Supplement: An Ensemble-Variational Inversion System for the Estimation of Ammonia Emissions using CrIS Satellite Ammonia Retrievals

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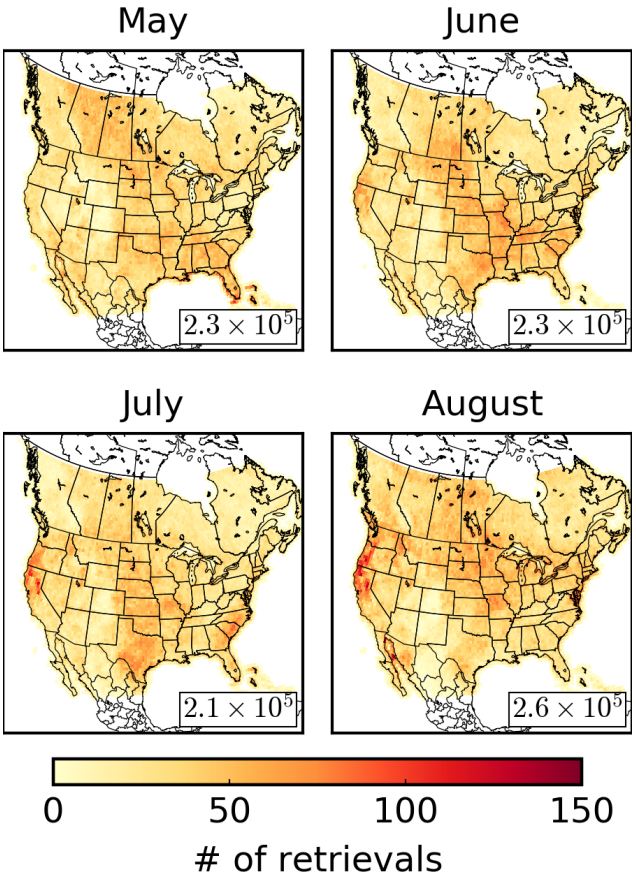

Figure S1. Number of CrIS ammonia retrievals used in the inversions within $0.5^\circ \times 0.5^\circ$ longitude/latitude bins for May to August 2016. The total number of retrievals for each month is displayed in the lower right corner of each panel.

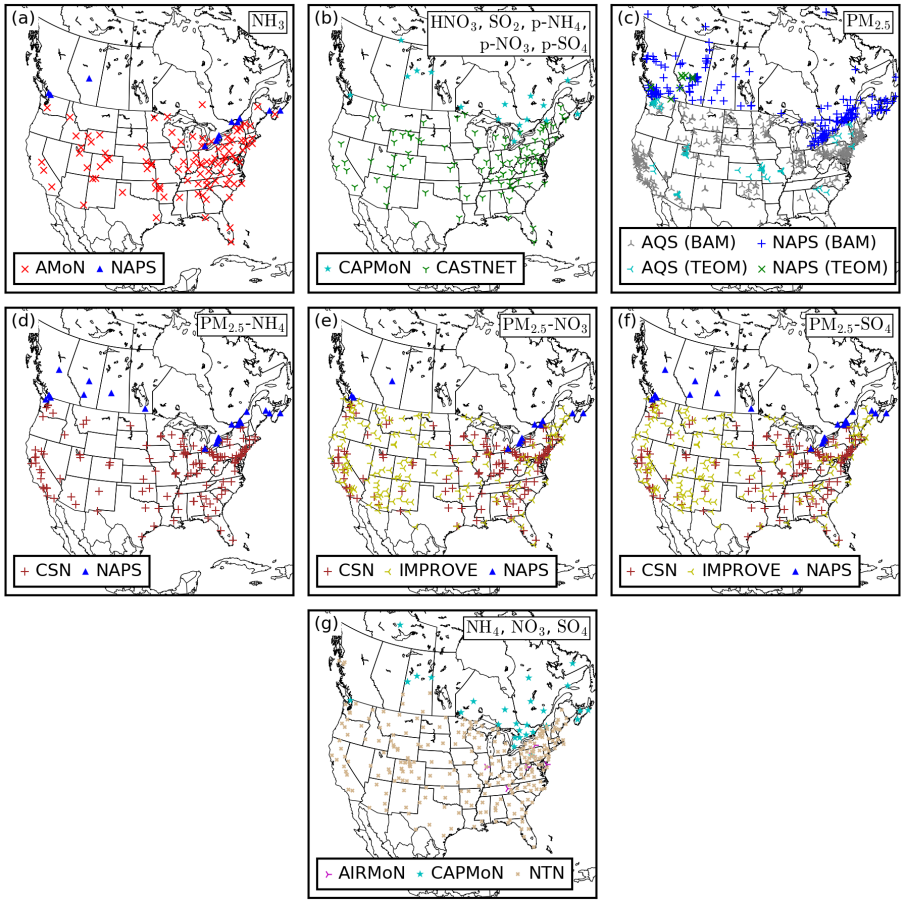

Figure S2. Observation sites measuring the atmospheric surface concentrations of (a) ammonia, (b) nitric acid, sulfur dioxide, and total ammonium, nitrate, and sulfate, (c) total $PM_{2.5}$, the $PM_{2.5}$ component of (d) ammonium, (e) nitrate, (f) sulfate, and precipitation concentrations of (g) ammonium, nitrate, and sulfate. The networks measuring each species is displayed in the legend of each panel.

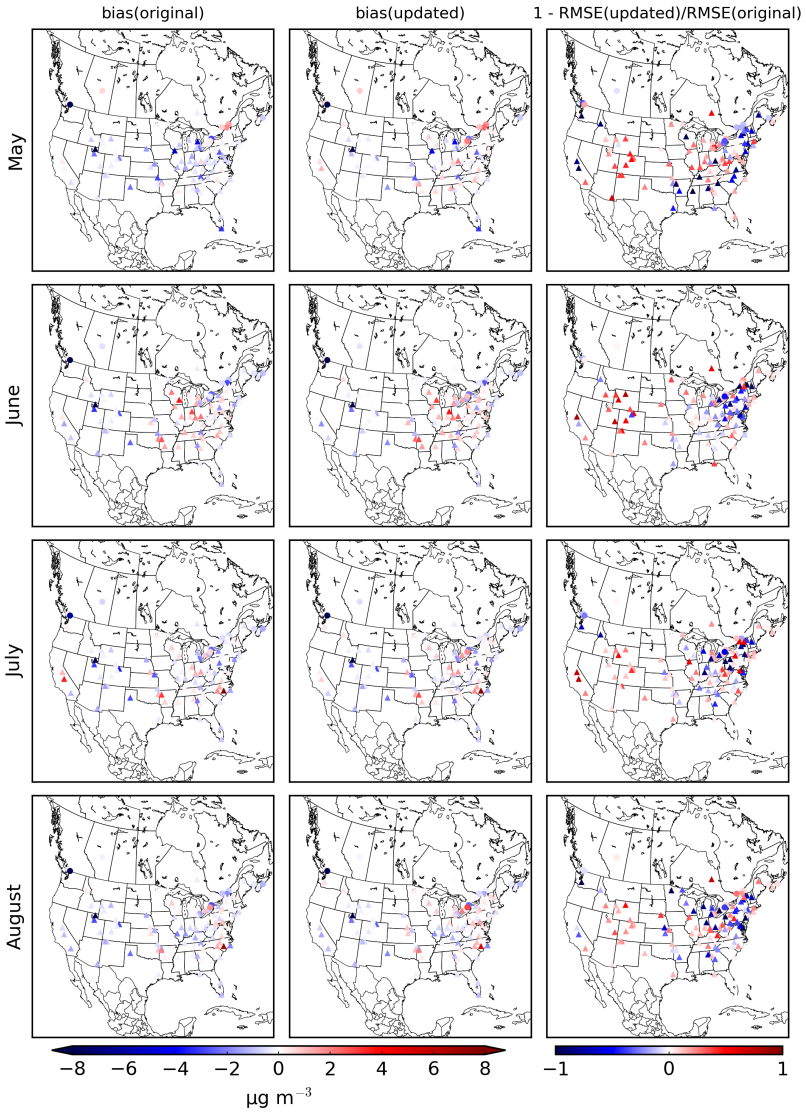

Figure S3. Comparisons of ammonia surface observations from the NAPS and AMoN networks with GEM-MACH surface fields. The left and center columns show bias values for each station when the original and updated ammonia emissions are used, respectively. The right column shows the relative improvement of the root-mean-square error (RMSE) for each station. NAPS and AMoN stations are denoted with circular and triangular markers, respectively.

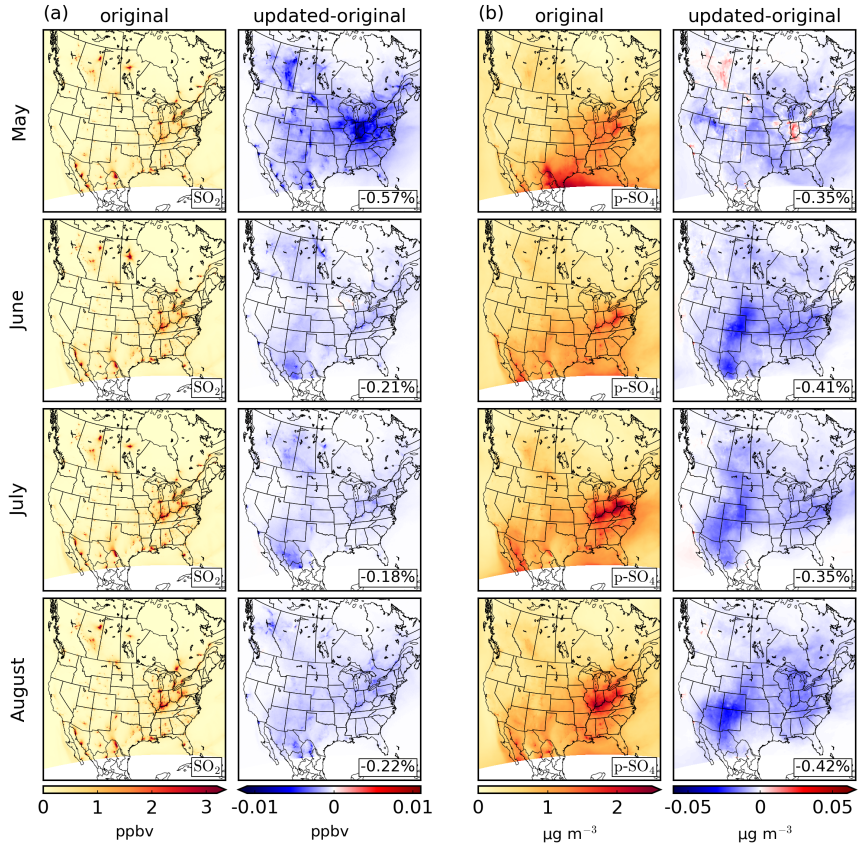

Figure S4. Monthly mean surface (a) SO_2 and (b) p-SO_4 fields from GEM-MACH for May to August 2016 on the $0.09^\circ \times 0.09^\circ$ model grid. In sub-figures of (a) and (b), the left columns show the mean surface field when the original ammonia emissions are used and the right columns show the mean difference between GEM-MACH run with the updated ammonia emissions from the inversions and the original emissions. For plots in the right column, the total difference over the model domain as a percentage of the original field is shown in the lower right corner. Plots for sulfate show the total sulfate mass over all aerosol size bins.

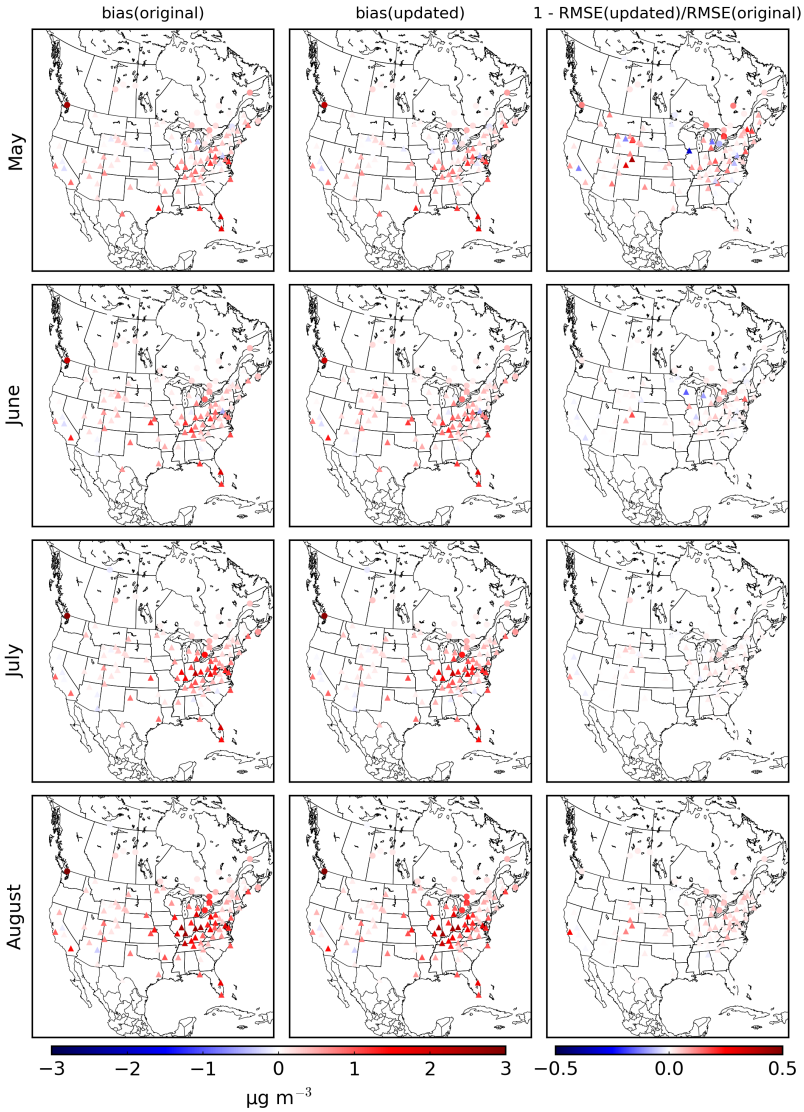

Figure S5. Same as Figure S3, but for HNO_3 observations from the CAPMoN and CASTNET networks, which are denoted by circular and triangular markers, respectively.

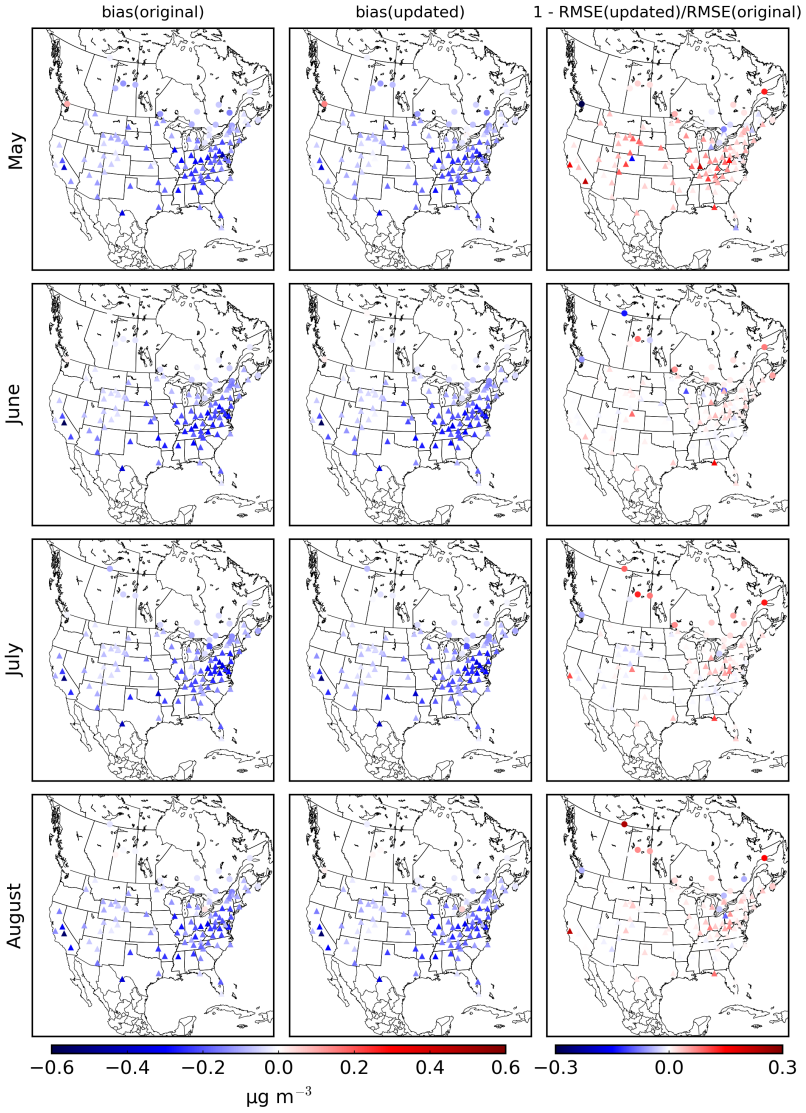

Figure S6. Same as Figure S5, but for p-NH_4.

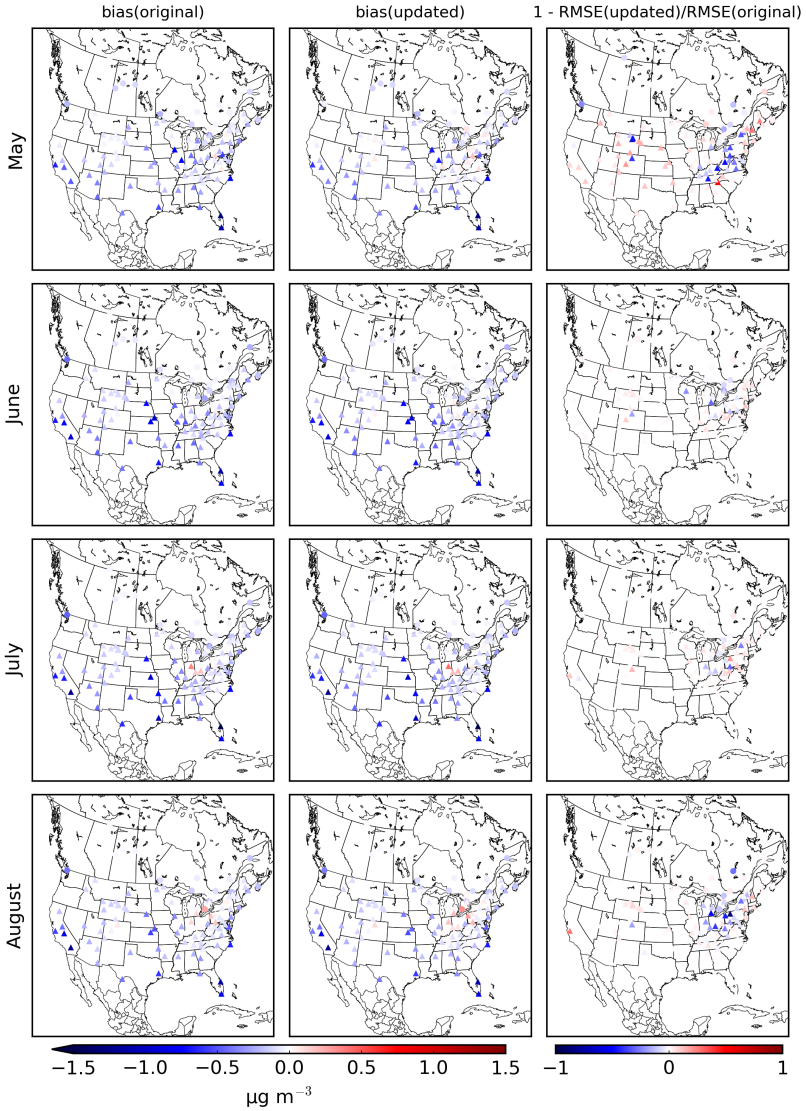

Figure S7. Same as Figure S5, but for p-NO_3.

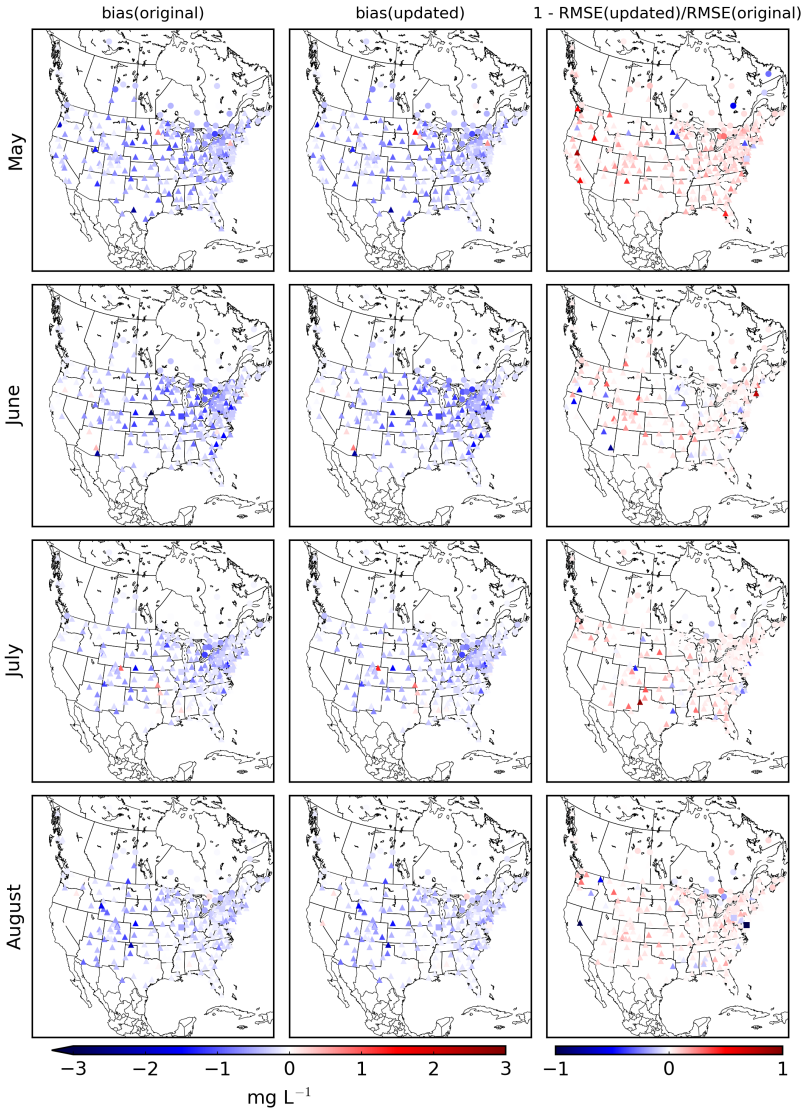

Figure S8. Same as Figure S3, but for precipitation-chemistry concentration observations of ammonium from the CAPMoN, NTN, and AIRMoN networks, which are denoted by circular, triangular, and square markers, respectively.

species	month	network	$\bar{O}$	NMB (%)			NSTD (%)			ρ			N
			($\mu g\,m^{-3}$)	original	updated	sig	original	updated	sig	original	updated	sig	
NH_3	May	NAPS	3.84	-22.9±12.5	-3.8±11.1	74	120.6±35.0	107.5±30.7	15	0.67±0.08	0.74±0.07	79	93
		AMoN	1.82	-43.0±4.6	-17.9±4.5	100	97.3±10.7	95.6±10.0	17	0.50±0.04	0.53±0.04	65	446
	June	NAPS	3.81	-46.3±15.1	-36.9±14.8	35	143.1±62.7	140.0±60.5	3	0.74±0.07	0.72±0.07	30	90
		AMoN	2.02	-4.2±5.2	3.7±5.0	73	99.2±11.0	96.2±10.8	31	0.44±0.05	0.48±0.05	61	368
	July	NAPS	3.14	-35.8±5.6	-29.2±7.2	53	55.4±8.5	71.2±12.4	54	0.88±0.05	0.78±0.06	100	98
		AMoN	1.87	-17.7±5.2	-13.9±4.7	41	99.7±14.7	89.7±14.3	42	0.53±0.04	0.56±0.04	62	363
	August	NAPS	3.48	-38.5±14.0	-29.5±15.3	34	133.5±48.1	145.6±51.1	17	0.64±0.08	0.51±0.09	93	91
		AMoN	1.79	-23.5±5.0	-12.2±4.8	90	95.4±16.3	92.3±16.2	6	0.55±0.04	0.57±0.04	43	370
p-NH_4	May	CAPMoN	0.20	-27.4±3.6	-22.0±3.9	70	86.1±6.0	92.1±6.8	43	0.61±0.03	0.58±0.03	72	570
		CASTNET	0.36	-42.5±1.9	-38.3±1.9	88	44.6±3.4	45.1±3.3	23	0.68±0.03	0.66±0.03	42	547
	June	CAPMoN	0.14	-31.4±3.9	-26.2±4.0	65	91.1±8.4	94.6±9.7	20	0.58±0.03	0.55±0.04	64	552
		CASTNET	0.34	-48.1±1.7	-47.6±1.7	15	35.9±1.5	36.0±1.5	4	0.73±0.03	0.73±0.03	21	465
	July	CAPMoN	0.19	-36.4±3.6	-31.9±3.6	63	84.5±15.0	85.5±14.7	11	0.64±0.03	0.63±0.03	35	558
		CASTNET	0.36	-42.3±1.6	-41.9±1.6	15	33.9±1.4	33.9±1.4	3	0.79±0.03	0.79±0.03	7	454
	August	CAPMoN	0.18	-21.8±3.3	-16.2±3.4	76	78.1±5.8	79.9±5.9	17	0.69±0.03	0.68±0.03	35	545
		CASTNET	0.36	-41.1±1.6	-39.7±1.7	45	38.3±1.5	38.8±1.5	1	0.70±0.03	0.70±0.03	28	553

Table S1. Comparison of surface observations of NH_3 and p-NH_4 with GEM-MACH using the original and updated ammonia emissions for May to August 2016. The normalized mean bias, normalized standard deviation of differences, and correlation coefficient are shown with their standard errors. The columns labeled 'sig' are the statistical significance (displayed as a percentage) of the difference between the original and updated statistical values. Values for ammonium are for the total mass over all aerosol sizes.

species	month	network	$\bar{O}$	NMB (%)			NSTD (%)			ρ			N
			(μg m^{-3})	original	updated	sig	original	updated	sig	original	updated	sig	
HNO_3	May	CAPMoN	0.39	71.2±9.6	64.0±8.6	43	228.4±32.8	205.9±27.9	37	0.63±0.03	0.65±0.03	51	571
		CASTNET	0.62	59.1±4.7	52.2±4.6	71	109.2±7.8	107.6±7.5	13	0.54±0.04	0.52±0.04	43	547
	June	CAPMoN	0.33	93.3±10.4	90.1±10.0	18	243.2±25.8	234.9±25.2	15	0.70±0.03	0.70±0.03	8	552
		CASTNET	0.76	53.7±3.1	52.8±3.1	16	67.7±3.2	67.2±3.1	11	0.68±0.03	0.68±0.03	3	465
	July	CAPMoN	0.37	113.6±11.5	111.7±11.2	9	270.5±34.2	265.6±33.5	9	0.71±0.03	0.71±0.03	8	558
		CASTNET	0.73	70.9±4.3	70.0±4.2	12	91.3±6.2	90.4±6.1	11	0.66±0.04	0.66±0.04	2	454
	August	CAPMoN	0.32	166.8±14.1	162.3±13.7	18	330.3±43.9	321.7±43.3	18	0.73±0.03	0.72±0.03	18	549
		CASTNET	0.65	117.3±6.4	113.9±6.2	30	149.7±10.7	146.3±10.6	23	0.53±0.04	0.53±0.04	7	553
p-NO_3	May	CAPMoN	0.26	-39.1±7.0	-28.7±7.4	69	166.4±16.3	176.6±16.4	23	0.53±0.04	0.53±0.04	9	570
		CASTNET	0.38	-54.2±4.6	-41.1±4.8	95	108.2±10.2	111.6±9.7	55	0.42±0.04	0.40±0.04	42	547
	June	CAPMoN	0.17	-72.9±6.5	-67.7±6.9	42	153.2±17.4	163.0±23.9	3	0.31±0.04	0.32±0.04	24	552
		CASTNET	0.33	-74.1±4.0	-71.9±4.0	31	86.0±4.5	87.1±4.5	16	0.21±0.05	0.20±0.05	20	465
	July	CAPMoN	0.17	-70.9±7.2	-66.6±7.3	33	170.0±17.4	172.5±17.6	4	0.28±0.04	0.29±0.04	21	558
		CASTNET	0.31	-78.2±5.0	-76.1±5.0	24	105.8±8.7	106.8±8.7	11	0.04±0.05	0.04±0.05	1	454
	August	CAPMoN	0.18	-57.8±6.5	-50.2±6.7	58	150.8±12.4	156.7±13.6	11	0.39±0.04	0.41±0.04	43	544
		CASTNET	0.27	-66.9±4.6	-60.0±4.8	70	107.8±7.0	112.2±7.1	43	0.01±0.04	0.01±0.04	3	553

Table S2. Same as Table S1, but for surface observations of HNO_3 and p-NO_3. Values for nitrate are for the total mass over all aerosol sizes.

species	month	network	$\bar{O}$	NMB (%)			NSTD (%)			ρ			N
			(μg m^{-3})	original	updated	sig	original	updated	sig	original	updated	sig	
SO_2	May	CAPMoN	0.23	169.7±18.0	168.1±17.9	5	428.7±80.5	427.9±80.4	2	0.48±0.04	0.48±0.04	0	569
		CASTNET	0.41	47.6±5.1	46.1±5.1	16	119.7±7.2	118.8±7.2	9	0.57±0.04	0.57±0.04	2	547
	June	CAPMoN	0.16	205.1±16.2	204.1±16.2	3	380.7±25.5	380.5±25.5	1	0.49±0.04	0.49±0.04	0	549
		CASTNET	0.56	18.7±5.2	18.5±5.2	3	112.1±7.5	112.0±7.5	1	0.44±0.04	0.44±0.04	1	465
	July	CAPMoN	0.18	223.1±17.5	222.3±17.5	3	412.1±33.4	411.8±33.4	1	0.49±0.04	0.49±0.04	0	557
		CASTNET	0.54	32.6±6.0	32.4±6.0	2	127.0±7.1	126.9±7.1	1	0.40±0.04	0.40±0.04	0	454
	August	CAPMoN	0.17	313.3±23.3	312.1±23.3	3	547.6±61.2	547.1±61.2	2	0.46±0.04	0.46±0.04	0	551
		CASTNET	0.47	55.9±7.6	55.5±7.5	3	177.6±13.7	177.5±13.7	1	0.37±0.04	0.37±0.04	0	553
p-SO_4	May	CAPMoN	0.56	-48.4±2.8	-48.8±2.8	8	66.8±4.4	67.3±4.5	5	0.70±0.03	0.70±0.03	17	570
		CASTNET	1.01	-57.3±1.7	-57.5±1.7	7	40.2±1.7	40.3±1.7	3	0.72±0.03	0.72±0.03	6	547
	June	CAPMoN	0.41	-40.3±3.3	-40.7±3.3	8	77.9±5.3	77.9±5.3	2	0.65±0.03	0.65±0.03	8	552
		CASTNET	1.03	-59.7±2.0	-60.1±2.0	11	43.7±1.9	43.7±1.9	2	0.73±0.03	0.74±0.03	19	465
	July	CAPMoN	0.60	-45.1±4.1	-45.5±4.1	6	96.1±18.1	96.2±18.2	1	0.62±0.03	0.62±0.03	3	558
		CASTNET	1.17	-55.1±1.9	-55.5±1.9	11	40.1±1.7	40.0±1.7	1	0.75±0.03	0.76±0.03	11	454
	August	CAPMoN	0.58	-39.1±3.3	-39.6±3.3	10	77.2±4.9	77.1±4.9	0	0.68±0.03	0.68±0.03	10	549
		CASTNET	1.08	-52.3±1.7	-52.8±1.7	15	39.9±1.4	39.9±1.4	1	0.71±0.03	0.71±0.03	10	553

Table S3. Same as Table S1, but for surface observations of SO_2 and p-SO_4. Values for sulfate are for the total mass over all aerosol sizes.

species	month	network	$\bar{O}$	NMB (%)			NSTD (%)			ρ			N
			($mg\,L^{-1}$)	original	updated	sig	original	updated	sig	original	updated	sig	
NH_4	May	CAPMoN	0.44	-73.2±6.1	-64.9±6.0	67	96.8±7.2	95.8±7.7	24	0.54±0.05	0.55±0.05	3	255
		NTN	0.48	-76.4±3.6	-68.5±3.9	86	98.1±10.2	104.6±13.1	9	0.21±0.04	0.21±0.04	4	727
		AIRMoN	0.58	-77.8±10.6	-71.1±10.6	35	85.3±8.7	85.8±8.8	3	0.27±0.12	0.29±0.12	13	65
	June	CAPMoN	0.34	-76.6±9.0	-73.6±8.9	19	141.3±18.3	140.7±18.0	4	0.46±0.06	0.46±0.06	1	248
		NTN	0.55	-74.5±3.9	-72.9±4.0	23	83.6±7.0	85.1±7.0	7	0.38±0.04	0.33±0.04	73	464
		AIRMoN	0.66	-70.3±13.9	-69.0±13.9	5	82.5±13.8	82.2±13.7	2	-0.00±0.17	-0.00±0.17	2	35
	July	CAPMoN	0.32	-64.4±8.4	-59.7±8.3	31	131.8±17.0	130.6±16.7	0	0.49±0.06	0.50±0.06	16	245
		NTN	0.42	-67.2±3.7	-63.9±3.7	47	86.6±8.4	88.0±8.8	5	0.30±0.04	0.30±0.04	0	552
		AIRMoN	0.38	-61.4±10.9	-58.2±11.1	16	69.9±7.0	71.0±7.0	14	0.30±0.15	0.29±0.15	4	41
	August	CAPMoN	0.29	-59.4±8.5	-51.5±8.9	48	135.1±14.1	141.4±16.1	12	0.26±0.06	0.25±0.06	12	253
		NTN	0.37	-75.2±4.4	-71.3±4.4	47	117.8±22.0	116.6±22.0	10	0.39±0.03	0.40±0.03	24	715
		AIRMoN	0.28	-75.7±15.0	-71.8±15.5	14	93.8±11.8	96.5±13.0	6	-0.02±0.16	-0.02±0.16	0	39
NO_3	May	CAPMoN	0.98	-81.6±6.3	-81.3±6.3	3	103.5±10.0	102.9±9.7	1	0.68±0.05	0.68±0.05	4	267
		NTN	0.95	-82.6±3.7	-82.4±3.7	2	99.9±21.1	100.1±21.2	1	0.26±0.04	0.25±0.04	23	726
		AIRMoN	1.16	-82.3±9.3	-82.3±9.3	0	74.9±10.4	74.9±10.4	0	0.10±0.13	0.09±0.13	1	65
	June	CAPMoN	0.74	-84.9±8.8	-84.9±8.8	0	141.0±25.8	141.0±25.8	0	0.53±0.05	0.53±0.05	1	255
		NTN	1.10	-85.4±3.5	-85.4±3.5	0	76.0±7.4	75.9±7.3	0	0.11±0.05	0.11±0.05	1	464
		AIRMoN	1.31	-83.2±11.7	-83.3±11.7	0	68.1±7.7	68.1±7.7	0	0.05±0.18	0.05±0.18	1	34
	July	CAPMoN	0.83	-77.1±7.3	-77.2±7.3	0	114.8±13.9	115.0±14.0	1	0.48±0.06	0.48±0.06	5	249
		NTN	1.16	-85.2±3.0	-85.2±3.0	1	70.3±6.0	70.3±6.0	0	0.22±0.04	0.22±0.04	2	550
		AIRMoN	1.32	-85.1±12.6	-85.1±12.6	0	82.8±18.7	82.7±18.7	0	0.08±0.16	0.08±0.16	0	43
	August	CAPMoN	0.71	-70.9±7.9	-70.8±7.9	0	126.0±17.3	126.2±17.3	0	0.24±0.06	0.24±0.06	2	257
		NTN	0.92	-83.9±3.5	-83.9±3.5	1	95.0±15.5	95.0±15.5	0	0.32±0.04	0.32±0.04	2	717
		AIRMoN	0.84	-85.7±9.4	-85.7±9.4	0	58.9±6.0	58.9±6.0	0	0.13±0.16	0.13±0.16	0	39

Table S4. Same as Table S1, but for precipitation-chemistry concentration observations of ammonium and nitrate.

species	month	network	$\bar{O}$	NMB (%)			NSTD (%)			ρ			N
			($\mathrm{mg\,L^{-1}}$)	original	updated	sig	original	updated	sig	original	updated	sig	
SO_4	May	CAPMoN	0.84	-82.3±10.5	-82.3±10.5	0	172.1±54.3	172.0±54.3	0	0.31±0.06	0.32±0.06	3	267
		NTN	0.74	-84.6±17.1	-84.3±17.1	1	461.8±224.7	461.6±224.7	0	0.10±0.04	0.10±0.04	20	726
		AIRMoN	0.77	-80.2±9.5	-80.0±9.5	1	76.6±14.0	76.5±14.0	1	0.12±0.13	0.13±0.12	4	65
	June	CAPMoN	0.51	-80.6±7.6	-80.6±7.6	0	121.3±24.6	121.2±24.6	0	0.34±0.06	0.34±0.06	3	255
		NTN	0.67	-85.0±3.6	-84.8±3.6	3	77.2±6.7	77.2±6.7	0	0.13±0.05	0.14±0.05	8	465
		AIRMoN	0.74	-76.2±13.8	-76.2±13.7	0	80.2±15.3	80.0±15.3	0	-0.12±0.18	-0.12±0.18	2	34
	July	CAPMoN	0.49	-75.1±6.7	-74.9±6.7	1	106.7±11.3	106.5±11.3	1	0.37±0.06	0.37±0.06	5	250
		NTN	0.67	-84.1±3.3	-83.9±3.3	2	77.6±7.5	77.6±7.5	0	0.23±0.04	0.23±0.04	0	550
		AIRMoN	0.76	-82.3±12.2	-82.2±12.2	1	79.9±11.6	79.8±11.6	0	0.10±0.16	0.10±0.16	2	43
	August	CAPMoN	0.52	-72.5±8.1	-72.3±8.1	2	129.4±21.9	129.6±21.9	0	0.20±0.06	0.20±0.06	3	257
		NTN	0.57	-83.1±6.6	-82.9±6.6	1	177.3±74.3	177.3±74.3	0	0.22±0.04	0.22±0.04	4	717
		AIRMoN	0.58	-83.1±11.4	-83.0±11.4	0	71.3±7.5	71.4±7.5	0	0.01±0.16	0.01±0.16	1	39

Table S5. Same as Table S1, but for precipitation-chemistry concentration observations of sulfate.