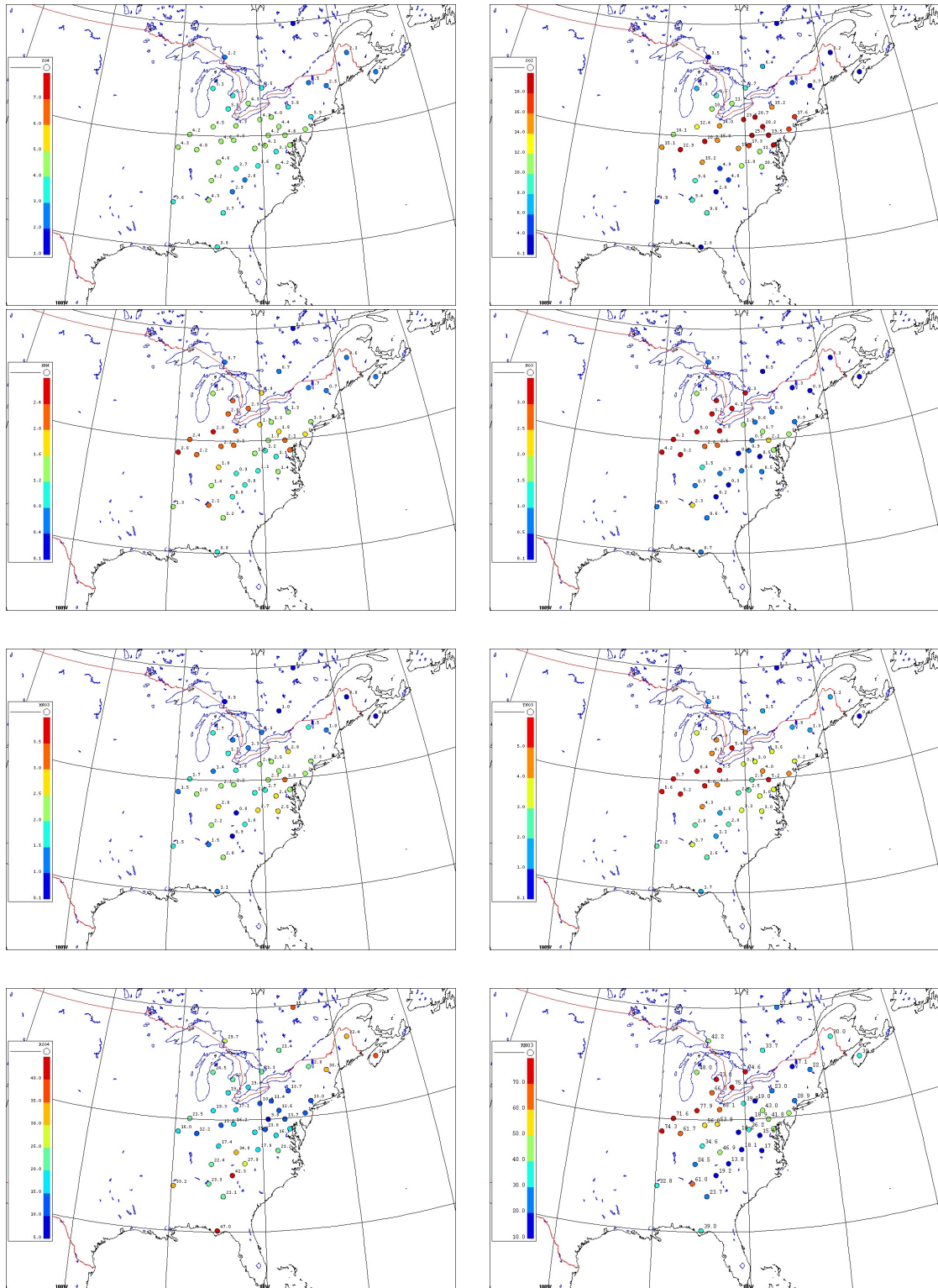
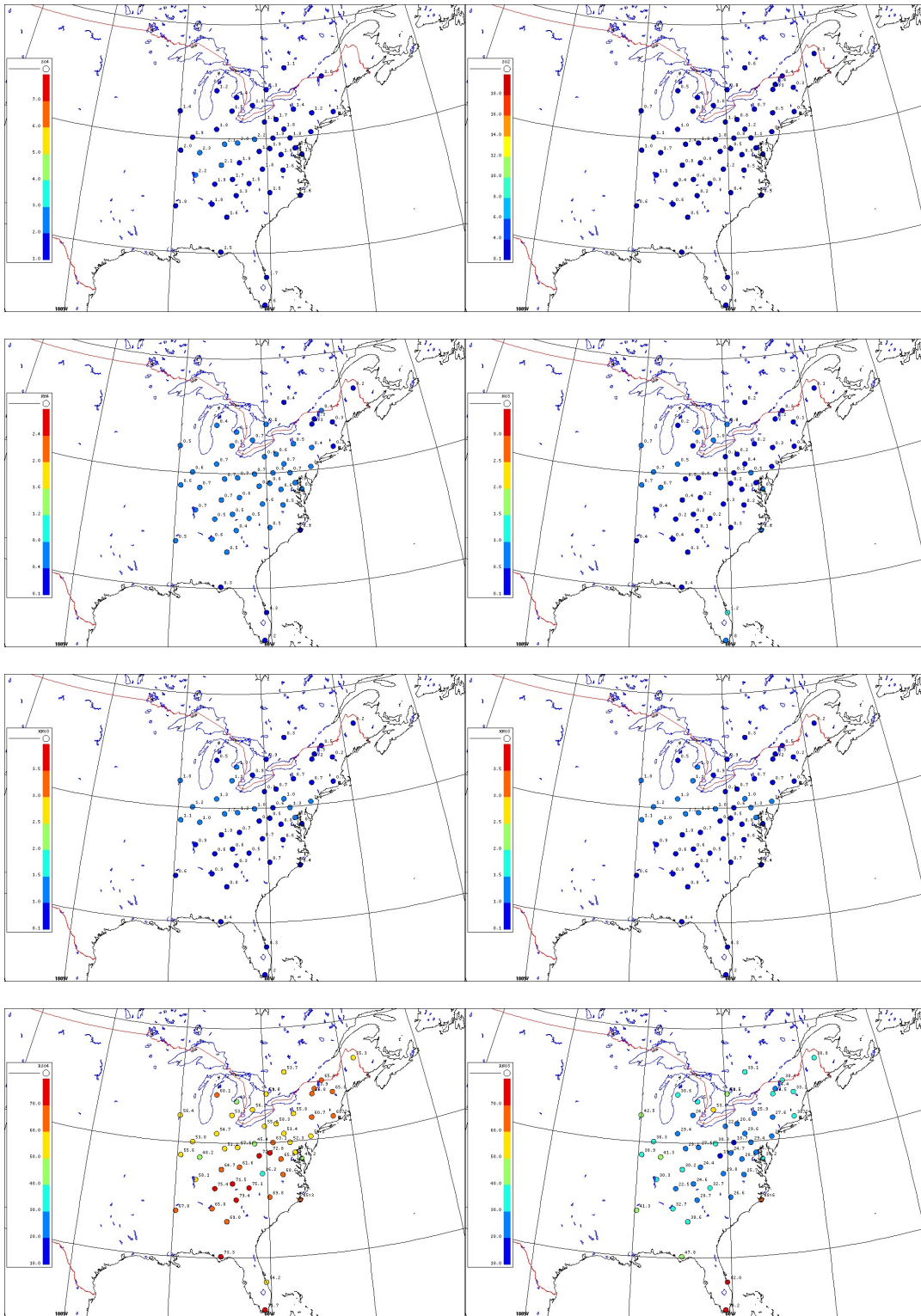



Fig. S1 Mean air concentrations (μg m^{-3}) of SO_2, SO_4^{2-}, NO_3^-, HNO_3, TNO_3 and NH_4^+, and mean ratios of RSO_4 (%) and RNO_3 (%) during 1989-1991 for (a) warm season and (b) cold season.

(a) Warm season

(b) Cold season

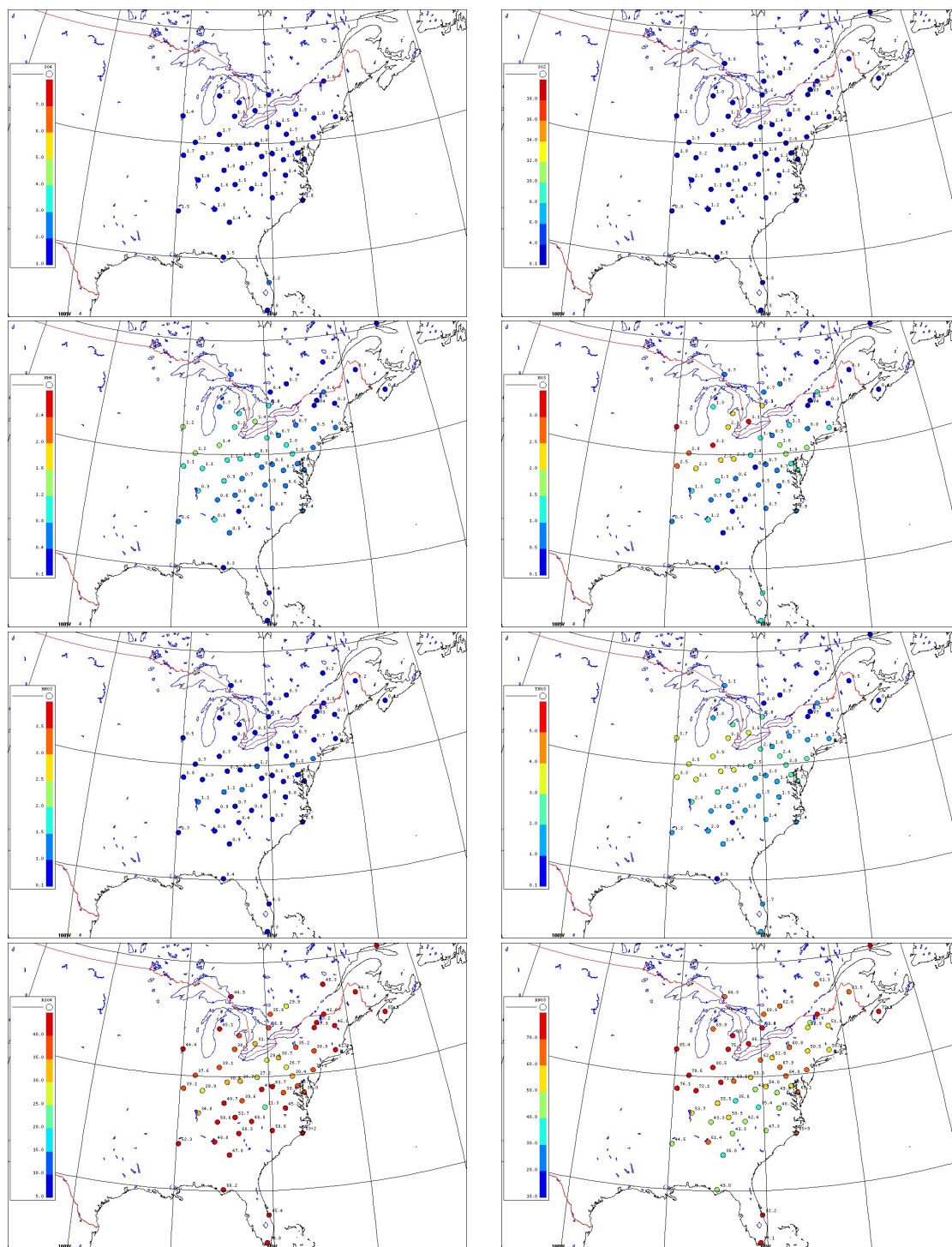
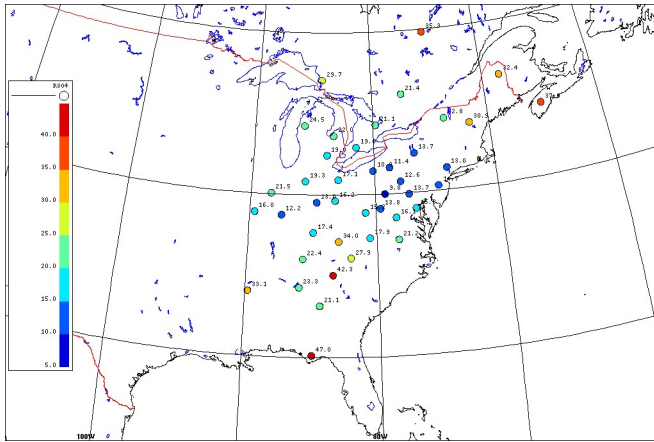
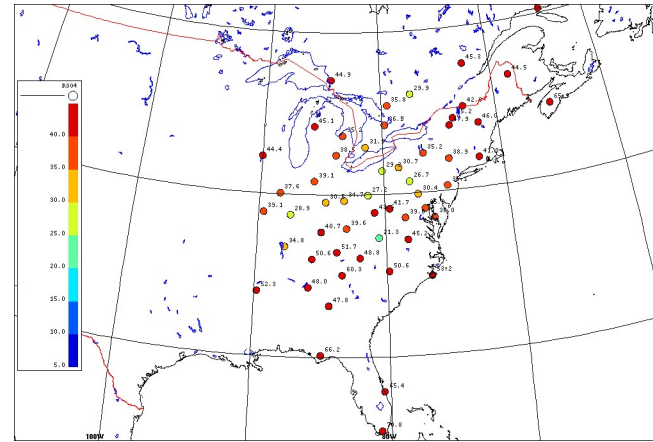
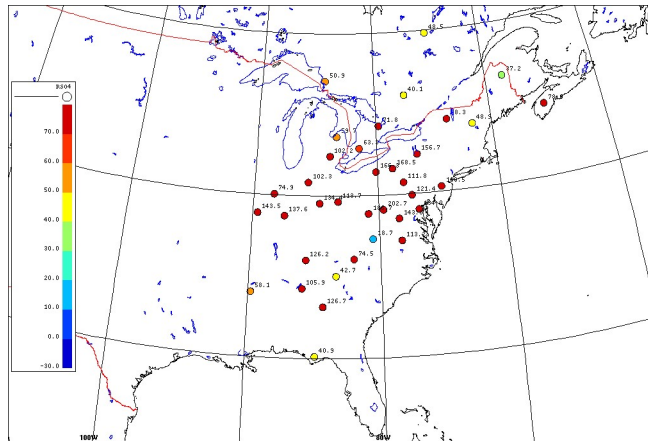
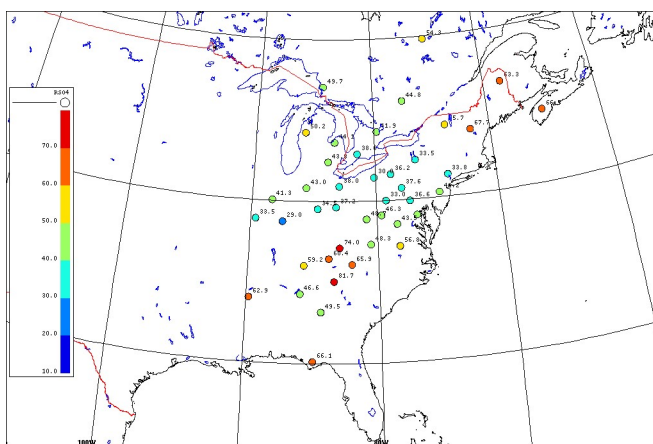
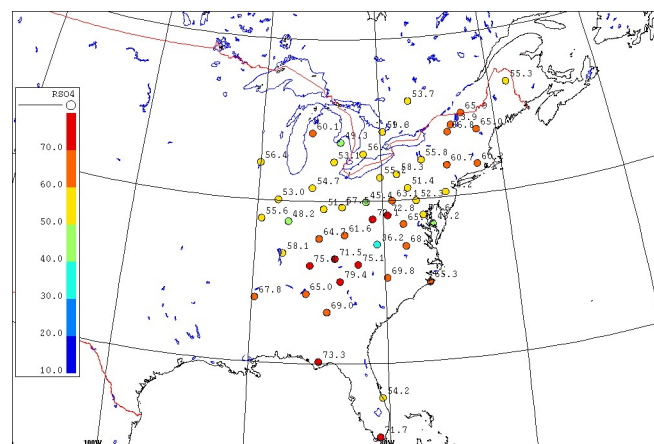
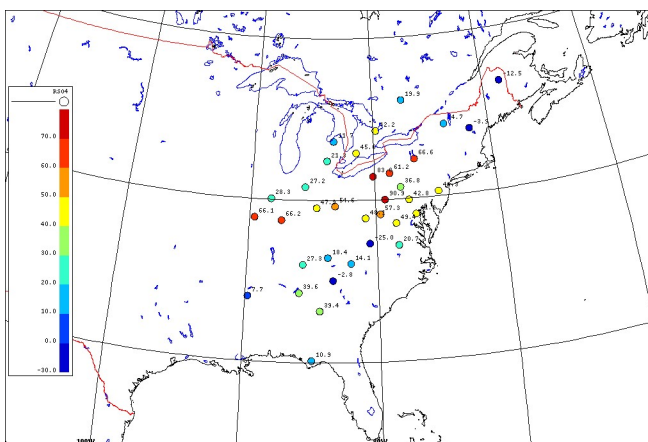
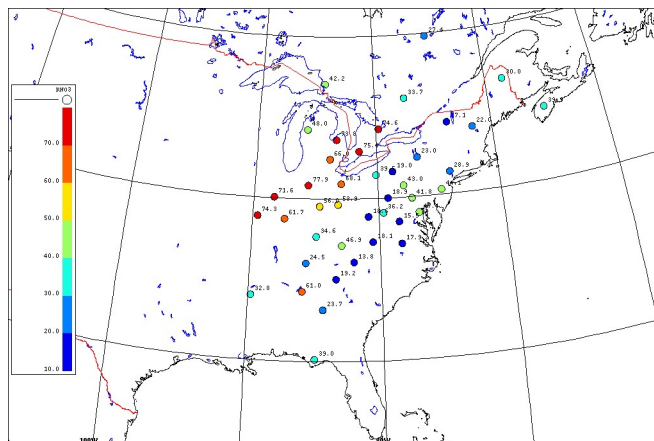
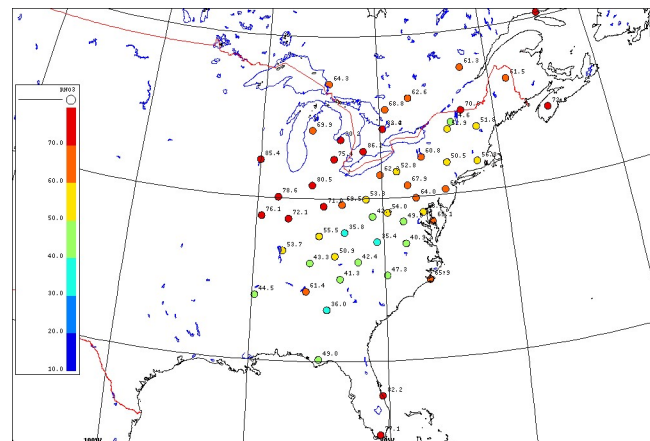
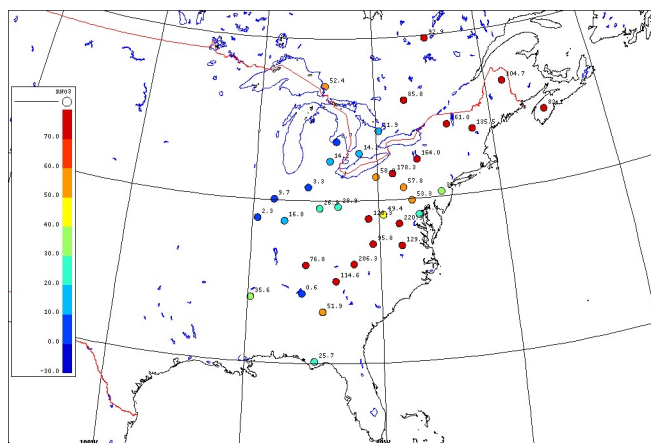
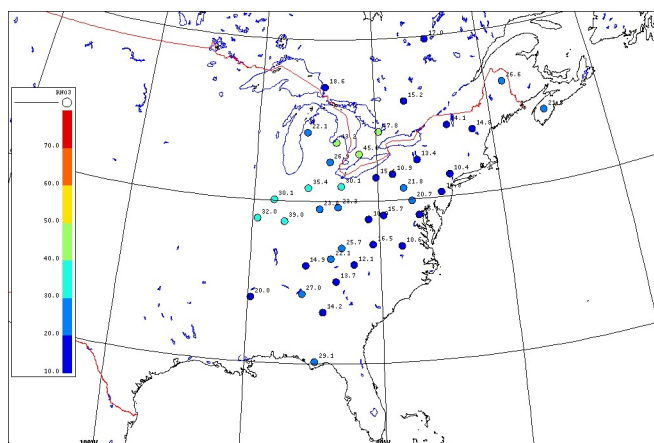
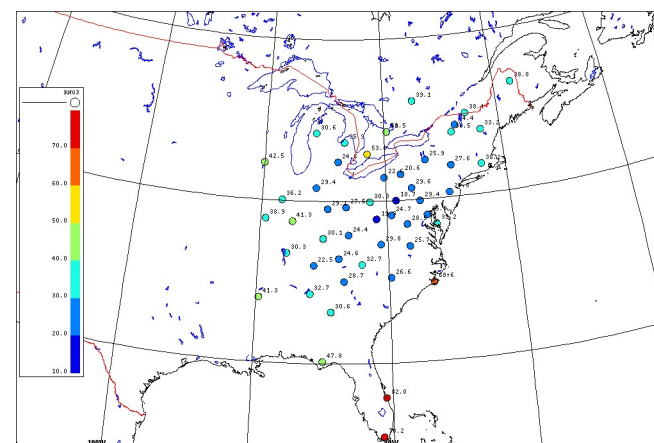
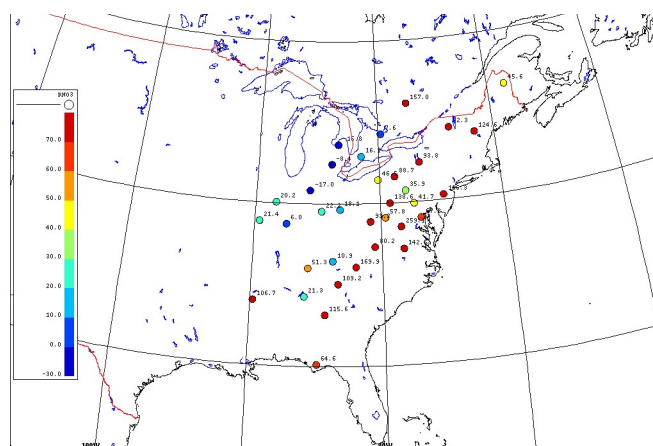



Fig. S2 Mean air concentrations (μg m^{-3}) of SO_2, SO_4^{2-}, NO_3^-, HNO_3, TNO_3, and NH_4^+, and mean ratios of RSO_4 (%) and RNO_3 (%) during 2014-2016 for (a) warm season and (b) cold season.

1989-1991

2014-2016

Change of RSO$_4$ (%): 2014-2016 vs. 1989-1991

(a) Cold season

1989-1991   2014-2016

Change of RSO$_4$ (%): 2014-2016 vs. 1989-1991

(b) Warm season
Fig. S3 RSO$_4$ of 1989-1991 and 2014-2016, and the change of RSO$_4$: 2014-2016 vs. 1989-1991.

1989-1991

2014-2016

Change of RNO$_3$: 2014-2016 vs. 1989-1991

(a) Cold season

1989-1991

2014-2016

Change of RNO$_3$: 2014-2016 vs. 1989-1991

(b) Warm season

Fig. S4 RNO$_3$ of 1989-1991 and 2014-2016, and change of RNO$_3$: 2014-2016 vs. 1989-1991

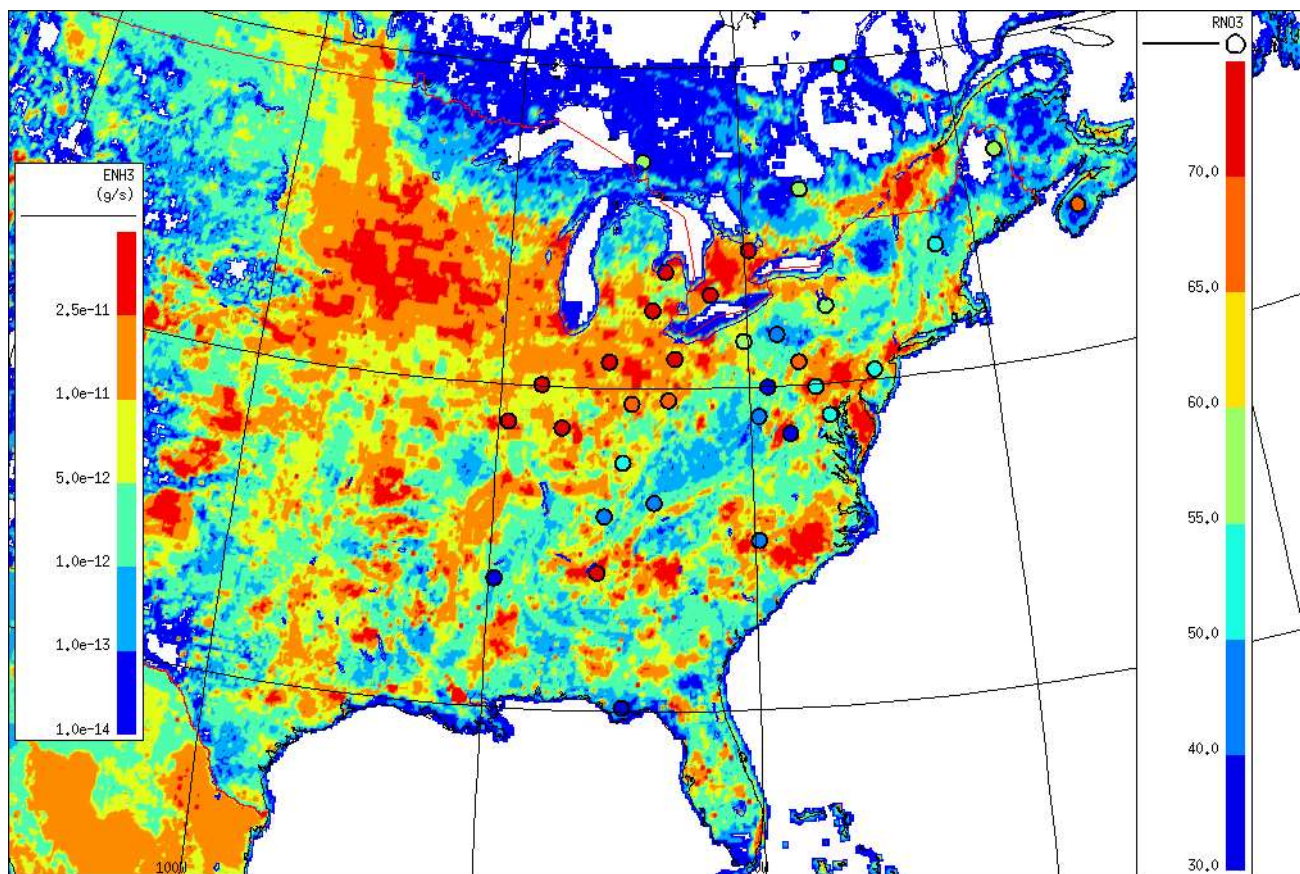



Fig. S5 Overlap of NH_3 emission rate for December 2010 from HTAP with RNO_3 (%) for the cold season of 2010.

TableS1. 3 years' averages of air concentrations ($\mu g\ m^{-3}$), RSO$_4$ (%) and RNO$_3$ (%) for 1989-1991 (a) Warm season

Region	STN	SO_4^{2-}	NO_3^-	NH_4^+	HNO_3	SO_2	TNO_3	RSO_4	RNO_3
1	ALG100	3.11	0.21	0.87	0.90	2.12	1.10	49.73	18.59
	CHA100	3.68	0.13	0.94	0.76	3.14	0.87	44.84	15.22
	CPS100	1.66	0.04	0.37	0.22	0.97	0.26	54.27	17.02
	KEJ100	3.13	0.16	0.59	0.58	1.06	0.72	66.52	21.75
	ASH135	2.42	0.18	0.65	0.52	0.94	0.69	63.27	26.63
	WST109	3.56	0.13	0.97	0.76	1.15	0.88	67.65	14.79
	AVE	**2.93**	**0.14**	**0.73**	**0.62**	**1.56**	**0.75**	**57.71**	**19.00**
	STD	0.76	0.06	0.24	0.24	0.89	0.28	9.47	4.51
2	EGB100	5.67	1.78	2.00	1.97	5.29	3.72	41.94	47.85
	LON100	7.09	2.07	2.42	2.50	7.49	4.53	38.63	45.63
	ALH157	8.23	1.43	2.90	3.04	10.87	4.42	33.47	32.01
	ANA115	6.34	0.96	2.17	2.65	5.39	3.57	43.80	26.86
	BVL130	8.07	1.45	2.70	3.37	7.63	4.77	41.29	30.15
	DCP114	9.82	1.15	3.23	3.83	11.02	4.91	37.22	23.30
	LYK123	8.19	1.51	2.86	3.55	8.90	5.00	37.96	30.10
	OXF122	9.85	1.14	3.17	3.67	12.41	4.75	34.55	23.83
	SAL133	7.92	1.70	2.94	3.09	7.01	4.74	42.99	35.43
	SND152	8.19	0.83	3.06	2.33	6.25	3.13	46.60	26.95
	UVL124	5.63	1.67	2.15	2.24	4.71	3.87	44.11	43.15
	VIN140	9.77	1.81	3.05	2.87	15.99	4.64	29.00	38.95
	AVE	**7.90**	**1.46**	**2.72**	**2.93**	**8.58**	**4.34**	**39.30**	**33.68**
	STD	1.49	0.37	0.43	0.60	3.39	0.61	5.14	8.47
3	ARE128	9.10	1.05	3.05	4.08	10.55	5.06	36.62	20.73
	BEL116	8.75	0.61	2.91	3.40	8.45	3.96	40.83	15.11
	CDR119	9.49	0.17	2.31	1.52	6.68	1.67	48.71	10.01
	CTH110	6.67	0.42	1.92	2.75	8.84	3.13	33.50	13.39
	KEF112	7.39	0.25	1.75	2.07	8.76	2.28	36.17	10.93
	LRL117	9.38	0.20	2.18	2.40	12.66	2.57	33.05	7.83
	MKG113	7.94	0.54	2.35	2.98	12.32	3.47	30.05	15.43
	PAR107	9.25	0.36	2.13	1.93	7.23	2.26	46.30	15.68
	PSU106	8.34	0.77	2.85	2.82	9.30	3.55	37.56	21.76
	WPB104	6.23	0.30	1.74	2.62	8.14	2.88	33.79	10.40
	WSP144	7.67	0.54	2.42	4.11	7.31	4.59	41.21	11.76
	AVE	**8.20**	**0.47**	**2.33**	**2.79**	**9.11**	**3.22**	**37.98**	**13.91**
	STD	1.12	0.27	0.45	0.83	1.98	1.04	5.77	4.40
4	COW137	6.44	0.13	1.55	0.80	0.98	0.91	81.66	13.70
	CVL151	6.83	0.37	1.84	1.51	2.66	1.85	62.93	19.98
	ESP127	8.57	0.29	2.41	1.70	3.94	1.97	59.23	14.89
	GAS153	7.90	0.32	2.19	2.09	5.42	2.38	49.50	14.17
	LCW121	7.99	0.31	1.95	0.89	1.87	1.18	74.01	25.71
	PED108	7.93	0.26	2.07	2.27	4.03	2.49	56.83	10.61
	PNF126	7.69	0.23	1.53	1.72	2.66	1.93	65.88	12.10
	SHN418	8.12	0.24	1.94	2.77	7.06	2.96	43.53	7.95
	SPD111	8.32	0.46	2.61	1.66	3.65	2.10	60.40	22.15
	SUM156	5.01	0.40	0.97	1.06	1.72	1.45	66.06	29.06
	VPI120	8.92	0.53	2.04	2.69	6.39	3.17	48.34	16.55
	AVE	**7.61**	**0.32**	**1.92**	**1.74**	**3.67**	**2.04**	**60.76**	**16.99**
	STD	1.12	0.11	0.45	0.67	1.96	0.70	11.26	6.53

(b) Cold season

Region	STN	SO_4^{2-}	NO_3^-	NH_4^+	HNO_3	SO_2	TNO_3	RSO_4	RNO_3
1	ALG100	2.20	0.69	0.69	0.94	3.48	1.61	29.74	42.19
	CHA100	2.58	0.50	0.72	0.98	6.36	1.46	21.38	33.69
	CPS100	1.72	0.09	0.29	0.23	2.10	0.31	35.32	27.43
	KEJ100	2.49	0.33	0.43	0.51	2.90	0.83	37.04	39.86
	ASH135	2.30	0.32	0.56	0.77	3.20	1.08	32.41	30.04
	WST109	2.46	0.29	0.71	1.02	3.70	1.29	30.90	21.98
	AVE	**2.29**	**0.37**	**0.57**	**0.74**	**3.62**	**1.10**	**31.13**	**32.53**
	STD	0.31	0.20	0.17	0.31	1.45	0.47	5.50	7.64
2	EGB100	3.49	3.25	1.86	1.13	8.73	4.36	21.12	74.57
	LON100	4.26	4.10	2.30	1.35	11.68	5.43	19.56	75.44
	ALH157	4.35	4.18	2.56	1.45	15.11	5.61	16.05	74.35
	ANA115	3.79	3.25	2.03	1.70	10.88	4.92	19.04	66.00
	BVL130	4.16	4.07	2.37	1.66	10.14	5.71	21.50	71.61
	DCP114	4.34	2.53	2.09	2.20	14.96	4.70	16.24	53.90
	LYK123	4.29	3.77	2.37	1.80	14.02	5.54	17.07	68.08
	OXF122	4.60	2.81	2.23	2.25	20.69	5.02	12.99	55.96
	SAL133	4.47	4.99	2.85	1.43	12.45	6.40	19.34	77.92
	UVL124	3.62	3.59	2.08	1.29	8.55	4.86	22.05	73.84
	VIN140	4.77	3.22	2.25	2.03	22.91	5.22	12.18	61.72
	AVE	**4.19**	**3.62**	**2.27**	**1.66**	**13.65**	**5.25**	**17.92**	**68.49**
	STD	0.40	0.70	0.27	0.37	4.64	0.56	3.32	8.18
3	ARE128	4.63	2.16	2.06	3.04	19.54	5.16	13.73	41.75
	BEL116	4.89	1.85	2.11	2.02	18.09	3.84	15.34	47.36
	CDR119	4.05	0.38	1.22	1.66	15.41	2.02	15.20	18.65
	CTH110	3.62	0.83	1.29	2.77	15.20	3.56	13.70	23.04
	KEF112	4.01	0.58	1.27	2.50	20.71	3.04	11.44	18.96
	LRL117	4.17	0.54	1.28	2.30	25.73	2.80	9.81	18.93
	MKG113	4.32	1.54	1.68	2.38	23.46	3.88	10.95	39.45
	PAR107	4.11	0.91	1.16	1.66	17.28	2.54	13.77	36.15
	PSU106	4.37	1.73	1.85	2.31	20.17	4.00	12.62	43.03
	WPB104	3.89	0.92	1.33	2.28	17.62	3.17	12.96	28.91
	WSP144	4.61	1.92	1.90	2.46	17.95	4.34	14.73	44.07
	AVE	**4.24**	**1.21**	**1.56**	**2.31**	**19.20**	**3.49**	**13.11**	**32.76**
	STD	0.37	0.64	0.36	0.42	3.22	0.89	1.78	11.28
4	COW137	2.86	0.22	0.85	0.94	2.62	1.14	42.27	19.25
	CVL151	3.57	0.73	1.25	1.53	4.90	2.24	33.09	32.83
	ESP127	4.15	0.69	1.40	2.17	9.58	2.83	22.36	24.51
	GAS153	3.72	0.61	1.22	2.03	9.56	2.60	21.07	23.69
	LCW121	3.69	0.69	0.94	0.80	4.91	1.47	33.96	46.89
	PED108	4.18	0.54	1.37	2.51	10.40	3.01	21.21	17.86
	PNF126	2.82	0.28	0.82	1.76	4.83	2.01	27.94	13.83
	SHN418	3.35	0.47	1.14	2.61	11.71	3.04	16.07	15.51
	SUM156	3.77	0.67	0.85	1.07	2.84	1.72	47.02	39.01
	VPI120	3.59	0.59	1.07	2.73	11.01	3.27	17.91	18.06
	AVE	**3.57**	**0.55**	**1.09**	**1.81**	**7.24**	**2.33**	**28.29**	**25.14**
	STD	0.46	0.18	0.22	0.71	3.54	0.73	10.52	10.98

Table S2. 3 years' averages of air concentrations (μg m^{-3}), RSO$_4$ (%) and RNO$_3$ (%) for 2014-2016. (a) Warm season

Region	STN	SO$_4^{2-}$	NO$_3^-$	NH$_4^+$	HNO$_3$	SO$_2$	TNO$_3$	RSO$_4$	RNO$_3$
1	CHA100	1.10	0.16	0.36	0.26	0.63	0.42	53.74	39.11
	ASH135	0.57	0.11	0.21	0.17	0.30	0.27	55.34	38.77
	WST109	0.73	0.10	0.26	0.21	0.26	0.31	65.00	33.23
	AVE	**0.80**	**0.12**	**0.28**	**0.21**	**0.40**	**0.33**	**58.03**	**37.04**
	STD	0.27	0.03	0.07	0.05	0.20	0.08	6.09	3.30
2	EGB100	1.54	0.96	0.64	0.92	0.70	1.87	59.64	51.49
	LON100	1.84	0.97	0.75	0.88	0.96	1.84	56.23	53.00
	ALH157	1.96	0.67	0.65	1.07	1.03	1.72	55.60	38.87
	ANA115	1.54	0.34	0.52	1.05	0.90	1.37	53.13	24.60
	BVL130	1.88	0.68	0.64	1.21	1.15	1.87	52.98	36.25
	DCP114	2.00	0.45	0.74	1.19	1.00	1.62	57.54	27.52
	OXF122	2.09	0.49	0.75	1.21	1.38	1.68	51.10	29.12
	SAL133	1.78	0.54	0.65	1.32	0.98	1.84	54.66	29.41
	SND152	1.80	0.45	0.62	0.94	0.64	1.37	65.04	32.68
	UVL124	1.35	0.56	0.56	1.02	0.92	1.57	49.25	35.88
	VIN140	2.33	0.73	0.73	1.05	1.70	1.76	48.21	41.31
	AVE	**1.83**	**0.62**	**0.66**	**1.08**	**1.03**	**1.68**	**54.85**	**36.38**
	STD	0.28	0.21	0.08	0.14	0.30	0.18	4.82	9.31
3	ARE128	1.77	0.52	0.66	1.28	1.07	1.78	52.30	29.38
	BEL116	1.67	0.37	0.52	1.13	0.82	1.48	57.64	25.26
	CDR119	1.89	0.10	0.63	0.41	0.48	0.50	72.12	19.34
	CTH110	1.38	0.25	0.52	0.71	0.72	0.94	55.80	25.94
	KEF112	1.68	0.17	0.56	0.67	0.82	0.84	58.33	20.62
	LRL117	1.94	0.15	0.65	0.65	0.77	0.78	63.09	18.69
	MCK131	2.09	0.42	0.74	1.00	0.77	1.40	64.69	30.15
	MKG113	1.81	0.26	0.61	0.89	0.99	1.13	55.22	22.61
	PAR107	1.92	0.18	0.60	0.55	0.47	0.71	72.83	24.74
	PSU106	1.83	0.42	0.68	1.02	1.20	1.42	51.39	29.56
	WSP144	1.43	0.49	0.45	1.29	0.68	1.76	58.25	27.79
	AVE	**1.77**	**0.30**	**0.60**	**0.87**	**0.80**	**1.16**	**60.15**	**24.92**
	STD	0.21	0.15	0.08	0.30	0.22	0.44	7.27	4.15
4	CND125	1.55	0.27	0.47	0.74	0.43	1.00	69.82	26.63
	COW137	1.27	0.12	0.42	0.29	0.22	0.40	79.35	28.66
	CVL151	1.77	0.40	0.53	0.58	0.56	0.97	67.76	41.29
	ESP127	1.86	0.18	0.53	0.63	0.40	0.80	75.41	22.53
	GAS153	1.57	0.33	0.48	0.76	0.46	1.08	68.98	30.56
	PED108	1.62	0.22	0.53	0.64	0.48	0.84	68.60	25.73
	PNF126	1.52	0.25	0.55	0.51	0.33	0.75	75.14	32.66
	SHN418	1.65	0.31	0.58	0.78	0.59	1.08	65.04	28.54
	SPD111	1.65	0.21	0.49	0.65	0.43	0.85	71.54	24.58
	SUM156	1.53	0.35	0.30	0.39	0.37	0.74	73.26	47.84
	VPI120	1.84	0.30	0.62	0.71	2.20	1.00	36.24	29.82
	AVE	**1.62**	**0.27**	**0.50**	**0.61**	**0.59**	**0.86**	**68.29**	**30.80**
	STD	0.17	0.08	0.09	0.16	0.54	0.20	11.39	7.51

(b) Cold season

Region	STN	SO_4^{2-}	NO_3^-	NH_4^+	HNO_3	SO_2	TNO_3	RSO_4	RNO_3
1	ALG100	0.95	0.72	0.43	0.41	0.78	1.12	44.89	64.29
	CHA100	0.83	0.55	0.35	0.33	1.30	0.87	29.95	62.60
	KEJ100	0.93	0.35	0.21	0.13	0.32	0.48	65.95	72.58
	ASH135	0.81	0.31	0.28	0.20	0.67	0.51	44.45	61.49
	WST109	0.84	0.30	0.32	0.28	0.66	0.57	46.02	51.77
	AVE	**0.87**	**0.44**	**0.32**	**0.27**	**0.75**	**0.71**	**46.25**	**62.54**
	STD	0.06	0.18	0.08	0.11	0.36	0.28	12.83	7.43
2	EGB100	1.40	2.31	1.02	0.47	1.63	2.77	36.28	83.41
	LON100	1.73	3.08	1.36	0.50	2.51	3.58	31.94	86.17
	ALH157	1.71	2.53	1.13	0.79	1.79	3.30	39.09	76.09
	ANA115	1.59	2.34	1.07	0.77	1.73	3.09	38.51	75.40
	BVL130	1.65	2.79	1.22	0.75	1.93	3.53	37.60	78.57
	DCP114	1.81	2.16	1.13	0.95	2.38	3.09	34.69	69.48
	OXF122	1.81	2.23	1.13	0.91	3.08	3.13	30.45	70.99
	SAL133	1.74	3.14	1.38	0.75	1.86	3.88	39.13	80.47
	UVL124	1.43	2.37	1.05	0.59	1.76	2.95	35.22	80.24
	VIN140	1.86	2.27	1.12	0.86	3.20	3.12	28.93	72.06
	AVE	**1.67**	**2.52**	**1.16**	**0.73**	**2.19**	**3.24**	**35.18**	**77.29**
	STD	0.16	0.36	0.12	0.16	0.58	0.33	3.67	5.47
3	ARE128	1.65	1.90	0.99	1.08	2.63	2.97	30.40	64.03
	BEL116	1.70	1.41	0.80	1.00	2.12	2.39	35.90	58.90
	CDR119	1.58	0.45	0.59	0.62	1.50	1.06	43.58	42.59
	CTH110	1.28	1.07	0.65	0.68	1.61	1.74	35.16	60.83
	KEF112	1.50	0.87	0.66	0.78	2.40	1.64	30.72	52.76
	MCK131	1.78	1.35	0.89	1.09	1.81	2.42	40.68	55.45
	MKG113	1.70	1.44	0.84	0.85	2.87	2.28	29.15	62.32
	PAR107	1.61	0.71	0.55	0.61	1.58	1.31	41.67	54.01
	PSU106	1.68	1.65	0.96	0.78	3.35	2.41	26.74	67.91
	WSP144	1.45	1.63	0.76	1.07	1.85	2.68	35.14	60.68
	AVE	**1.59**	**1.25**	**0.77**	**0.86**	**2.17**	**2.09**	**34.91**	**57.95**
	STD	0.15	0.46	0.15	0.19	0.62	0.62	5.71	7.12
4	CND125	1.40	0.68	0.57	0.77	0.92	1.44	50.62	47.32
	COW137	0.99	0.28	0.39	0.39	0.44	0.66	60.31	41.31
	CVL151	1.55	0.54	0.56	0.69	0.93	1.22	52.31	44.51
	ESP127	1.63	0.68	0.64	0.90	1.09	1.57	50.56	43.32
	GAS153	1.42	0.50	0.52	0.90	1.06	1.38	47.77	35.99
	PED108	1.40	0.53	0.56	0.77	1.18	1.29	45.22	40.94
	PNF126	1.06	0.44	0.43	0.60	0.74	1.03	48.76	42.37
	SHN418	1.12	0.70	0.50	0.71	1.21	1.40	39.05	49.77
	SPD111	1.50	0.73	0.62	0.71	0.96	1.43	51.72	50.86
	SUM156	1.52	0.43	0.34	0.43	0.52	0.85	66.24	49.03
	VPI120	1.38	0.52	0.52	0.97	3.41	1.47	21.26	35.37
	AVE	**1.36**	**0.55**	**0.51**	**0.71**	**1.13**	**1.25**	**48.53**	**43.71**
	STD	0.21	0.14	0.10	0.18	0.80	0.29	11.53	5.24