Supplement of Geosci. Model Dev. Discuss., 8, 645–671, 2015
http://www.geosci-model-dev-discuss.net/8/645/2015/
doi:10.5194/gmdd-8-645-2015-supplement

Geoscientific Model Development
Discussions
Open Access

Supplement of

Development of $PM_{2.5}$ source impact spatial fields using a hybrid source apportionment air quality model

C. E. Ivey et al.

Correspondence to: C. E. Ivey (sunni.ivey@gmail.com)

Note A

During the chosen period of study, the CSN network performed thermal optical transmittance analyses to determine carbon species concentrations. As a result, organic and elemental carbon concentrations were converted to IMPROVE or total optical reflectance equivalents using previously derived methods (Malm et al. 2011). Additionally, measurements below detection limit are handled by setting the concentration to ½ of the measurement detection limit (MDL) and setting the measurement uncertainty to ⅔ of the MDL (Marmur et al. 2006).

Table S1. January 2004 domain-wide $PM_{2.5}$ emissions and uncertainties.

Source Categories	Emissions (metric tons/day)	Uncertainty (factor)	Rank
Agricultural Burning	323.5	± 5	12
Aircraft Emissions	25.1	± 1.5	25
Biogenic Emissions	0.0	± 1.5	33
Coal CM[ŧ]	1274.3	± 1.1	4
Diesel CM[ŧ]	2.0	± 1.3	30
Dust	5300.9	± 10	1
Fuel Oil CM[ŧ]	228.6	± 1.3	17
Livestock Emissions	1.3	± 1.3	31
Liquid Petroleum Gas CM[ŧ]	13.7	± 1.3	26
Lawn Waste Burning	759.9	± 5	6
Metal Processing	171.2	± 1.3	18
Meat Cooking	309.5	± 1.5	14
Mexican CM[ŧ]	13.2	± 1.5	27
Mineral Processing	315.9	± 1.3	13
Natural Gas CM[ŧ]	619.7	± 1.3	8
NR[§] Diesel CM[ŧ]	353.7	± 1.5	11
NR[§] Fuel Oil CM[ŧ]	33.8	± 1.5	24
NR[§] Gasoline CM[ŧ]	238.3	± 1.5	16
NR[§] Liquid Petroleum Gas CM[ŧ]	2.9	± 1.5	29
NR[§] Natural Gas CM[ŧ]	0.3	± 1.5	32
Other NR Sources	5.8	± 1.5	28
Open Fires	398.5	± 5	9
Onroad Diesel CM[ŧ]	358.3	± 1.5	10
Onroad Gasoline CM[ŧ]	159.7	± 1.5	20
Other CM[ŧ] Sources	169.6	± 1.5	19
Other PM Sources	785.0	± 1.5	5
Prescribed Burning	725.3	± 10	7
Railroad Emissions	64.4	± 1.5	21
Seasalt	1893.4	± 1.5	3
Solvent Emissions	61.3	± 1.5	22
Wildfires	45.7	± 10	23
Woodfuel Burning	245.8	± 1.5	15
Woodstoves	3407.4	± 5	2

[§]NR = Nonroad, [ŧ]CM = Combustion

Table S2. Median and mean hybrid and hybrid-kriging adjustment factors for withheld CSN receptors

Source Categories	$\widetilde{R_{Hyb}}$	$\widetilde{R_{HK}}$	$\overline{R_{Hyb}}$	$\overline{R_{HK}}$	**RMSE**
Agricultural Burning	0.881	0.778	0.669	0.683	0.316
Aircraft Emissions	1.000	1.000	0.996	0.997	0.022
Biogenic Emissions	1.001	1.002	0.986	0.988	0.126
Coal CM[ǂ]	0.954	0.966	0.937	0.941	0.074
Diesel CM[ǂ]	1.000	1.000	1.000	1.000	0.004
Dust	0.100	0.105	0.131	0.133	0.185
Fuel Oil CM[ǂ]	0.985	0.969	0.956	0.956	0.100
Livestock Emissions	1.001	0.999	0.990	0.994	0.094
Liquid Petroleum Gas CM[ǂ]	1.000	1.000	0.999	0.999	0.009
Lawn Waste Burning	0.100	0.107	0.285	0.156	1.738
Metal Processing	0.778	0.742	0.677	0.687	0.322
Meat Cooking	0.985	0.973	0.931	0.937	0.168
Mexican CM[ǂ]	1.000	1.000	0.999	1.001	0.017
Mineral Processing	0.875	0.874	0.857	0.845	0.165
Natural Gas CM[ǂ]	0.604	0.620	0.599	0.591	0.269
NR[§] Diesel CM[ǂ]	1.001	0.995	0.964	0.979	0.132
NR[§] Fuel Oil CM[ǂ]	0.999	0.996	0.985	0.990	0.055
NR[§] Gasoline CM[ǂ]	1.000	0.998	0.963	0.975	0.139
NR[§] Liquid Petroleum Gas CM[ǂ]	1.000	1.000	0.997	0.998	0.017
NR[§] Natural Gas CM[ǂ]	1.000	1.000	0.999	1.000	0.007
Other NR Sources	1.000	1.000	1.000	0.999	0.005
Open Fires	0.704	0.652	0.584	0.590	0.270
Onroad Diesel CM[ǂ]	0.983	0.976	0.944	0.960	0.115
Onroad Gasoline CM[ǂ]	0.873	0.884	0.826	0.857	0.285
Other CM[ǂ] Sources	0.954	0.911	0.874	0.867	0.254
Other PM Sources	0.667	0.654	0.632	0.641	0.279
Prescribed Burning	0.959	0.942	1.047	0.894	1.930
Railroad Emissions	1.001	0.998	0.992	0.993	0.046
Seasalt	0.995	0.993	0.988	0.986	0.034
Solvent Emissions	0.943	0.920	0.843	0.852	0.262
Wildfires	0.866	0.866	0.791	0.787	0.250
Woodfuel Burning	0.953	0.939	0.854	0.881	0.264
Woodstoves	0.100	0.128	0.322	0.184	1.768

[§]NR = Nonroad, [ǂ]CM = Combustion

Table S3. Mean observations and model simulations for January 2004 for withheld CSN monitors.

	OBS		CMAQ-DDM		HYB		SH	
	Avg.	**Std.**	**Avg.**	**Std.**	**Avg.**	**Std.**	**Avg.**	**Std.**
$PM_{2.5}$	11.7	8.3	16.3	11	8.59	4.7	9.20	5.7
OC	2.05	2.1	3.28	3.3	1.23	0.84	1.39	1.1
EC	0.727	0.64	0.944	1.3	0.55	0.59	0.627	0.90
NO_3	2.96	3.8	2.05	2.3	1.87	2.0	1.87	2.1
NH_4	1.38	1.3	1.45	0.99	1.20	0.79	1.24	0.83
SO_4	2.07	1.4	2.78	1.9	2.32	1.6	2.38	1.7
Na	0.0602	0.069	0.0960	0.079	0.0339	0.019	0.0394	0.028
Mg	0.0127	0.0017	0.0255	0.020	0.0960	0.061	0.0111	0.091
Al	0.0149	0.015	0.187	0.16	0.0397	0.022	0.0451	0.029
Si	0.0814	0.066	0.568	0.50	0.113	0.070	0.149	0.017
P	0.0046	0.0019	0.0064	0.0054	0.0021	0.0011	0.0026	0.0018
Cl	0.0436	0.10	0.369	0.33	0.0798	0.056	0.0931	0.077
K	0.0725	0.068	0.429	0.46	0.0738	0.062	0.0946	0.13
Ca	0.0407	0.060	0.182	0.14	0.0445	0.023	0.0501	0.030
V	0.022	0.0030	0.0015	0.0014	4.64 E-4	3.1E-4	5.68E-4	5.0E-4
Fe	0.0707	0.16	0.160	0.14	0.0368	0.022	0.0447	0.036
Cu	0.0027	0.0034	0.0035	0.053	0.0015	0.0011	0.0020	0.0022
Zn	0.0209	0.070	0.0099	0.013	0.0029	0.0021	0.0035	0.0032
Se	0.0018	0.0016	0.0017	0.011	0.0012	7.1E-4	0.0012	7.7E-4
Pb	0.0054	0.011	0.0019	0.0022	5.60E-4	3.8E-4	7.42E-4	7.8E-4

Note: All averages and standard deviations are expressed in µg m^{-3}.

Table S4. Mean observations and model simulations for January 2004 for SEARCH monitors.

	OBS		**CMAQ-DDM**		**HYB**	
	Avg	Std.	Avg	Std.	Avg	Std.
$PM_{2.5}$	11.2	5.3	14.7	9.8	9.04	5.3
OC	2.81	2.4	3.05	2.8	1.66	1.5
EC	0.831	0.81	0.711	0.76	0.457	0.48
NO_3	1.08	0.78	1.01	1.5	0.927	1.4
NH_4	1.12	0.51	1.22	0.92	1.01	0.70
SO_4	2.73	1.6	3.78	2.3	3.30	2.0
Al	0.0062	0.0058	0.143	0.12	0.0342	0.027
Si	0.0292	0.026	0.388	0.28	0.0936	0.064
K	0.0600	0.027	0.279	0.24	0.0504	0.035
Ca	0.0281	0.031	0.141	0.11	0.0396	0.032
Fe	0.0368	0.044	0.132	0.14	0.0328	0.031
Cu	0.0013	0.0018	0.0043	0.0077	0.0016	0.0017
Zn	0.0157	0.030	0.0108	0.013	0.0036	0.0028
Se	8.00E-04	8.00E-04	0.0013	0.0013	9.00E-04	9.00E-04
Pb	0.0027	0.0041	0.0020	0.0033	5.00E-04	7.00E-04

Note: All averages and standard deviations are expressed in $\mu g\ m^{-3}$.

Table S5. Mean observations and model simulations for January 2004 for IMPROVE monitors.

	OBS		CMAQ-DDM		HYB	
	Avg	Std.	Avg	Std.	Avg	Std.
$PM_{2.5}$	6.25	3.8	11.51	8.2	6.63	4.4
OC	1.39	1.2	2.33	2.3	0.959	1.2
EC	0.298	0.22	0.532	0.52	0.283	0.30
NO_3	1.25	1.4	1.41	1.6	1.23	1.4
NH_4	0.894	0.40	0.984	0.66	0.870	0.57
SO_4	1.64	1.1	2.43	1.7	2.09	1.4
Al	0.0128	0.017	0.103	0.099	0.0268	0.022
Si	0.0629	0.039	0.297	0.27	0.0781	0.067
K	0.0416	0.027	0.291	0.30	0.0615	0.060
Ca	0.0218	0.015	0.101	0.094	0.0297	0.024
Fe	0.0198	0.021	0.0860	0.094	0.0234	0.021
Cu	7.00E-04	6.0E-04	0.0020	0.0043	0.0011	0.0019
Zn	0.00840	0.0078	0.0085	0.010	0.0035	0.0066
Se	9.00E-04	0.001	0.0011	0.0013	8.00E-04	8.0E-04
Pb	0.00190	0.0024	0.0011	0.0016	4.00E-04	5.0E-04

Table S6. Linear regression and correlation coefficients for model simulations vs. observations for January 2004 for withheld CSN monitors. Regression equation: $\boldsymbol{Conc_{model} = \alpha + \beta \cdot Conc_{obs}}$

	CMAQ-DDM vs. OBS					**HYB vs. OBS**					**SH vs. OBS**				
	α	SE_α	**β**	SE_β	**r**	**α**	SE_α	**β**	SE_β	**r**	**α**	SE_α	**β**	SE_β	**r**
$PM_{2.5}$	11.2	4.3	0.43	0.30	0.321	5.5	1.7	0.27	0.12	0.470	6.4	2.1	0.24	0.15	0.350
OC	2.49	1.0	0.39	0.36	0.242	0.73	0.22	0.25	0.076	0.605	1.01	0.33	0.18	0.11	0.351
EC	0.61	0.43	0.46	0.45	0.234	0.34	0.20	0.29	0.20	0.314	0.41	0.31	0.30	0.32	0.217
NO_3	1.3	0.63	0.25	0.13	0.409	1.1	0.54	0.25	0.11	0.454	1.2	0.55	0.25	0.12	0.440
NH_4	0.98	0.30	0.34	0.16	0.454	0.78	0.23	0.31	0.12	0.516	0.81	0.24	0.31	0.13	0.492
SO_4	0.77	0.56	0.97	0.23	0.706	0.48	0.46	0.89	0.19	0.744	0.55	0.48	0.89	0.19	0.730
Na	0.11	0.024	-0.17	0.27	-0.145	0.032	0.0059	0.030	0.065	0.110	0.040	0.0087	-0.016	0.096	-0.039
Mg	0.025	0.0057	0.038	0.27	0.033	0.0091	0.0018	0.039	0.084	0.109	0.011	0.0027	0.025	0.13	0.046
Al	0.16	0.050	1.86	2.4	0.182	0.033	0.0070	0.44	0.33	0.301	0.041	0.0093	0.27	0.44	0.145
Si	0.37	0.18	2.46	1.7	0.320	0.079	0.024	0.42	0.23	0.392	0.13	0.061	0.20	0.59	0.078
P	0.0081	0.0033	-0.36	0.67	-0.124	0.0024	6.90E-4	-0.058	0.14	-0.098	0.0035	0.011	-0.21	0.22	-0.216
Cl	0.35	0.081	0.46	0.72	0.147	0.065	0.011	0.33	0.098	0.620	0.086	0.019	0.16	0.17	0.221
K	0.42	0.16	0.11	1.6	0.016	0.041	0.019	0.45	0.19	0.496	0.098	0.043	-0.041	0.43	-0.022
Ca	0.13	0.0035	1.22	0.48	0.513	0.038	0.0060	0.16	0.083	0.420	0.044	0.008	0.16	0.11	0.316
V	0.0015	4.0E-4	-3.77E-4	0.11	-8.0E-4	4.25E-4	8.8E-5	0.018	0.024	0.173	5.45E-4	1.44E-4	0.011	0.039	0.063
Fe	0.15	0.035	0.16	0.20	0.180	0.034	0.0053	0.046	0.031	0.326	0.042	0.0090	0.038	0.053	0.166
Cu	0.0036	0.0016	-0.022	0.37	-0.014	0.0013	3.2E-4	0.078	0.074	0.238	0.0018	6.4E-4	0.071	0.15	0.110
Zn	0.0099	0.0033	4.8E-4	0.045	0.003	0.0029	5.2E-4	0.0011	0.0072	0.037	0.0034	7.4E-4	0.0032	0.011	0.070
Se	0.0015	3.9E-4	0.082	0.16	0.116	9.8E-4	2.5E-4	0.11	0.10	0.235	0.0010	2.7E-4	0.11	0.11	0.225
Pb	0.0017	5.7E-4	0.027	0.046	0.135	5.3E-4	9.7E-5	0.0052	0.0078	0.153	6.13E-4	7.9E-4	0.024	0.015	0.345

Table S7. Linear regression and correlation coefficients for model predictions vs. observations for January 2004 for SEARCH monitors. Regression equation: $Conc_{model} = \alpha + \beta \cdot Conc_{obs}$

	CMAQ-DDM vs. OBS					SH vs. OBS				
	α	SE_α	β	SE_β	r	α	SE_α	β	SE_β	r
$PM_{2.5}$	6.40	5.2	0.746	0.42	0.404	6.67	2.7	0.332	0.22	0.357
OC	1.72	0.93	0.473	0.25	0.413	1.18	0.47	0.264	0.13	0.445
EC	0.353	0.23	0.431	0.20	0.462	0.263	0.14	0.307	0.12	0.519
NO_3	0.222	0.59	0.730	0.44	0.371	0.497	0.59	0.604	0.44	0.315
NH_4	0.760	0.53	0.411	0.43	0.227	0.827	0.40	0.294	0.32	0.216
SO_4	0.794	0.74	1.10	0.23	0.752	0.840	0.55	0.978	0.17	0.807
Al	0.0882	0.037	8.86	4.4	0.446	0.028	0.0097	1.89	1.1	0.379
Si	0.214	0.087	5.95	2.2	0.556	0.0755	0.022	1.12	0.58	0.437
K	0.135	0.14	2.40	2.1	0.271	0.0412	0.020	0.253	0.31	0.200
Ca	0.0820	0.031	2.10	0.74	0.581	0.0281	0.0095	0.613	0.23	0.559
Fe	0.0395	0.028	2.52	0.49	0.788	0.0206	0.0086	0.473	0.15	0.615
Cu	7.00E-04	0.0018	2.85	0.81	0.657	0.0012	5.00E-04	0.507	0.24	0.470
Zn	0.0088	0.0033	0.130	0.098	0.313	0.0035	7.00E-04	0.0296	0.022	0.323
Se	0.0012	4.00E-04	0.0946	0.39	0.060	0.001	3.00E-04	0.0657	0.28	0.060
Pb	0.0013	9.00E-04	0.265	0.19	0.335	5.00E-04	2.00E-04	0.0593	0.042	0.330

Table S8. Linear regression and correlation coefficients for model predictions vs. observations for January 2004 for IMPROVE monitors. Regression equation: $\boldsymbol{Conc_{model} = \alpha + \beta \cdot Conc_{obs}}$

	CMAQ-DDM vs. OBS					SH vs. OBS				
	α	SE_α	**β**	SE_β	**r**	**α**	SE_α	**β**	SE_β	**r**
$PM_{2.5}$	3.53	1.4	1.28	0.19	0.590	1.96	0.72	0.747	0.098	0.637
OC	1.11	0.35	0.877	0.19	0.456	0.359	0.18	0.432	0.097	0.439
EC	0.189	0.084	1.15	0.23	0.479	0.0862	0.049	0.661	0.13	0.472
NO_3	0.431	0.17	0.782	0.092	0.684	0.346	0.15	0.712	0.081	0.696
NH_4	0.369	0.31	0.689	0.32	0.422	0.355	0.27	0.576	0.28	0.406
SO_4	0.617	0.25	1.11	0.13	0.692	0.451	0.20	0.997	0.10	0.728
Al	0.0894	0.013	1.05	0.62	0.182	0.023	0.0029	0.295	0.13	0.232
Si	0.180	0.054	1.86	0.73	0.268	0.0554	0.014	0.361	0.18	0.208
K	0.194	0.059	2.33	1.2	0.209	0.0274	0.011	0.820	0.22	0.370
Ca	0.0387	0.016	2.83	0.62	0.448	0.0155	0.0043	0.648	0.16	0.400
Fe	0.0455	0.012	2.05	0.42	0.466	0.0143	0.0028	0.461	0.095	0.469
Cu	3.00E-04	7.0E-04	2.42	0.78	0.319	5.00E-04	3.00E-04	0.842	0.35	0.256
Zn	0.0049	0.002	0.434	0.13	0.339	0.002	0.001	0.182	0.090	0.214
Se	7.00E-04	2.0E-04	0.403	0.13	0.321	5.00E-04	1.00E-04	0.330	0.083	0.399
Pb	8.00E-04	2.0E-04	0.143	0.075	0.204	3.00E-04	1.00E-04	0.0305	0.022	0.151

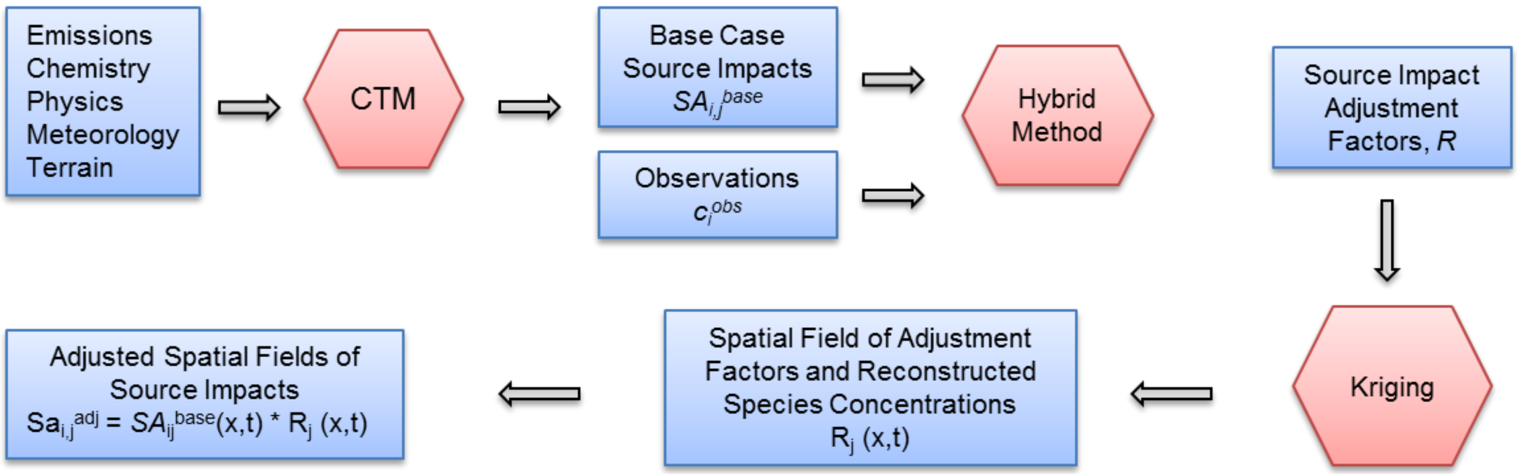

Figure S1. Flowchart of the inputs for the spatial hybrid source apportionment method. Hybrid adjustment factors are kriged to produce spatial fields of improved source impact estimates.

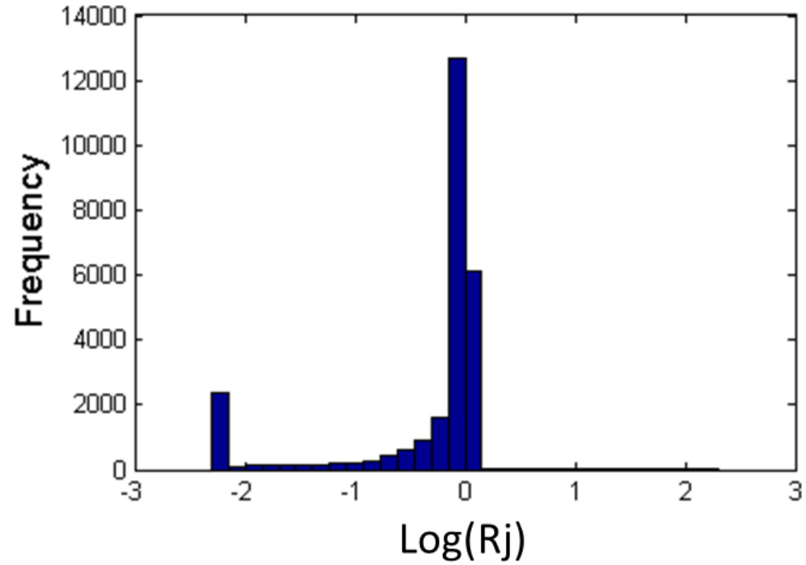

Figure S2. Distribution of Rj values generated by hybrid analysis of CSN data for January 2004 (n = 26,400; 0.1 < Rj < 10). Note that the x-axis is on a log scale.

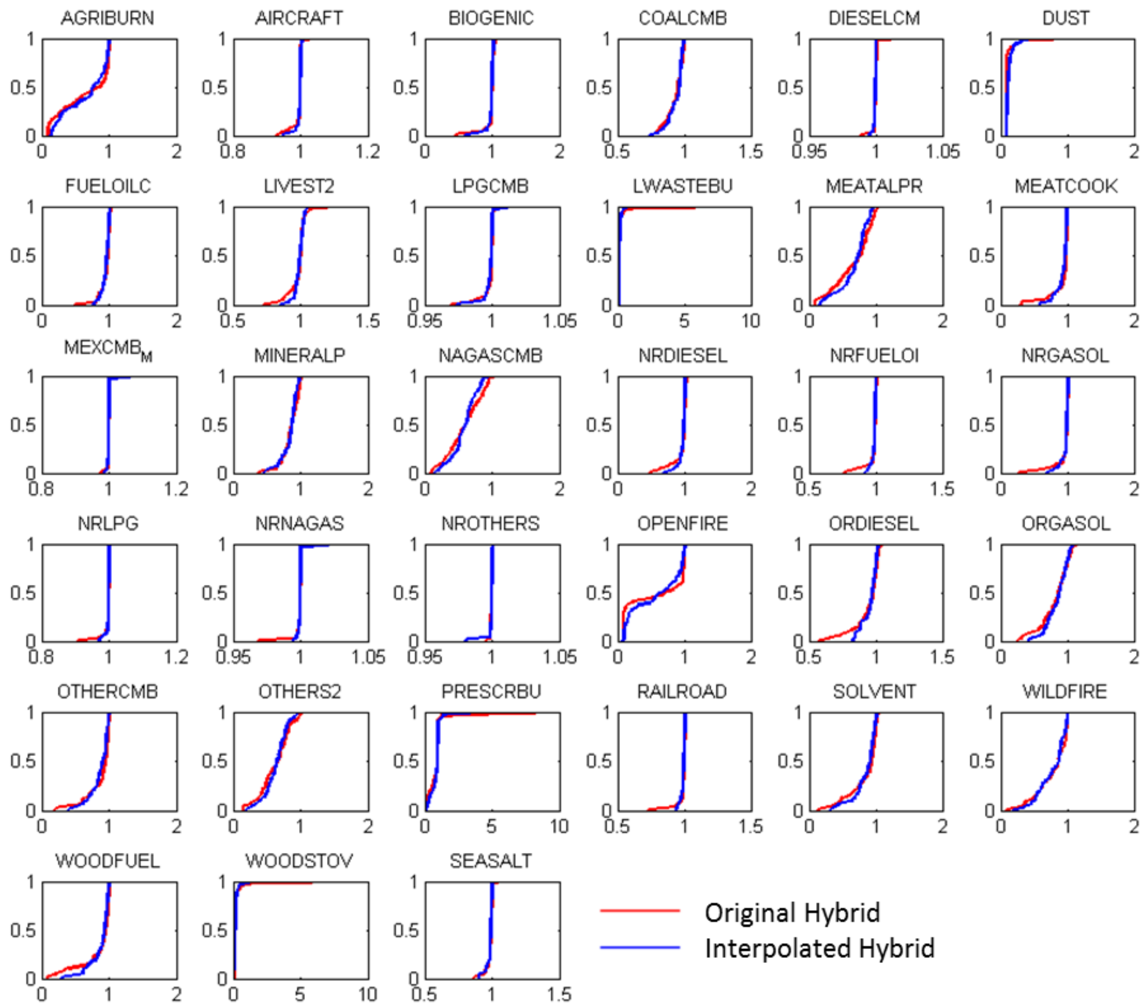

Figure S3. Cumulative distributions of original and interpolated hybrid adjustment factors for witheld CSN monitors.

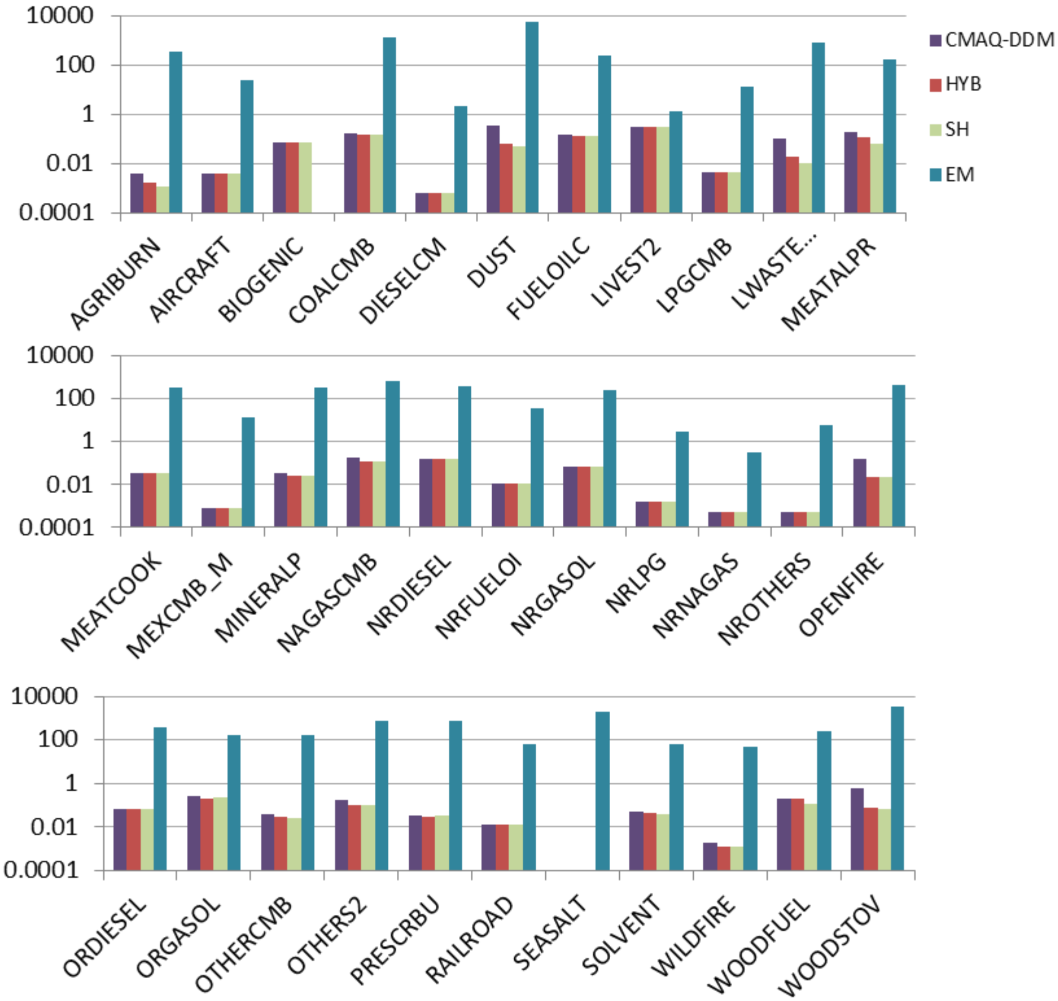

Figure S4. Average source impacts (µg m-3) at withheld CSN receptors for January 2004 for CMAQ-DDM, hybrid (HYB), and spatial hybrid (SH) applications. Also shown are the domain-wide PM2.5 emissions (EM) for each source (metric tons per day).

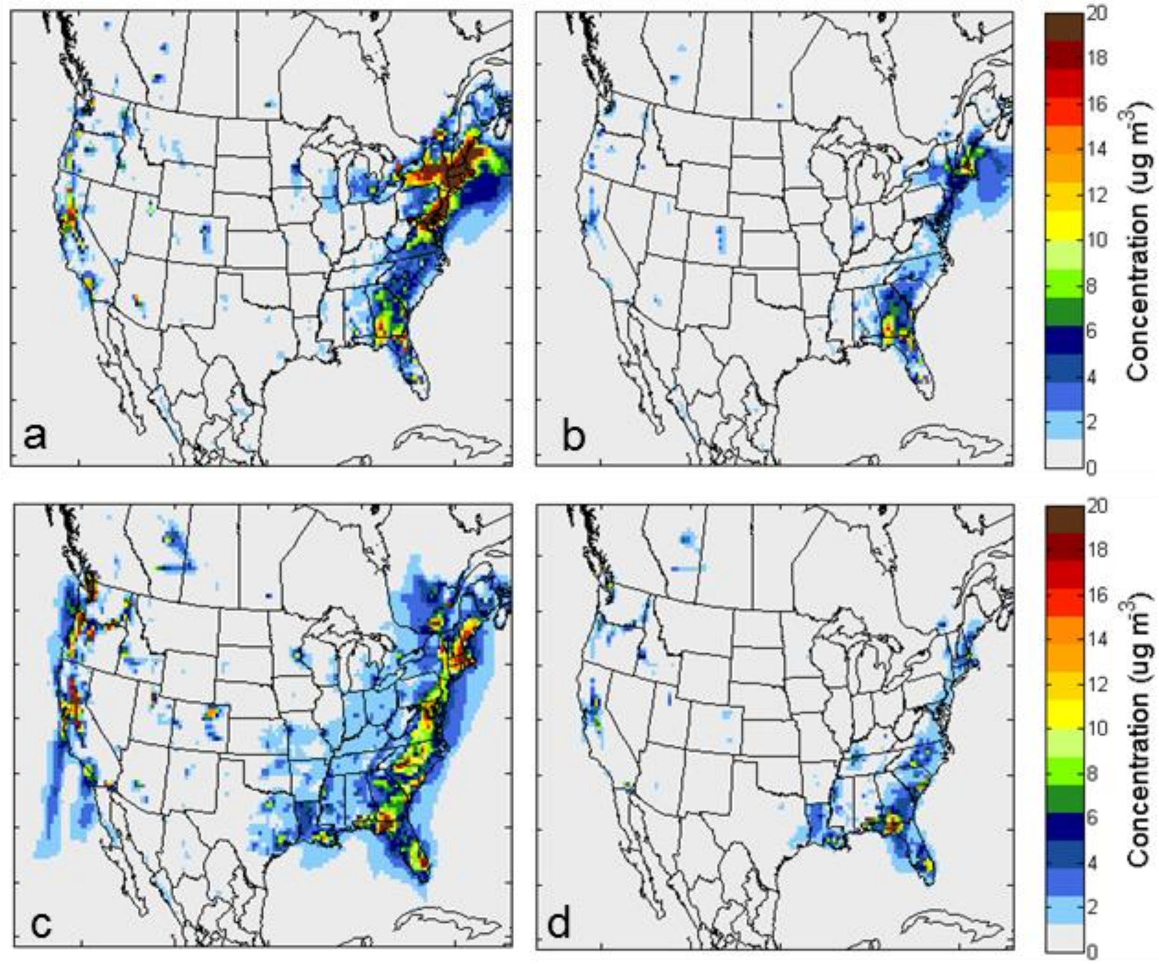

Figure S5. Hybrid adjustment of biomass burning impacts on $PM_{2.5}$ on January 4th and 22nd in 2004. Biomass burning fields are produced by aggregating source impacts from agricultural burning, lawn waste burning, open fires, prescribed burning, wildfires, woodfuel and woodstove burning. (a) CMAQ-DDM spatial field for January 4th. (b) SH spatial field for January 4th. (c) CMAQ-DDM spatial field for January 22nd. (d) SH spatial field for January 22nd.

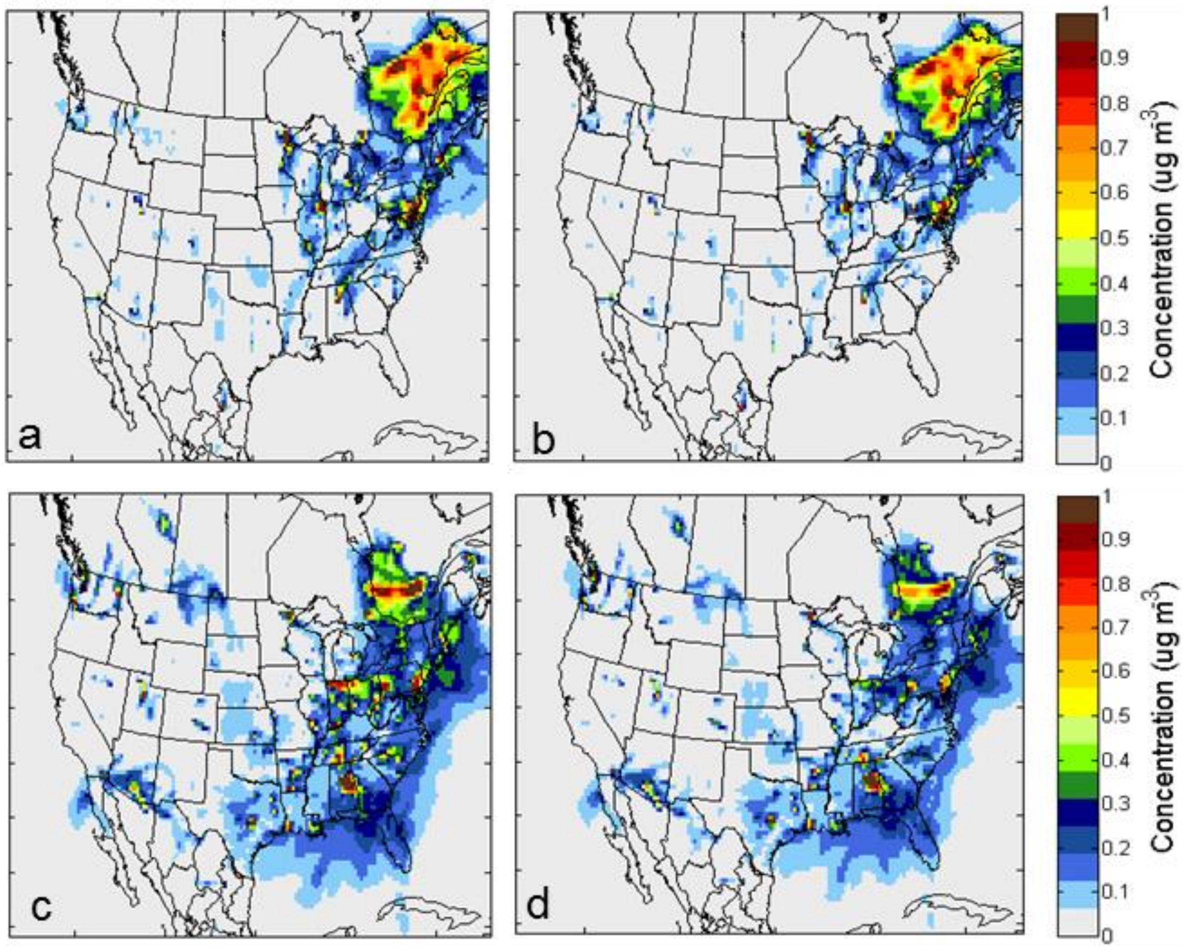

Figure S6. Hybrid adjustment of metals processing impacts on $PM_{2.5}$ on January 4th and 22nd in 2004. (a) CMAQ-DDM spatial field for January 4th. (b) SH spatial field for January 4th. (c) CMAQ-DDM spatial field for January 22nd. (d) SH spatial field for January 22nd.

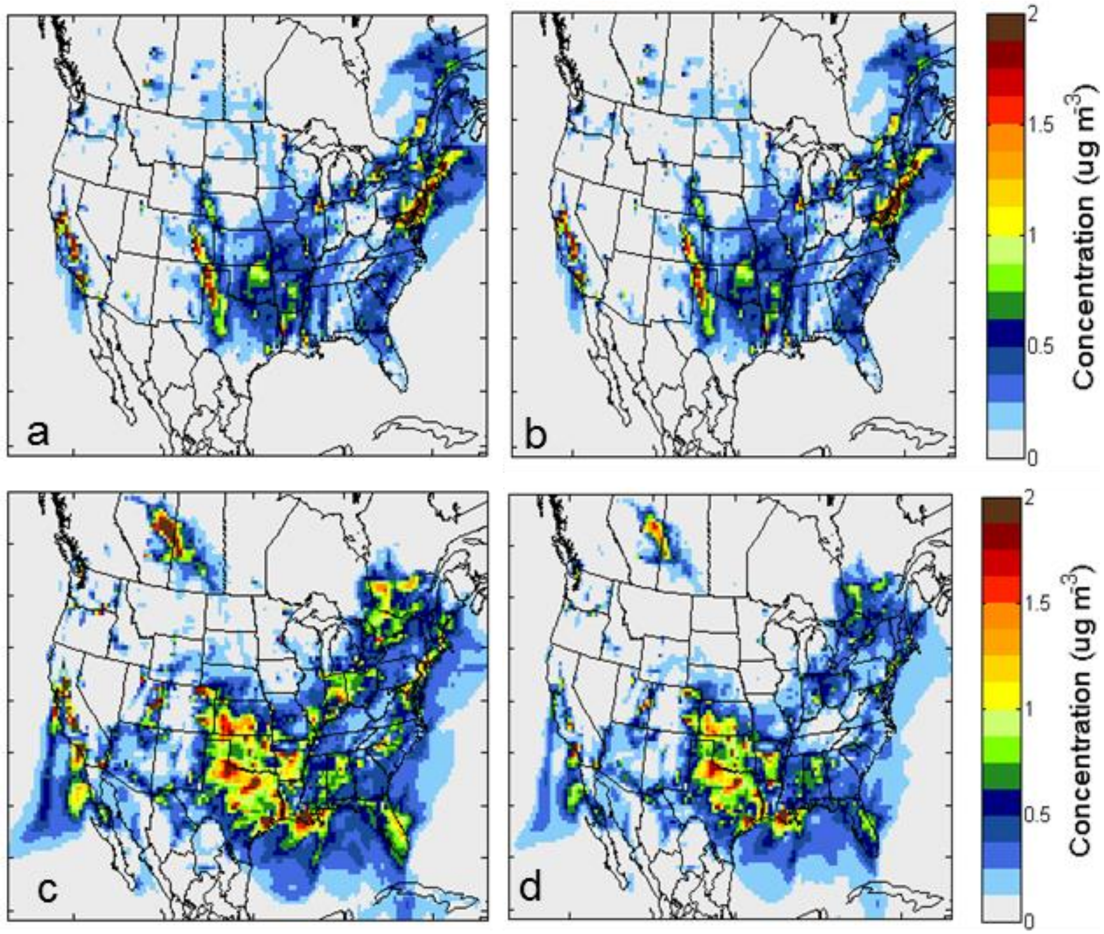

Figure S7. Hybrid adjustment of natural gas combustion (point and area sources) source impact fields on January 4th and 22nd in 2004. (a) CMAQ-DDM spatial field for January 4th. (b) SH spatial field for January 4th. (c) CMAQ-DDM spatial field for January 22nd. (d) SH spatial field for January 22nd.

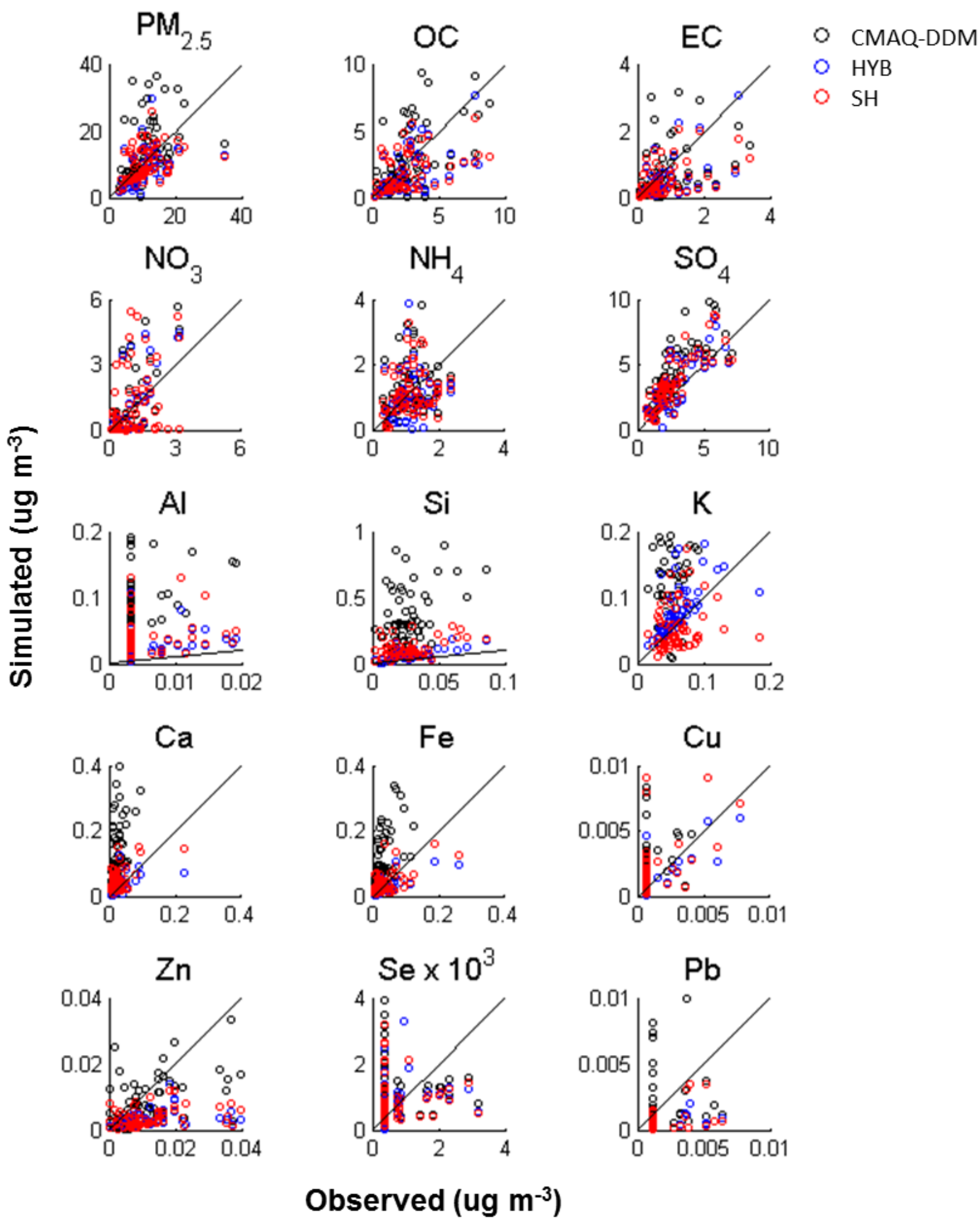

Figure S8. Simulated concentrations vs. observations for withheld CSN receptors (n = 75). Note that many of the metals are set at their measurement detection limit, hence their poor correlation. Plots are drawn with the 1-to-1 line.

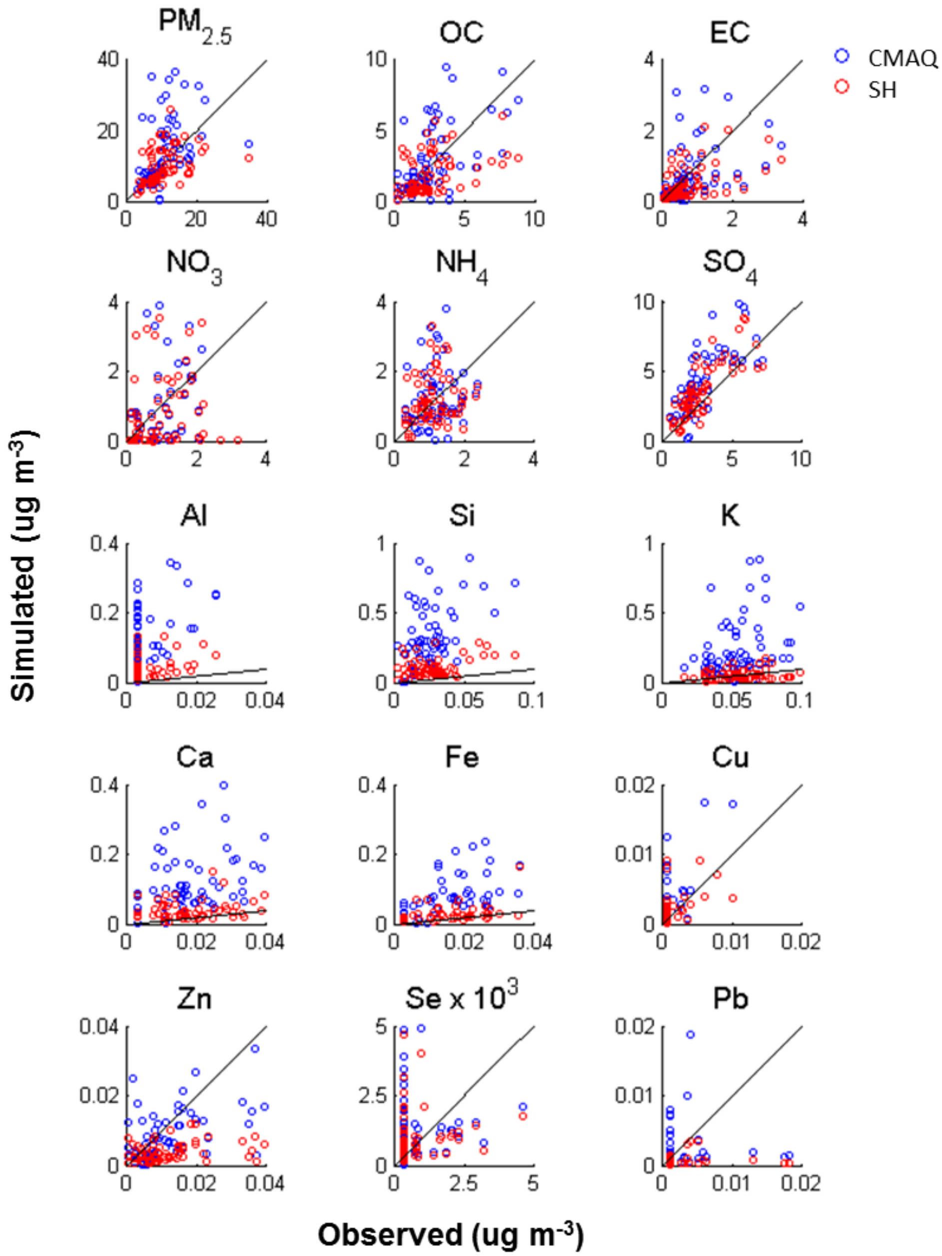

Figure S9. Simulated concentrations vs. observations for SEARCH receptors. Plots are drawn with the 1-to-1 line.

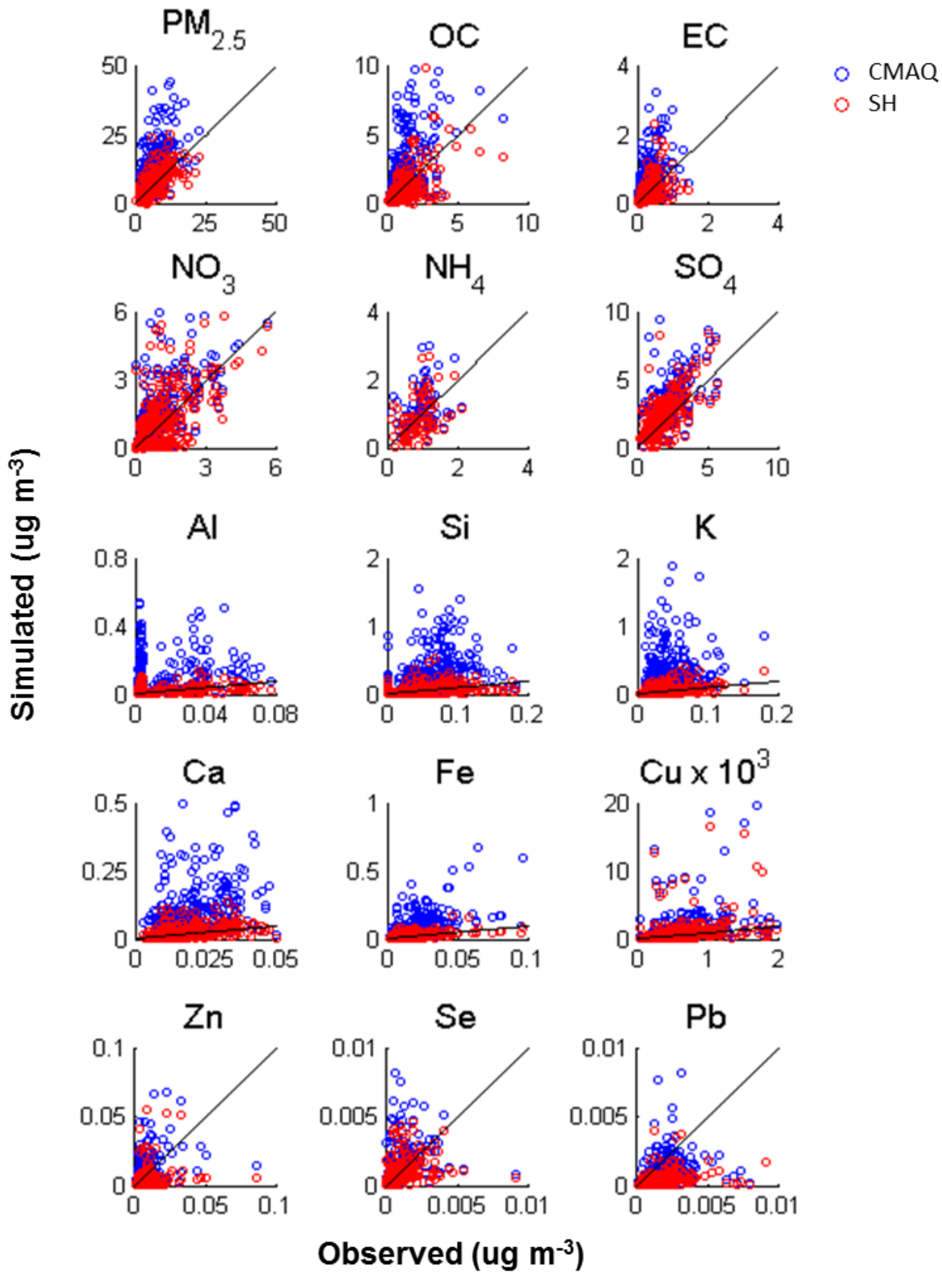

Figure S10. Simulated concentrations vs. observations for IMPROVE receptors. Plots are drawn with the 1-to-1 line.