



Supplement of

Effects of emission reductions on organic aerosol in the southeastern United States

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Table S1. Correlations (r^2) among mean OC, EC, and SO₄ based on annual, seasonal, and monthly averaging periods and on daily measurements. Summer is defined as June through August, winter is December through February, autumn is September through November, and spring is March through May.

Site	Pair	Annual (n=15)	Seasonal (n=60)	Monthly (n=180)	Monthly- Winter (n=45)	Monthly- Summer (n=45)	Daily All (n = 1382- 3993)	Daily 2009-13 (n = 110 - 558)	Daily- Jan (n = 106 - 338)	Daily- July (n = 150 - 345)
BHM	OC-EC	0.931	0.856	0.803	0.841	0.696	0.719	0.692	0.836	0.553
BHM	OC-SO ₄	0.843	0.425	0.331	0.305	0.690	0.211	0.127	0.057	0.514
BHM	EC-SO ₄	0.834	0.349	0.258	0.219	0.603	0.135	0.046	0.064	0.362
CTR	OC-EC	0.736	0.470	0.508	0.653	0.402	0.648	0.602	0.805	0.533
CTR	OC-SO ₄	0.772	0.492	0.350	0.293	0.360	0.181	0.181	0.024	0.520
CTR	EC-SO ₄	0.846	0.317	0.223	0.311	0.428	0.166	0.104	0.051	0.561
GFP	OC-EC	0.894	0.749	0.718	0.689	0.711	0.680	0.708	0.712	0.541
GFP	OC-SO ₄	0.730	0.181	0.158	0.086	0.554	0.166	0.112	0.057	0.361
GFP	EC-SO ₄	0.801	0.054	0.054	0.146	0.487	0.084	0.086	0.097	0.189
JST	OC-EC	0.910	0.762	0.706	0.756	0.537	0.645	0.668	0.661	0.343
JST	OC-SO ₄	0.854	0.276	0.191	0.350	0.552	0.099	0.084	0.144	0.365
JST	EC-SO ₄	0.876	0.200	0.153	0.315	0.680	0.076	0.019	0.174	0.335
OAK	OC-EC	0.731	0.666	0.647	0.904	0.397	0.842	0.594	0.964	0.605
OAK	OC-SO ₄	0.702	0.110	0.129	0.257	0.523	0.060	0.066	0.016	0.402
OAK	EC-SO ₄	0.591	0.004	0.021	0.332	0.483	0.052	0.037	0.026	0.359
OLF	OC-EC	0.840	0.701	0.574	0.673	0.517	0.590	0.684	0.748	0.520
OLF	OC-SO ₄	0.721	0.361	0.316	0.321	0.651	0.238	0.103	0.100	0.559
OLF	EC-SO ₄	0.875	0.349	0.223	0.239	0.800	0.168	0.036	0.121	0.342
PNS	OC-EC	0.930	0.858	0.767	0.770	0.539	0.724	0.902	0.779	0.490
PNS	OC-SO ₄	0.568	0.006	0.030	0.089	0.700	0.088	0.043	0.024	0.569
PNS	EC-SO ₄	0.592	0.000	0.002	0.059	0.679	0.041	0.050	0.024	0.307
YRK	OC-EC	0.839	0.636	0.546	0.774	0.489	0.554	0.651	0.705	0.413
YRK	OC-SO ₄	0.800	0.619	0.451	0.454	0.568	0.304	0.215	0.104	0.502
YRK	EC-SO ₄	0.764	0.414	0.293	0.453	0.782	0.253	0.097	0.096	0.391

14 Table S2. PCA1 and PCA2 orthogonal factors for SEARCH BHM daily data. PCA1: n =
 15 364, variance explained = 78%. PCA2: n = 1513, variance explained = 77%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.817	0.058	0.12	0.171	0.164	0.156	0.001	0.288
CO_max1	0.619	0.144	0.058	0.284	0.16	0.062	-0.039	0.323
NO _x	0.806	0.069	0.285	0.065	-0.034	0.295	-0.029	0.188
EC	0.744	0.068	-0.091	0.321	0.16	0.155	0.051	0.137
OC	0.784	0.063	-0.159	0.238	0.315	0.087	0.122	0.073
NO _z	0.409	0.062	0.127	-0.011	-0.007	-0.044	0.021	0.631
K	0.223	0.054	-0.035	-0.019	0.128	0.005	0.814	0.095
Al	0.106	0.949	-0.036	-0.063	0.006	0.078	0.06	0.016
Si	0.135	0.86	-0.069	0.116	0.048	0.3	0.105	0.21
Fe	0.394	0.373	0.186	0.002	0.043	0.677	-0.056	-0.135
Ca	0.237	0.199	-0.02	0.256	0.274	0.17	0.19	0.552
O ₃ _max8	0.282	0.062	-0.577	0.197	0.486	-0.112	-0.055	0.08
NH ₃	0.552	0.129	-0.395	0.087	0.284	0.078	0.101	0.022
NO ₃	0.144	-0.044	0.833	0.028	0.122	0.06	-0.048	0.108
SO ₂ _max1	0.296	-0.012	-0.055	0.861	0.168	0.01	0.037	0.08
SO ₂	0.389	-0.002	-0.013	0.828	0.187	0.086	0.012	0.101
NH ₄	0.198	-0.03	0.117	0.122	0.945	0.017	0.018	0.047
SO ₄	0.111	0.056	-0.088	0.137	0.929	0.07	0.108	0.072
Na	-0.108	0.255	0.289	0.376	-0.051	-0.026	0.417	-0.266
Cl	0.649	-0.034	0.306	-0.001	-0.178	0.221	0.306	-0.106
Mg	-0.12	0.184	-0.116	0.161	0.013	0.527	0.544	0.426
Cu	0.399	0.137	0.072	-0.083	0.068	0.761	0.081	-0.124
Zn	0.083	0.052	-3.25E-04	0.106	0.001	0.858	0.015	0.188
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.91	0.054	-0.127	0.14	0.062	0.212		
CO_max1	0.815	0.12	-0.066	0.188	0.053	0.109		
NO _x	0.81	0.035	-0.264	0.116	-0.169	0.306		
EC	0.758	0.082	0.094	0.228	0.136	0.28		
OC	0.745	0.068	0.195	0.148	0.303	0.204		
NO _z	-0.159	0.07	0.02	0.085	0.493	0.1		
nsK	0.7	0.312	0.054	0.177	0.145	-0.043		
Al	-0.036	0.921	0.105	-0.038	-0.036	-0.017		
Si	0.309	0.894	0.075	0.109	0.028	0.126		
Fe	0.289	0.566	-0.029	0.104	0.04	0.653		
Ca	0.334	0.557	0.033	0.119	0.134	0.428		
O ₃ _max8	0.267	0.086	0.78	0.101	0.422	-0.01		
O ₃ _24hr	-0.042	0.004	0.823	0.019	0.39	-0.091		
NO ₃	0.197	-0.077	-0.751	0.04	0.263	0.009		
SO ₂ _max1	0.307	0.062	0.038	0.908	0.121	0.058		
SO ₂	0.482	0.088	-0.001	0.793	0.196	0.129		
NH ₄	0.285	0.002	0.05	0.075	0.905	-0.04		
SO ₄	0.26	0.072	0.204	0.081	0.854	-0.002		
Cu	0.145	-0.02	-0.031	-0.012	-0.018	0.72		
Zn	0.224	0.14	-0.047	0.112	0.083	0.764		

17 Table S3. PCA1 and PCA2 orthogonal factors for SEARCH CTR daily data. PCA1: n =
18 383, variance explained = 79%. PCA2: n = 1258, variance explained = 79%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.742	-0.14	-0.118	0.194	0.205	0.238	-0.132	
CO_max1	0.669	-0.066	-0.158	0.144	0.177	0.049	-0.108	
NO _x	0.238	-0.094	-0.531	0.453	-0.152	0.361	-0.197	
EC	0.794	-0.017	0.132	0.111	0.145	0.141	-0.081	
OC	0.784	0.169	0.348	0.011	0.196	-0.002	-0.037	
NO _z	0.566	-0.016	0.055	0.487	0.19	0.342	-0.053	
K	0.769	0.085	0.095	-0.064	0.017	0.021	0.163	
Al	-0.037	0.951	0.018	-0.103	-0.054	-0.127	0.077	
Si	-0.053	0.963	0.057	-0.07	-0.036	-0.109	0.092	
Fe	0.057	0.955	0.044	0.065	0.08	0.064	0.084	
Ca	0.088	0.51	0.065	0.231	0.256	0.199	0.186	
O ₃ _max8	0.395	-0.068	0.593	0.299	0.231	0.255	-0.117	
NH ₃	0.409	0.148	0.689	-0.025	-0.092	0.121	0.058	
NO ₃	0.27	-0.046	-0.535	0.308	-0.057	0.297	0.219	
SO ₂ _max1	0.041	0.01	0.011	0.902	0.066	-0.06	-0.076	
SO ₂	0.141	0.024	-0.102	0.917	0.142	0.051	-0.078	
NH ₄	0.305	0.011	0.014	0.145	0.893	0.117	-0.051	
SO ₄	0.254	0.091	0.077	0.073	0.927	0.01	-3.62E-04	
Na	-0.039	0.046	0.022	-0.025	0.018	-0.012	0.883	
Cl	-0.083	0.027	-0.085	-0.097	-0.023	0.084	0.86	
Mg	0.01	0.352	0.026	-0.065	-0.049	-0.109	0.791	
Cu	0.032	-0.004	0.068	-0.094	0.025	0.791	-0.033	
Zn	0.293	-0.042	-0.081	0.173	0.107	0.741	0.047	
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.682	-0.139	0.016	0.446	0.045	0.234		
CO_max1	0.715	-0.082	0.017	0.397	0.002	0.169		
NO _x	0.173	-0.074	-0.211	0.836	-0.025	0.039		
EC	0.728	-0.044	0.217	0.189	0.357	-0.012		
OC	0.765	0.057	0.311	-0.012	0.347	-0.105		
NO _z	0.37	-0.017	0.522	0.41	0.222	0.035		
nsK	0.741	0.26	0.19	0.023	-0.001	-0.056		
Al	-0.03	0.936	-0.133	-0.169	-0.061	-0.021		
Si	0.005	0.967	-0.039	-0.133	-0.024	-0.067		
Fe	0.063	0.952	0.026	0.078	0.077	0.081		
Ca	0.069	0.575	0.383	0.287	0.049	0.086		
O ₃ _max8	0.244	9.53E-07	0.862	-0.014	0.333	0.016		
O ₃ _24hr	0.204	-0.021	0.879	-0.089	0.269	0.016		
NO ₃	0.231	-0.021	-0.27	0.614	0.083	0.063		
SO ₂ _max1	0.038	0.012	0.2	0.808	0.058	-0.203		
SO ₂	0.103	-0.001	0.208	0.878	0.09	-0.162		
NH ₄	0.233	-0.007	0.281	0.166	0.897	-0.038		
SO ₄	0.137	0.031	0.289	0.038	0.917	-0.047		
Cu	0.009	0.025	-0.003	-0.163	-0.091	0.852		
Zn	0.243	0.053	0.18	0.587	0.11	0.496		

19 Table S4. PCA1 and PCA2 orthogonal factors for SEARCH GFP daily data. PCA1: n =
 20 100, variance explained = 79%. PCA2: n = 376, variance explained = 79%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.927	-0.083	0.17	0.005	0.044		-0.133	0.103
CO_max1	0.915	-0.035	0.112	-0.044	-0.045		-0.165	2.63E-04
NO _x	0.904	-0.015	-0.06	0.212	-0.162		-0.074	0.033
EC	0.753	-0.068	0.262	0.042	0.266		-0.21	0.251
OC	0.554	-0.066	0.389	0.111	0.201		-0.282	0.4
NO _z	0.45	0.087	0.421	0.031	0.306		-0.007	0.132
K	0.342	-0.083	0.35	-0.049	0.212		0.214	0.608
Al	-0.105	0.98	-0.001	-0.033	0.047		0.072	0.056
Si	-0.083	0.985	0.021	-0.029	0.041		0.05	0.06
Fe	0.124	0.98	0.02	0.032	0.039		0.033	0.043
Ca	0.059	0.195	-0.066	0.002	-0.051		-0.042	0.881
O ₃ _max8	0.202	0.019	0.747	-0.221	0.165		-0.291	-0.007
NH ₃	0.28	0.062	0.628	-0.023	-0.186		-0.047	0.043
NO ₃	0.52	-0.088	-0.152	0.049	0.433		0.245	0.286
SO ₂ _max1	0.044	-0.025	-0.065	0.94	-0.003		-0.12	-0.023
SO ₂	0.125	-0.024	-0.041	0.956	0.019		-0.021	0.019
NH ₄	0.17	-0.039	0.04	0.043	0.952		-0.078	0.023
SO ₄	-0.005	0.149	0.069	-0.002	0.931		-0.042	-0.006
Na	-0.077	0.087	0.068	0.034	-0.111		0.809	-0.081
Cl	-0.114	-0.028	-0.236	-0.109	0.053		0.826	0.046
Mg	-0.374	0.352	-0.075	-0.225	-0.039		0.62	0.185
Cu	0.448	0.071	-0.437	-0.114	-0.167		-0.176	0.038
Zn	0.744	0.007	0.135	0.38	0.08		-0.016	-0.058
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.913	-0.109	0.053	0.022	0.158	0.083		
CO_max1	0.93	-0.056	0.019	-0.013	0.014	0.057		
NO _x	0.853	-0.027	-0.279	0.212	-0.095	0.188		
EC	0.679	0.098	0.098	0.095	0.408	0.192		
OC	0.538	0.132	0.295	0.17	0.498	0.256		
NO _z	0.553	-0.017	0.098	0.196	0.16	-0.126		
nsK	0.126	0.413	0.003	-0.083	0.447	0.388		
Al	-0.087	0.971	-0.062	-0.034	-0.011	-0.069		
Si	-0.078	0.983	-0.035	-0.035	-0.012	-0.043		
Fe	0.009	0.972	-0.051	0.021	-0.005	0.048		
Ca	0.024	0.887	0.117	0.004	0.073	0.159		
O ₃ _max8	0.233	0.01	0.879	-0.088	0.287	0.056		
O ₃ _24hr	-0.053	-0.047	0.895	-0.132	0.247	-0.019		
NO ₃	0.369	0.009	-0.482	-0.142	0.449	0.159		
SO ₂ _max1	0.11	-0.019	-0.044	0.944	0.025	0.059		
SO ₂	0.209	-0.031	-0.109	0.92	0.093	0.049		
NH ₄	0.206	-0.064	0.174	0.081	0.901	-0.048		
SO ₄	0.05	0.025	0.284	0.078	0.849	-0.104		
Cu	-0.01	0.049	0.027	-0.027	-0.148	0.793		
Zn	0.198	-0.005	-0.046	0.14	0.122	0.655		

21 Table S5. PCA1 and PCA2 orthogonal factors for SEARCH JST daily data. PCA1: n =
 22 516, variance explained = 78%. PCA2: n = 2593, variance explained = 78%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.953	0.014	-0.012	0.122	0.015		0.08	0.039
CO_max1	0.88	0.018	0.057	0.096	-0.102		0.029	0.012
NO _x	0.93	0.047	-0.097	0.211	-0.109		0.064	0.051
EC	0.883	0.059	0.175	0.108	0.133		0.08	0.103
OC	0.741	0.031	0.226	0.015	0.279		0.314	0.035
NO _z	0.447	0.02	0.373	-0.041	0.16		-0.059	0.27
K	0.346	0.064	0.071	-0.022	0.122		0.748	0.011
Al	-0.065	0.954	-0.006	-0.094	-0.004		0.097	0.025
Si	-0.045	0.97	0.014	-0.044	-0.01		0.055	0.034
Fe	0.532	0.778	0.046	0.128	0.018		0.034	0.088
Ca	0.165	0.523	0.218	0.358	0.124		0.201	-0.07
O ₃ _max8	0.017	0.009	0.741	-0.101	0.473		0.027	-0.108
NH ₃	0.29	0.086	0.654	0.066	-0.128		0.017	0.143
NO ₃	0.426	-0.119	-0.653	0.003	0.122		0.044	0.147
SO ₂ _max1	0.185	0.014	0.007	0.932	0.135		-0.017	-0.016
SO ₂	0.343	0.004	-0.07	0.883	0.131		-0.006	-0.014
NH ₄	0.155	-0.006	-0.082	0.143	0.951		0.044	0.002
SO ₄	0.007	0.072	0.135	0.144	0.942		0.058	0.009
Na	0.053	0.088	-0.026	-0.008	-0.025		0.216	0.866
Cl	0.329	0.035	-0.403	0.07	-0.011		0.691	0.138
Mg	-0.109	0.457	0.15	-0.039	0.014		0.668	0.222
Cu	0.535	0.047	-0.007	0.132	-0.024		0.169	-0.327
Zn	0.749	-0.001	-0.202	0.233	0.114		0.174	-0.043
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.935	0.06	0.041	0.162	-0.017			
CO_max1	0.893	0.041	-0.007	0.179	-0.07			
NO _x	0.897	0.086	0.125	0.195	-0.126			
EC	0.881	0.089	0.043	0.107	0.199			
OC	0.799	0.104	0.019	-0.011	0.329			
NO _z	0.354	-0.032	0.019	0.172	0.12			
nsK	0.409	0.391	0.258	-0.195	0.168			
Al	-0.137	0.933	-0.052	-0.081	-0.043			
Si	0.033	0.961	-0.076	0.019	0.039			
Fe	0.554	0.716	-0.053	0.069	0.045			
Ca	0.386	0.585	-0.094	0.276	0.171			
O ₃ _max8	0.071	0.118	-0.672	-0.04	0.646			
O ₃ _24hr	-0.169	0.048	-0.671	-0.131	0.584			
NO ₃	0.218	-0.104	0.831	0.125	0.096			
SO ₂ _max1	0.145	0.031	0.071	0.924	0.034			
SO ₂	0.379	0.021	0.151	0.86	0.033			
NH ₄	0.17	-0.031	0.107	0.132	0.935			
SO ₄	0.093	0.023	-0.089	0.111	0.921			
Cu	0.115	0.034	-0.014	0.017	-0.006			
Zn	0.587	0.091	0.193	0.05	0.038			

23 Table S6. PCA1 and PCA2 orthogonal factors for SEARCH OAK daily data. PCA1: n =
 24 100, variance explained = 78%. PCA2: n = 707, variance explained = 78%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.623	-0.087	0.114	0.127	-0.043	0.551	-0.13	
CO_max1	0.788	-0.004	0.005	-0.091	-0.25	0.061	-0.068	
NO _x	0.085	0.078	-0.056	0.243	-0.09	0.778	0.054	
EC	0.55	-0.165	0.114	0.256	0.353	0.127	-0.058	
OC	0.58	-0.001	0.289	0.321	0.383	0.142	-0.183	
NO _z	0.223	-0.048	0.438	0.344	0.157	0.651	-0.128	
K	0.353	0.028	0.501	0.106	0.291	0.276	0.114	
Al	-0.031	0.981	-0.018	-0.062	-0.032	-0.027	0.045	
Si	-0.017	0.98	0.022	-0.064	-0.042	-0.036	0.04	
Fe	-0.035	0.982	0.009	-0.048	0.005	-0.003	0.04	
Ca	-0.07	0.775	0.151	0.041	4.30E-04	0.137	0.194	
O ₃ _max8	0.17	-0.182	0.71	0.135	0.315	0.147	-0.148	
NH ₃	-0.029	0.363	0.814	-0.064	-0.077	-0.038	-0.043	
NO ₃	0.129	-0.011	-0.011	-0.018	-0.044	0.743	0.309	
SO ₂ _max1	0.052	-0.059	0.045	0.921	-0.026	0.126	0.037	
SO ₂	0.129	-0.052	0.052	0.824	0.077	0.423	-0.031	
NH ₄	0.037	-0.084	0.128	-0.013	0.934	0.095	-0.089	
SO ₄	-0.056	0.05	0.06	0.008	0.942	-0.082	0.02	
Na	-0.093	0.088	-0.036	0.008	-0.013	-0.025	0.958	
Cl	-0.054	-0.046	-0.031	0.016	-0.089	-0.055	0.929	
Mg	-0.09	0.311	-0.065	-0.041	0.006	0.041	0.88	
Cu	-0.02	0.029	0.075	0.044	0.031	0.696	0.004	
Zn	0.114	-0.075	0.092	0.155	0.121	0.6	-0.254	
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.797	-0.151	0.076	0.191	0.062	-0.064		
CO_max1	0.813	-0.058	0.034	0.013	-0.082	-0.129		
NO _x	0.505	-0.126	-0.502	0.422	-0.204	0.033		
EC	0.774	-0.018	0.142	0.068	0.279	0.074		
OC	0.75	0.037	0.275	0.033	0.294	-0.012		
NO _z	0.691	-0.031	0.253	0.236	0.173	0.115		
nsK	0.465	0.485	0.172	0.061	0.096	0.317		
Al	-0.085	0.97	-0.08	-0.046	-0.033	-0.033		
Si	-0.076	0.981	-0.026	-0.041	-0.03	-0.016		
Fe	-0.069	0.977	-0.029	-0.023	0.018	0.02		
Ca	-0.016	0.892	0.114	0.002	0.023	0.095		
O ₃ _max8	0.198	0.014	0.901	0.096	0.223	0.043		
O ₃ _24hr	0.229	-0.026	0.908	0.112	0.143	0.085		
NO ₃	0.598	6.90E-05	-0.307	-0.004	-0.006	0.288		
SO ₂ _max1	0.02	-0.01	0.061	0.924	0.06	-0.011		
SO ₂	0.291	-0.039	0.1	0.887	0.095	0.026		
NH ₄	0.29	-0.025	0.165	0.065	0.898	0.072		
SO ₄	0.061	0.026	0.22	0.072	0.941	-0.041		
Cu	-0.183	0.102	0.132	-0.06	-0.188	0.766		
Zn	0.194	-0.014	-0.029	0.061	0.177	0.628		

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26 Table S7. PCA1 and PCA2 orthogonal factors for SEARCH OLF daily data. PCA1: n =
 27 327, variance explained = 78%. PCA2: n = 948, variance explained = 76%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.755	-0.175	0.333	0.014	0.297	-0.056	-0.093	
CO_max1	0.464	-0.043	0.438	-0.276	0.271	0.141	-0.079	
NO _x	0.761	-0.033	-0.155	-0.04	-0.138	0.208	-0.174	
EC	0.739	-0.114	0.318	0.173	0.138	-0.008	-0.117	
OC	0.674	-0.086	0.5	0.123	0.27	0.022	-0.094	
NO _z	0.395	-0.002	0.374	0.495	0.086	-0.476	-0.099	
K	0.292	0.272	0.31	0.135	0.118	-0.078	0.303	
Al	-0.106	0.958	-0.014	-0.076	0.013	0.051	0.104	
Si	-0.102	0.964	-0.026	-0.048	1.60E-04	0.089	0.115	
Fe	-0.06	0.958	-0.031	-0.022	0.03	0.117	0.143	
Ca	0.002	0.788	0.052	0.062	-0.058	-0.115	0.095	
O ₃ _max8	0.134	-0.195	0.635	0.303	0.347	-0.084	-0.046	
NH ₃	0.039	0.105	0.801	-0.001	-0.117	0.096	-0.011	
NO ₃	0.694	0.129	-0.151	0.277	-0.104	-0.353	0.179	
SO ₂ _max1	0.064	-0.01	0.06	0.822	0.248	0.095	-0.077	
SO ₂	0.191	-0.039	0.06	0.862	0.227	0.056	-0.1	
NH ₄	0.219	-0.07	0.056	0.262	0.877	-0.057	-0.072	
SO ₄	0.011	0.094	0.054	0.206	0.926	0.042	0.016	
Na	-0.206	0.325	-0.085	-0.217	0.089	-0.113	0.781	
Cl	0.008	-0.007	-0.025	0.044	-0.129	0.181	0.732	
Mg	-0.218	0.483	0.001	-0.199	0.007	-0.095	0.738	
Cu	0.287	0.157	0.165	0.298	-0.027	0.668	0.099	
Zn	0.645	-0.143	-0.004	0.423	0.021	0.149	-0.008	
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		
CO	0.785	-0.152	0.006	0.195	0.309	0.129		
CO_max1	0.854	-0.027	0.122	0.076	0.066	0.022		
NO _x	0.676	-0.04	-0.222	0.481	-0.058	-0.023		
EC	0.606	-0.055	-0.051	0.072	0.564	0.099		
OC	0.509	-0.038	0.16	0.055	0.637	0.157		
NO _z	0.096	-0.028	-0.08	0.011	0.663	0.013		
nsK	0.141	0.328	-0.14	-0.079	0.272	0.588		
Al	-0.062	0.97	-0.042	-0.056	-0.082	-0.004		
Si	-0.062	0.984	-0.036	-0.035	-0.063	-0.001		
Fe	-0.041	0.974	-0.032	-0.009	-0.018	0.047		
Ca	-0.043	0.928	0.069	0.02	0.056	0.1		
O ₃ _max8	0.178	-0.024	0.737	-0.074	0.537	0.11		
O ₃ _24hr	0.038	-0.072	0.757	-0.144	0.467	0.188		
NO ₃	0.397	-0.008	-0.503	0.024	0.292	0.326		
SO ₂ _max1	0.15	0.008	0.003	0.945	0.103	-0.002		
SO ₂	0.205	-0.032	-0.07	0.9	0.242	0.044		
NH ₄	0.114	-0.012	0.139	0.122	0.876	-2.62E-04		
SO ₄	-0.023	0.07	0.215	0.103	0.826	-0.091		
Cu	-0.069	0.1	0.206	-0.118	-0.276	0.71		
Zn	0.133	-0.091	0.006	0.102	0.231	0.732		

29 Table S8. PCA1 and PCA2 orthogonal factors for SEARCH PNS daily data. PCA1: n =
30 44, variance explained = 84%. PCA2: n = 445, variance explained = 79%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.959	0.044	0.036	0.054	0.029		-0.186	
CO_max1	0.929	0.071	0.021	-0.012	-0.121		-0.178	
NO _x	0.919	0.084	-0.069	0.08	-0.142		-0.233	
EC	0.908	-0.008	-0.087	0.067	0.279		-0.222	
OC	0.859	-0.029	0.104	0.067	0.266		-0.27	
NO _z	0.608	-0.142	0.072	0.002	0.61		-0.205	
K	0.726	-0.042	0.083	0.112	0.315		0.223	
Al	-0.109	0.971	0.009	-0.035	-0.044		-0.009	
Si	-0.09	0.971	-0.036	-0.04	-0.051		-0.011	
Fe	0.212	0.88	0.061	0.053	0.017		0.153	
Ca	-0.06	0.276	0.371	0.056	-0.203		-0.04	
O ₃ _max8	0.154	-0.049	0.845	0.023	0.351		-0.072	
NH ₃	0.341	0.539	0.343	-0.063	-0.063		-0.433	
NO ₃	0.654	0.004	-0.52	-0.019	0.405		0.06	
SO ₂ _max1	-0.035	0.021	-0.079	0.918	-0.02		-0.222	
SO ₂	0.072	-0.041	0.061	0.933	0.097		-0.257	
NH ₄	0.113	-0.057	-0.028	0.064	0.961		-0.143	
SO ₄	-0.037	-0.012	0.092	0.028	0.958		-0.026	
Na	-0.244	0.022	0.049	-0.137	-0.105		0.932	
Cl	-0.03	-0.044	-0.252	-0.172	-0.047		0.876	
Mg	-0.28	0.089	0.08	-0.145	-0.111		0.906	
Cu	0.354	-0.011	0.224	0.425	0.028		0.151	
Zn	0.851	-0.001	0.071	0.011	-0.091		-0.009	
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		Other
CO	0.893	-0.051	-0.001	0.135		0.027		-0.051
CO_max1	0.826	-0.043	0.014	0.164		0.086		-0.156
NO _x	0.862	0.01	-0.173	0.215		0.044		-0.255
EC	0.91	0.001	0.089	0.077		-0.01		0.079
OC	0.842	-0.014	0.251	0.07		-0.08		0.163
NO _z	0.079	-0.03	0.033	0.119		0.107		0.827
nsK	0.659	0.314	0.058	-0.067		0.046		0.233
Al	-0.091	0.959	0.005	-0.026		-0.081		-0.027
Si	-0.099	0.962	0.011	-0.039		-0.08		-0.016
Fe	0.195	0.914	0.008	0.02		0.017		0.002
Ca	0.142	0.672	0.077	0.123		0.266		0.016
O ₃ _max8	0.095	0.067	0.934	0.01		0.12		-0.068
O ₃ _24hr	-0.205	0.008	0.889	-0.113		0.115		-0.07
NO ₃	0.681	0.017	-0.141	-0.038		-0.094		0.293
SO ₂ _max1	0.135	0.007	-0.03	0.954		-0.022		0.058
SO ₂	0.207	0.05	-0.032	0.937		0.009		0.078
NH ₄	0.282	-0.003	0.64	0.056		-0.488		0.374
SO ₄	0.117	0.045	0.677	0.017		-0.497		0.338
Cu	0.153	0.049	0.068	-0.006		0.755		0.153
Zn	0.695	0.105	0.005	0.081		0.153		0.11

32 Table S9. PCA1 and PCA2 orthogonal factors for SEARCH YRK daily data. PCA1: n =
 33 426, variance explained = 79%. PCA2: n = 1435, variance explained = 79%.

PCA1	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.743	-0.098	0.468	-0.073	0.198	0.058	0.024	0.105
CO_max1	0.728	-0.061	0.38	-0.079	0.118	0.166	0.064	0.104
NO _x	0.809	-0.093	-0.023	0.239	-0.131	0.046	0.07	-0.007
EC	0.308	-0.043	0.715	-0.012	0.208	0.076	0.14	0.294
OC	0.12	0.075	0.773	-0.01	0.306	0.115	0.158	0.269
NO _z	0.392	0.039	0.699	0.211	0.136	-0.126	-0.101	0.038
K	0.132	0.353	0.488	0.059	0.043	0.05	0.317	0.299
Al	-0.088	0.955	-0.014	-0.064	-0.028	0.004	0.076	0.007
Si	-0.123	0.971	-0.003	-0.015	-0.011	0.024	0.069	0.012
Fe	-0.045	0.957	0.055	0.022	0.074	0.031	0.088	0.041
Ca	0.084	0.53	0.025	0.126	0.118	0.112	0.205	-0.377
O ₃ _max8	-0.234	0.026	0.79	0.024	0.259	-0.043	-0.219	-0.089
NH ₃	-0.012	0.02	0.623	0.093	-0.125	0.163	0.135	-0.443
NO ₃	0.782	-0.05	-0.081	0.148	-0.01	-0.123	0.047	0.111
SO ₂ _max1	0.061	0.008	0.044	0.952	0.099	0.029	0.016	-0.005
SO ₂	0.192	-0.007	0.084	0.942	0.114	0.007	-0.009	0.087
NH ₄	0.124	0.012	0.25	0.142	0.922	-0.04	-0.011	0.037
SO ₄	-0.07	0.07	0.253	0.097	0.932	-0.021	0.01	0.032
Na	0.024	0.15	0.116	0.011	-0.134	-0.116	0.802	0.071
Cl	0.183	0.092	-0.105	0.007	0.154	0.157	0.78	-0.102
Mg	-0.14	0.564	0.163	-0.038	-0.017	0.016	0.583	-0.092
Cu	0.028	0.091	0.071	0.03	-0.049	0.93	0.021	0.074
Zn	0.234	-0.037	0.218	0.137	0.052	0.144	-0.005	0.668
PCA2	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Other	
CO	0.836	-0.058	0.294	0.032		0.042		0.092
CO_max1	0.837	-0.021	0.242	0.024		0.054		0.047
NO _x	0.823	-0.061	-0.222	0.212		-0.089		0.012
EC	0.588	-0.009	0.581	0.029		0.147		0.101
OC	0.384	0.108	0.724	0.024		0.233		0.149
NO _z	0.294	-0.034	0.617	0.248		0.039		0.221
nsK	0.255	0.36	0.194	-0.049		0.516		0.221
Al	-0.12	0.936	-0.108	-0.07		0.069		-0.105
Si	-0.089	0.975	0.012	-0.02		0.05		-0.032
Fe	-0.023	0.965	0.084	-0.011		0.043		0.023
Ca	-0.032	0.65	0.301	0.086		0.004		0.412
O ₃ _max8	-0.217	0.039	0.883	0.043		0.058		0.125
O ₃ _24hr	-0.307	0.027	0.835	0.045		0.113		0.192
NO ₃	0.703	-0.089	-0.064	0.179		-0.048		0.093
SO ₂ _max1	0.092	-0.006	0.103	0.936		-0.021		-0.028
SO ₂	0.271	-0.03	0.186	0.892		-0.037		0.115
NH ₄	0.302	0.028	0.825	0.094		-0.152		-0.088
SO ₄	0.121	0.07	0.858	0.071		-0.151		-0.106
Cu	-0.094	0.019	-0.062	-0.027		0.876		-0.047
Zn	0.181	0.012	0.112	0.05		0.029		0.889

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35 Table S10. PCA3 orthogonal factors for SEARCH JST daily data. PCA3, 1999 – 2008: n
36 = 426, variance explained = 79%. PCA2: n = 1435, variance explained = 79%.

PCA3	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.916	0.032	0.165	0.107	-0.073	0.039		0.087
CO_max1	0.86	0.023	0.113	0.13	-0.108	0.036		0.1
NO _x	0.875	0.043	0.199	0.16	-0.182	0.079		0.008
EC	0.841	0.056	0.256	0.074	0.157	0.029		0.038
OC	0.731	0.077	0.319	-0.026	0.265	0.004		0.084
NO _z	0.188	-0.04	0.05	0.059	0.075	-0.01		0.922
nsK	0.32	0.33	0.5	-0.108	0.057	-0.009		0.059
Al	-0.012	0.929	-0.025	-0.036	0.015	-0.013		-0.068
Si	0.153	0.94	0.005	0.029	0.112	0.003		0.018
Fe	0.652	0.622	0.116	0.073	0.125	0.087		-0.006
Ca	0.363	0.53	0.123	0.239	0.223	0.082		0.123
O ₃ _max8	0.088	0.142	-0.225	-0.049	0.859	0.071		0.153
O ₃ _24hr	-0.163	0.087	-0.238	-0.13	0.802	0.068		0.162
NO ₃	0.077	-0.187	0.774	0.146	-0.257	-0.07		0.028
SO ₂ _max1	0.131	0.033	-0.01	0.941	-0.004	-0.007		-0.024
SO ₂	0.332	0.006	0.117	0.881	-0.057	-0.035		0.071
NH ₄	0.13	-0.095	0.345	0.063	0.848	-0.081		-0.056
SO ₄	0.098	-0.016	0.149	0.05	0.905	-0.057		-0.104
Cu	0.126	0.006	0.012	0.001	0.005	0.961		-0.008
Zn	0.425	0.059	0.42	0.067	-0.011	0.146		-0.029
Alkanes	0.907	0.015	0.043	0.019	-0.087	-0.028		0.035
Aromatics	0.917	0.03	-0.008	0.011	-0.021	-0.033		0.069
Alpha pinene	0.803	0.04	-0.158	-0.047	-0.009	-0.088		-0.091
Isoprene	0.013	0.373	-0.282	0.054	0.583	-0.041		-0.136

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40 Table S11. PCA4 (including NH₃) and PCA5 (excluding NH₃; including daily average
 41 O₃) orthogonal factors for SEARCH CTR daily data, 2004 - 2013. PCA4: n = 724,
 42 variance explained = 76%. PCA5: n = 751, variance explained = 79%.

PCA4	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
CO	0.733	-0.135	0.019	0.223	-0.044	0.313		
CO_max1	0.731	-0.076	0.026	0.196	0.01	0.177		
NO _x	0.219	-0.093	-0.258	0.82	-0.097	0.102		
EC	0.672	-0.039	0.07	0.187	0.45	-0.067		
OC	0.71	0.094	0.24	-0.043	0.458	-0.137		
NO _z	0.52	-0.003	0.089	0.359	0.425	0.188		
nsK	0.71	0.298	0.057	-0.054	0.162	-0.067		
Al	-0.052	0.946	-0.017	-0.131	-0.095	-0.058		
Si	-0.041	0.964	0.014	-0.111	-0.056	-0.087		
Fe	0.026	0.963	-0.006	0.012	0.05	0.076		
Ca	0.131	0.698	0.132	0.176	0.178	0.079		
O ₃ _max8	0.252	0.009	0.548	-0.005	0.604	0.164		
NH ₃	0.251	0.082	0.754	0.111	-0.122	-0.027		
NO ₃	0.329	-0.004	-0.488	0.442	-0.07	0.054		
SO ₂ _max1	0.045	0.016	0.164	0.874	0.13	-0.073		
SO ₂	0.103	0.001	0.089	0.923	0.177	-0.059		
NH ₄	0.196	0.009	-0.097	0.138	0.909	-0.027		
SO ₄	0.081	0.048	-0.047	0.03	0.934	-0.045		
Cu	0.03	0.018	0.05	-0.175	-0.096	0.839		
Zn	0.31	0.011	-0.132	0.4	0.181	0.607		
PCA5	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals		Other
CO	0.689	-0.141	0.071	0.239	-0.104	0.381		
CO_max1	0.725	-0.078	0.041	0.217	-0.055	0.221		
NO _x	0.178	-0.101	-0.25	0.823	-0.035	0.189		
EC	0.705	-0.044	0.154	0.217	0.392	-0.042		
OC	0.756	0.089	0.267	-0.013	0.368	-0.128		
NO _z	0.461	-0.005	0.415	0.401	0.265	0.248		
nsK	0.739	0.29	0.114	-0.014	0.091	-0.038		
Al	-0.038	0.944	-0.107	-0.138	-0.046	-0.05		
Si	-0.021	0.964	-0.057	-0.112	-0.026	-0.086		
Fe	0.024	0.962	0.004	0.014	0.059	0.084		
Ca	0.132	0.702	0.226	0.193	0.072	0.054		
O ₃ _max8	0.251	0.023	0.894	0.03	0.266	0.065		
O ₃ _24hr	0.21	-0.01	0.905	-0.02	0.213	0.054		
NO ₃	0.191	-0.025	-0.362	0.456	0.149	0.309		
SO ₂ _max1	0.077	0.024	0.149	0.876	0.012	-0.131		
SO ₂	0.114	0.005	0.112	0.926	0.094	-0.078		
NH ₄	0.206	-0.001	0.197	0.167	0.917	0.014		
SO ₄	0.122	0.042	0.227	0.059	0.913	-0.051		
Cu	0.008	0.022	0.058	-0.205	-0.134	0.77		
Zn	0.242	0.005	0.055	0.404	0.166	0.655		

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45 Table S12. Mean OC associated with components identified by PCA1 (2008 – 2013) and
 46 PCA2 (1999 – 2013). NS = not statistically significant, NA = not applicable (component
 47 not present in PCA). Units are percent. Mean concentrations are listed in Table 3.

PCA	Site	N	Combustion	Crustal	Seasonal	SO ₂	SO ₄	Metals	Salt	Other
1	BHM	364	44.7%	3.0%	13.2%	11.5%	14.8%	4.9%	4.6%	3.3%
1	CTR	383	52.7%	10.7%	23.0%		13.6%			
1	GFP	100	47.3%		22.4%	7.5%	12.4%		-20.4% ^a	30.8%
1	JST	516	38.5%	5.7%	17.3%	2.5%	9.5%		22.6%	3.9%
1	OAK	100	21.6%		27.0%	20.0%	28.6%	17.3%	-14.6% ^a	
1	OLF	327	43.8%		30.8%	9.5%	16.0%			
1	PNS	44	88.2%		14.9%		25.3%		-28.5% ^a	
1	YRK	426	5.8%	5.8%	45.2%		10.8%	6.6%	6.2%	19.5%
2	BHM	1513	43.4%	5.1%	12.7%	10.3%	15.4%	13.0%		
2	CTR	1258	50.5%	4.0%	23.2%		22.2%			
2	GFP	376	32.9%	6.4%	16.9%	9.6%	22.8%	11.4%		
2	JST	2593	62.9%	7.8%	1.5% ^b		24.6% ^b	3.2%		
2	OAK	707	58.6%		18.4%		23.0%			
2	OLF	948	35.1%	-2.6% ^a	10.8%	3.9%	44.2%	8.7%		
2	PNS	445	61.3%		17.8% ^c	6.7%				14.2%
2	YRK	1435	24.6%	6.5%	45.3% ^c	1.6%		12.6%		9.4%

- 48 a. Negative due to inverse associations of OC with crustal and salt components at
 49 (Tables S7 and S13).
 50 b. JST PCA2 seasonal OC is associated with NO₃; JST PCA2 SO₄ component
 51 includes OC associated with O₃ (Table 2).
 52 c. PNS and YRK PCA2 seasonal components include OC associated with SO₄
 53 (Table 2).

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Table S13. Statistically significant regression results for multiple regression of OC on PCA1 factor scores. Factor composition is defined in Tables S2 through S9. Units for mean OC are $\mu\text{g m}^{-3}$. Factors are normalized (mean 0, variance 1), so units are $\mu\text{g m}^{-3}$ factor-unit⁻¹ for $\Delta\text{OC}/\Delta$ factor. NS = not significant; NA = not applicable (component not present).

Parameter	BHM	CTR	GFP	JST	OAK	OLF	PNS	YRK
N	364	383	100	516	100	327	44	426
r ²	0.836	0.799	0.732	0.772	0.605	0.797	0.892	0.802
	2.904	2.422	2.129	3.783	2.545	2.274	2.734	2.840
Mean OC	± 0.040	± 0.029	± 0.078	± 0.052	± 0.102	± 0.039	± 0.107	± 0.040
	1.429	1.028	0.918	1.703	0.784	0.971	2.011	0.213
$\Delta\text{OC}/\Delta\text{Combustion}$	± 0.040	± 0.031	± 0.077	± 0.048	± 0.101	± 0.038	± 0.128	± 0.035
	0.100	0.201	NS	0.140	NS	-0.116	NS	0.121
$\Delta\text{OC}/\Delta\text{Crustal}$	± 0.043	± 0.028		± 0.023		± 0.024		± 0.021
	0.371	0.455	0.607	0.406	0.424	0.641	0.342	1.270
$\Delta\text{OC}/\Delta\text{Seasonal}$	± 0.048	± 0.031	± 0.069	± 0.038	± 0.076	± 0.033	± 0.109	± 0.037
	0.413	NS	0.161	0.087	0.461	0.284	NS	NS
$\Delta\text{OC}/\Delta\text{SO}_2$	± 0.046		± 0.076	± 0.038	± 0.087	± 0.053		
	0.570	0.263	0.349	0.735	0.594	0.409	0.672	0.634
$\Delta\text{OC}/\Delta\text{SO}_4$	± 0.042	± 0.028	± 0.093	± 0.056	± 0.102	± 0.047	± 0.137	± 0.049
	0.182	NS	NS	NA	0.250	NS	NA	0.432
$\Delta\text{OC}/\Delta\text{Metals}$	± 0.039				± 0.095			± 0.033
	0.159	NS	-0.549	0.669	-0.268	-0.122	-0.532	0.212
$\Delta\text{OC}/\Delta\text{Salt}$	± 0.039		± 0.095	± 0.044	± 0.073	± 0.027	± 0.092	± 0.032
	0.132	NA	0.569	0.134	NA	NA	NA	0.131
$\Delta\text{OC}/\Delta\text{Other}^a$	± 0.051		± 0.074	± 0.031				± 0.022

a. "Other" is predominantly NO_x, Ca, Mg at BHM, Na at JST, Zn at YRK.

64 Table S14. Statistically significant regression results for multiple regression of OC on
65 PCA2 factor scores. Factor composition is defined in Tables S2 through S9. Units for
66 mean OC are $\mu\text{g m}^{-3}$. Factor scores are normalized (mean 0, variance 1), so the units are
67 $\mu\text{g m}^{-3}$ factor-unit⁻¹ for $\Delta\text{OC}/\Delta$ factor. NS = not significant; NA = not applicable (PCA
68 component not present).

Parameter	BHM	CTR	GFP	JST	OAK	OLF	PNS	YRK
N	1513	1258	376	2593	707	948	445	1435
r ²	0.750	0.800	0.728	0.751	0.682	0.711	0.800	0.750
	3.938	2.990	2.296	4.035	2.625	2.302	2.668	3.000
Mean OC	± 0.034	± 0.023	± 0.038	± 0.024	± 0.041	± 0.025	± 0.040	± 0.023
	2.097	1.442	0.920	1.882	1.641	0.687	1.685	0.626
$\Delta\text{OC}/\Delta\text{Combustion}$	± 0.036	± 0.024	± 0.049	± 0.023	± 0.047	± 0.023	± 0.042	± 0.021
	0.205	0.105	0.176	0.253		-0.054		0.180
$\Delta\text{OC}/\Delta\text{Crustal}$	± 0.035	± 0.024	± 0.036	± 0.024	NS	± 0.027	NS	± 0.023
	0.492	0.555	0.436	0.051	0.596	0.241	0.519	1.193
$\Delta\text{OC}/\Delta\text{Seasonal}$	± 0.034	± 0.022	± 0.038	± 0.023	± 0.043	± 0.025	± 0.042	± 0.022
	0.420		0.274			0.084	0.146	0.045
$\Delta\text{OC}/\Delta\text{SO}_2$	± 0.033	NS	± 0.045	NS	NS	± 0.023	± 0.037	± 0.021
	0.841	0.635	0.776	0.790	0.656	0.937		
$\Delta\text{OC}/\Delta\text{SO}_4$	± 0.039	± 0.023	± 0.042	± 0.023	± 0.044	± 0.026	NS	NS
	0.635		0.315	0.103		0.218		0.389
$\Delta\text{OC}/\Delta\text{Metals}$	± 0.042	NS	± 0.032	± 0.040	NS	± 0.026	NS	± 0.022
$\Delta\text{OC}/\Delta\text{Salt}$	NA	NA	NA	NA	NA	NA	NA	NA
							0.375	0.237
$\Delta\text{OC}/\Delta\text{Other}^a$	NA	NA	NA	NA	NA	NA	± 0.039	± 0.020

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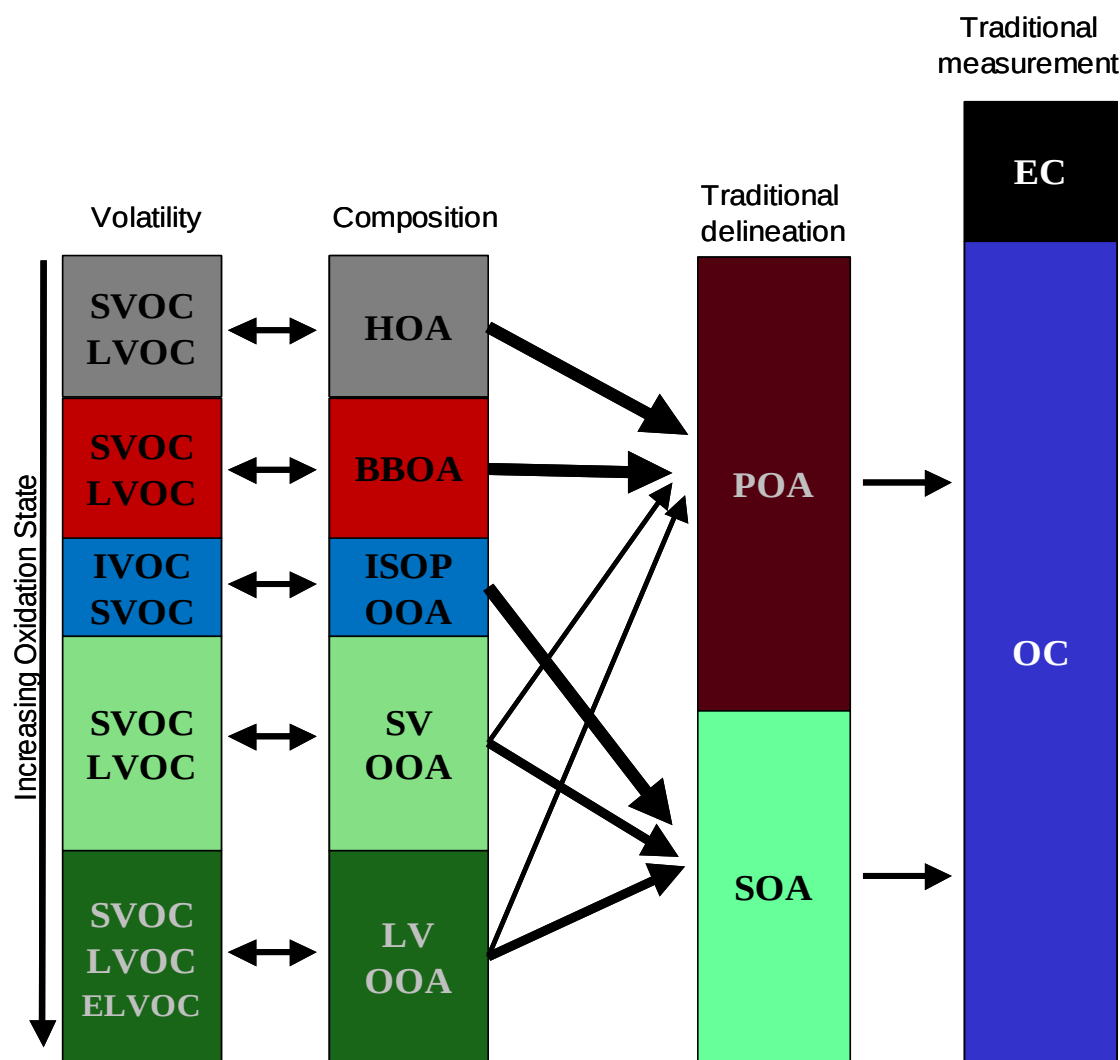


Figure S1. Graphical depiction of various categorizations of OA. Traditional measurements and delineation: EC, OC, POA, SOA as defined in the text. Composition-based categories: HOA, hydrocarbon-like OA; BBOA, biomass burning OA; ISOP OOA, isoprene-derived oxidized OA; SVOOA, semi-volatile oxidized OA (also, less-oxidized OA); LVOOA, low-volatility oxidized OA (also, more-oxidized OA). Volatility-based categories (Donahue et al., 2009; 2012): SVOC, semi-volatile OC; LVOC, low-volatility OC; IVOC, intermediate volatility OC; ELVOC, extremely low-volatility OA. Arrows denote correspondences. Composition categories (HOA, BBOA, SVOA) map to similar volatility ranges (SVOC, LVOC), but differ based on oxidation state (Donahue et al., 2012). The traditional categorization (POA, SOA), e.g., as used in emission inventories, tends to map some SVOA and LVOOA, principally organic material emitted in the condensed phase but subject to volatilization and oxidation, within POA. Traditional OC measurements do not distinguish among composition-based or volatility-based categories. Statistical analysis of multi-species data sets may identify correlations of OC with combustion products (e.g., CO, EC, NO_x) suggestive of less-oxidized OA, or with O₃ and SO₄, suggestive of more-oxidized OA.

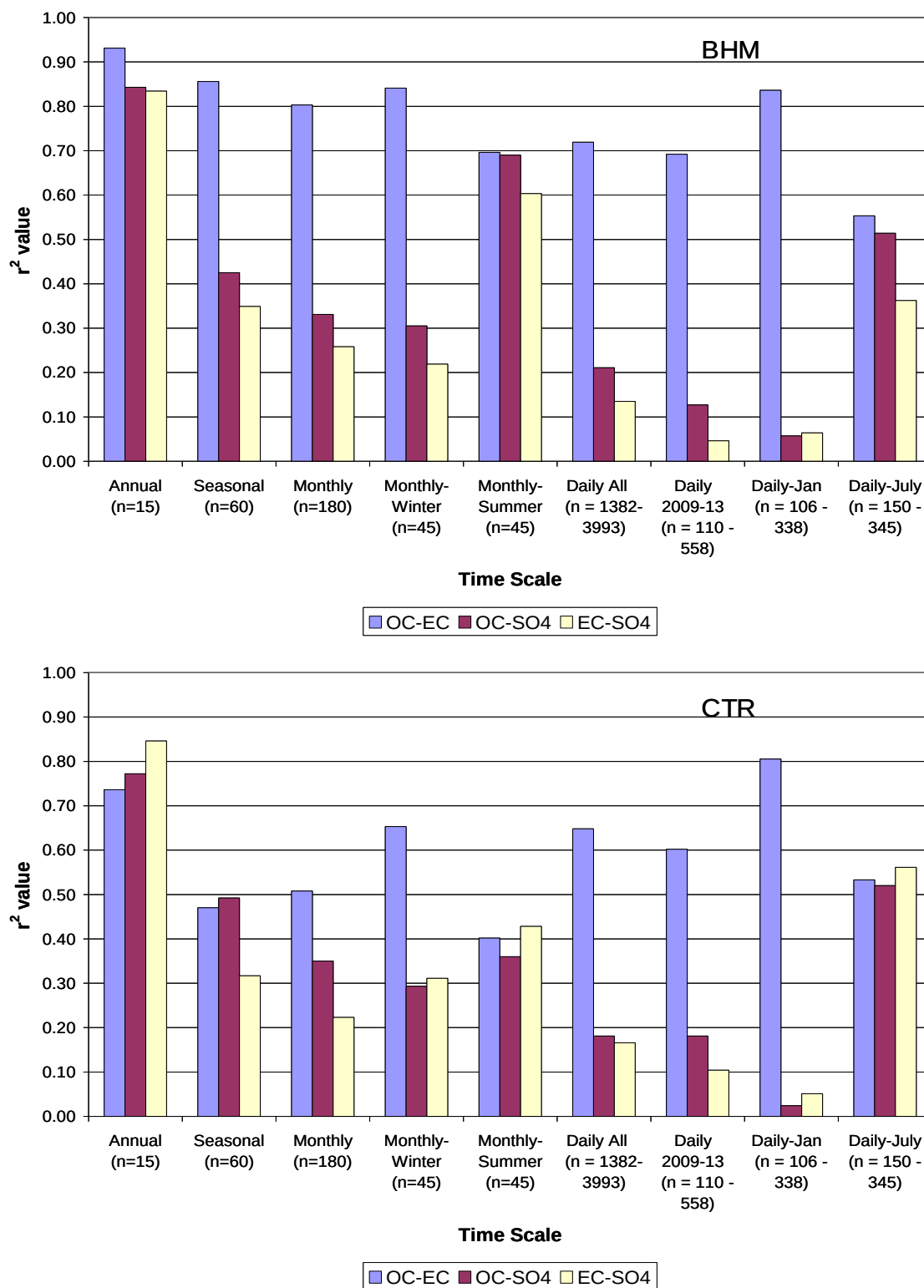


Figure S2. Correlations (r^2) among mean OC, EC, and SO_4 based on annual, seasonal, and monthly averaging periods and on daily measurements. Summer is defined as June through August, winter is December through February, autumn is September through November, and spring is March through May.

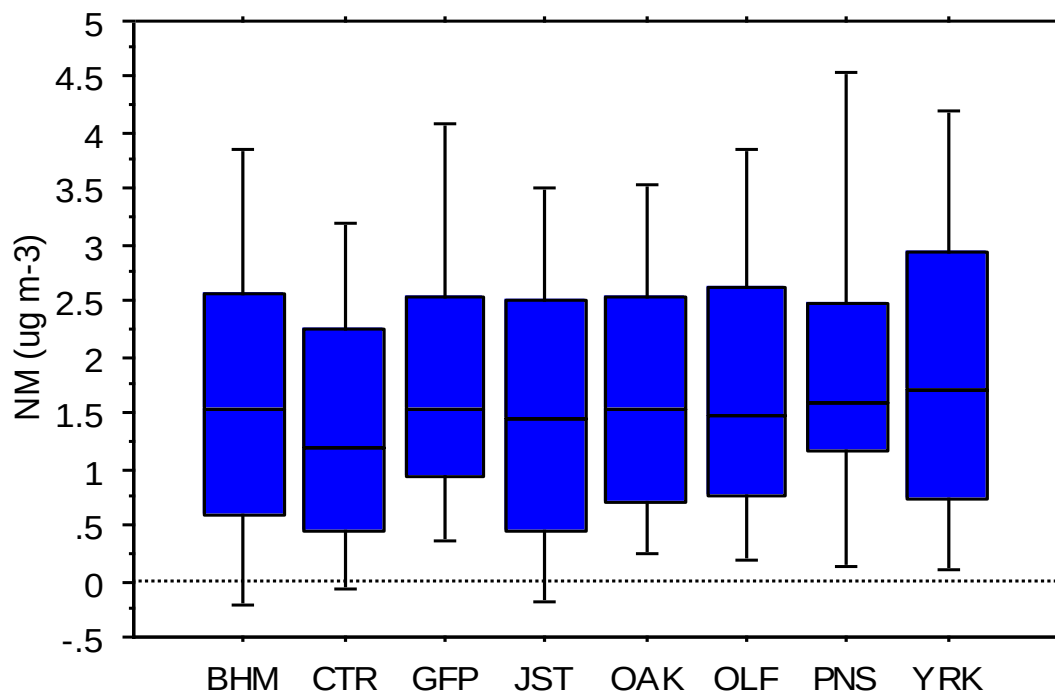


Figure S3. Distributions of non-measured (NM) $PM_{2.5}$ computed from 2009 – 2013 daily data as described in the text. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. No measurements were made at PNS after 2009, at OAK after 2010, and at GFP after 2012 (GFP EC and OC data were not available after 2010).

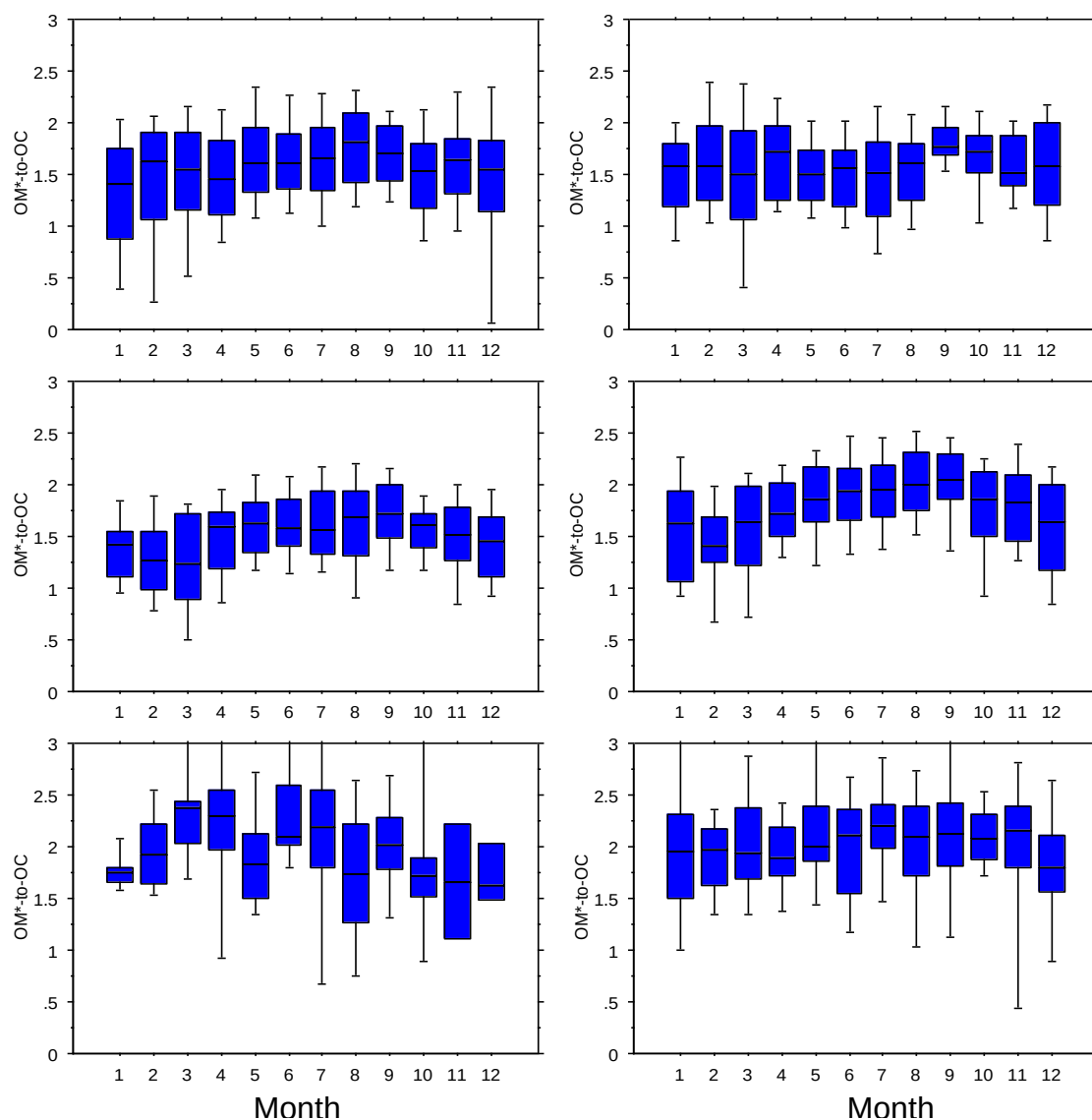


Figure S4. Distributions of daily OM*/OC vs. month at SEARCH sites, 2009 – 2013. GFP OC and EC measurements were available only for 2009 – 2010 (n = 90). Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles.

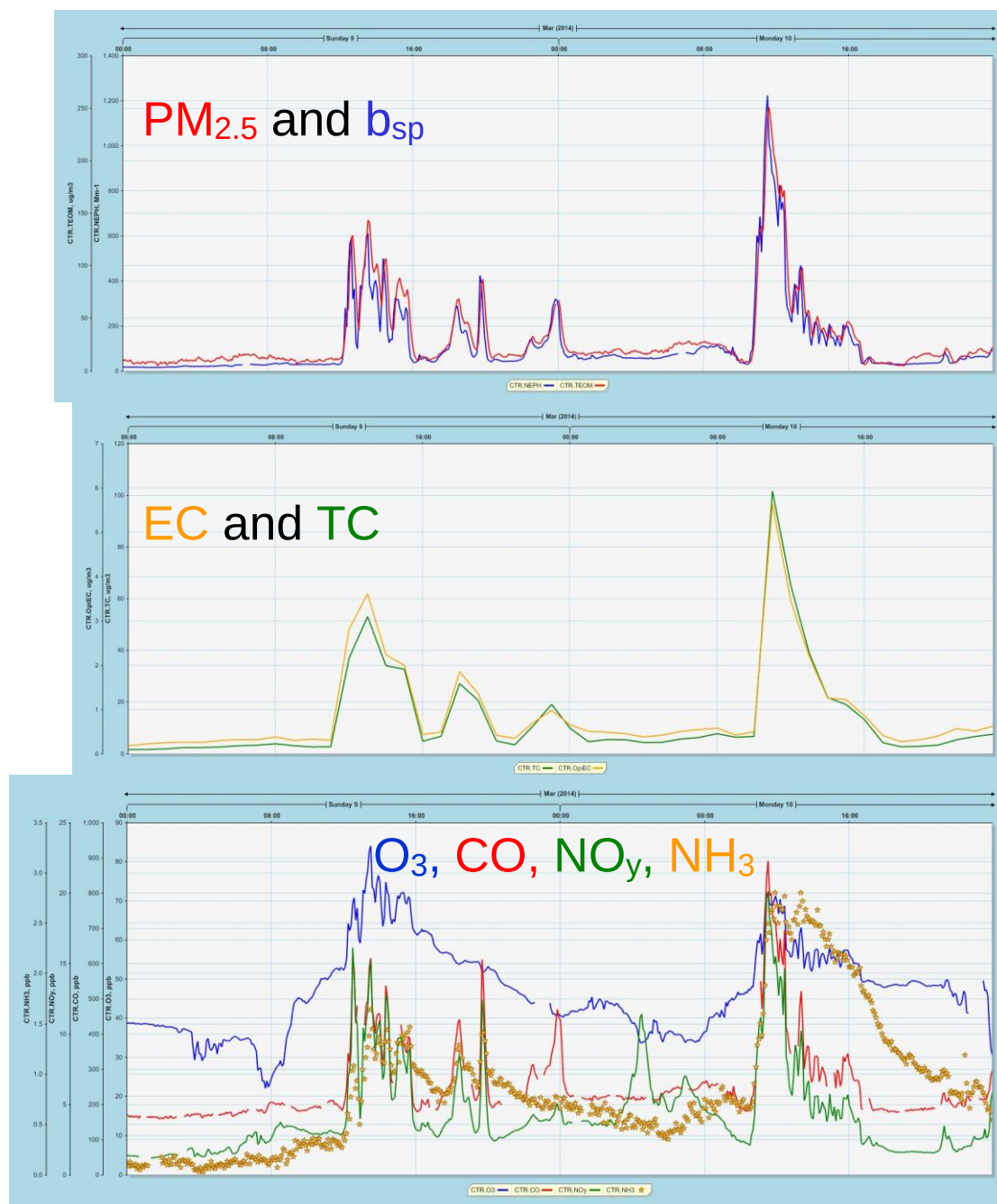


Figure S5. Observed biomass-burning impacts at CTR on March 9 – 10, 2014. The burn was located ~10 km northwest of CTR on a ~1000 acre parcel. EC and TC data are hourly; all other measurements are 5-minute resolution.

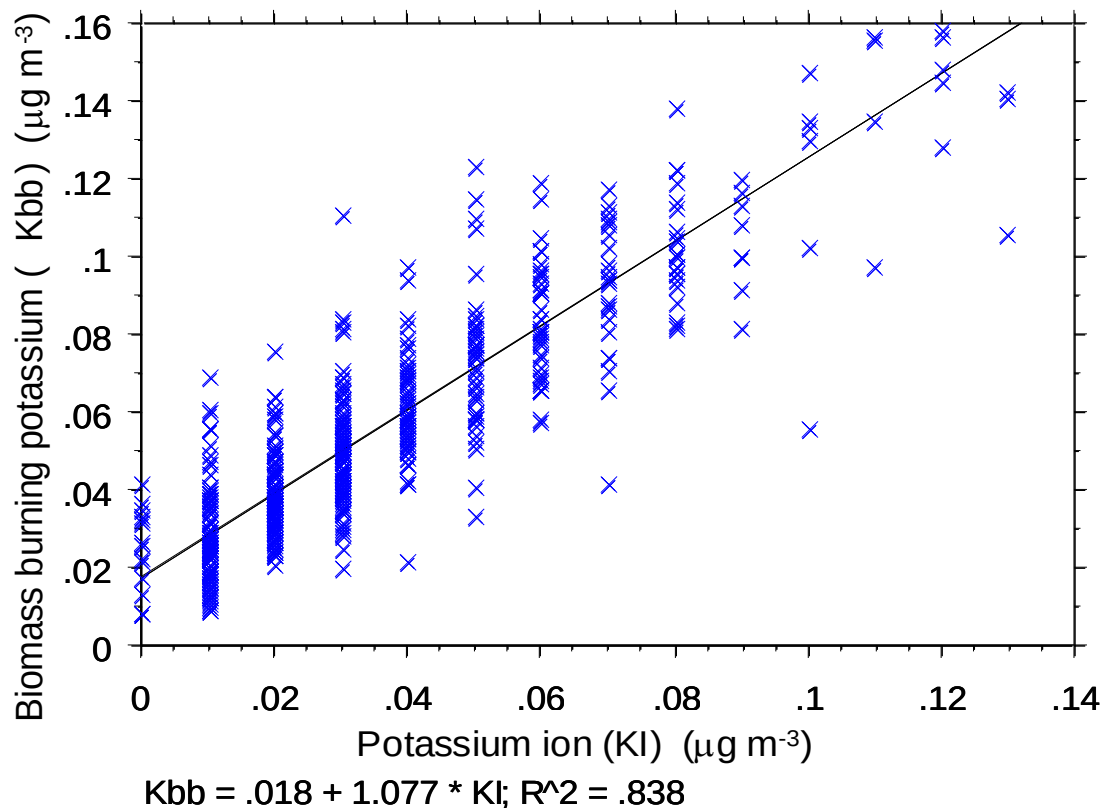
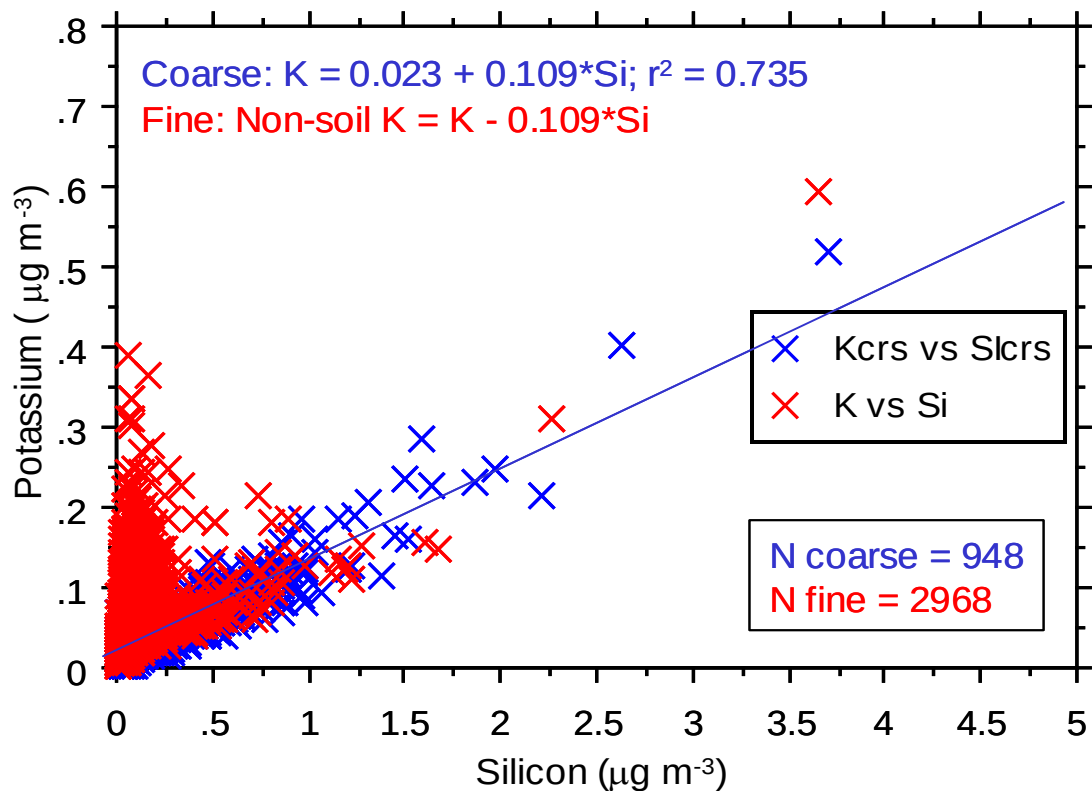


Figure S6. $\text{PM}_{\text{coarse}}$ and $\text{PM}_{2.5}$ K vs. Si at CTR (top) and computed non-soil K (inferred as biomass-burning K) vs. water soluble K (K ion) (bottom).

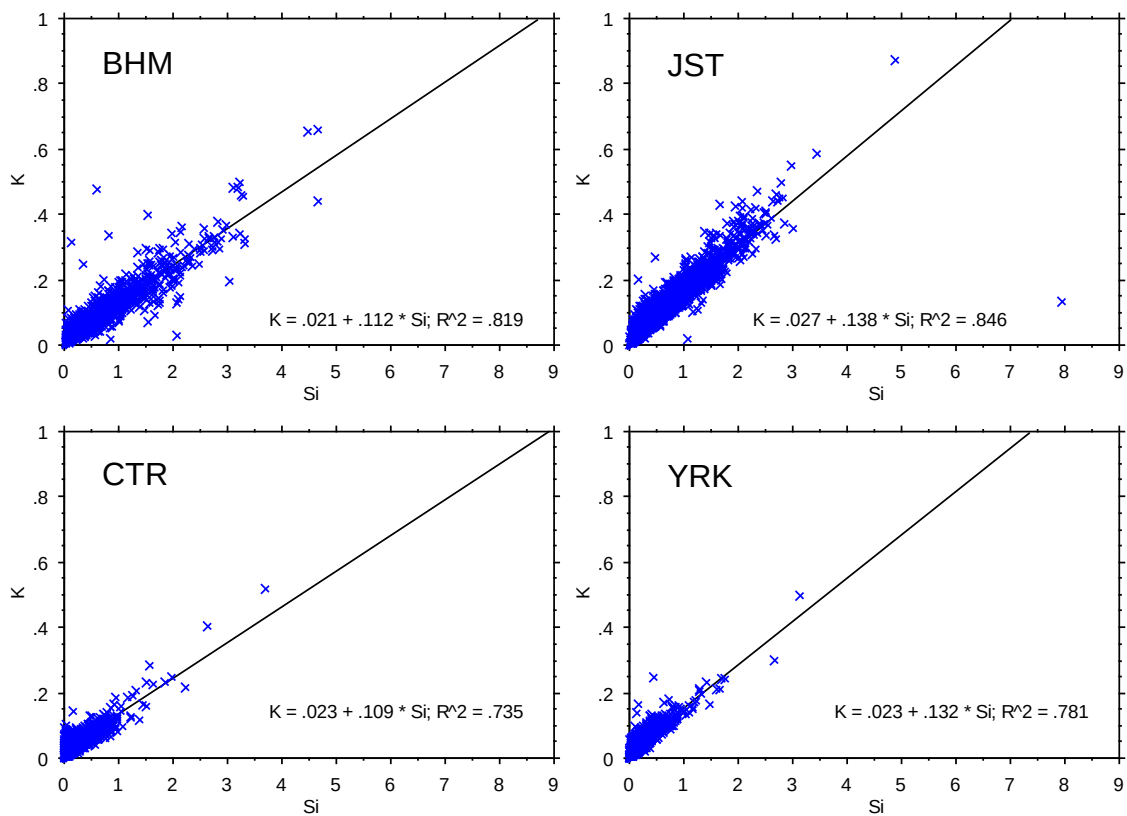


Figure S7. $PM_{10-2.5}$ K vs. Si. Units are $\mu g m^{-3}$.

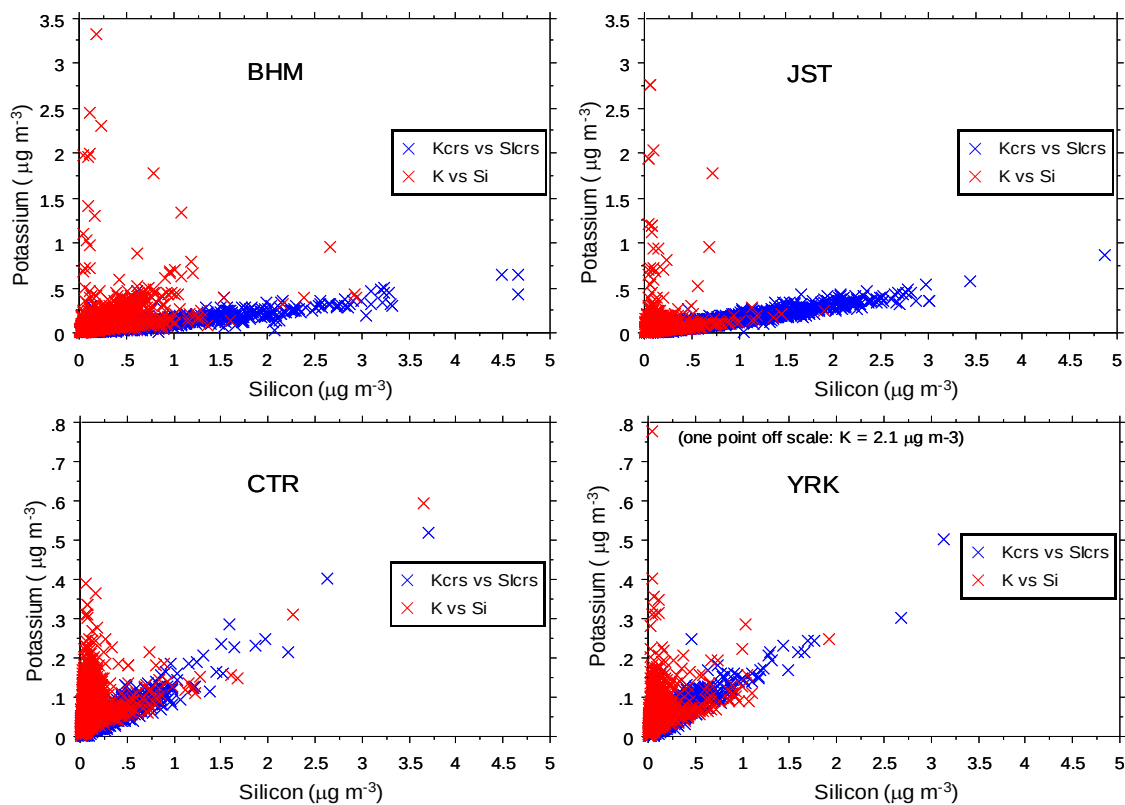


Figure S8. PM_{crs} ($\text{PM}_{10-2.5}$) K vs. Si and $\text{PM}_{2.5}$ K vs. Si. Units are $\mu\text{g m}^{-3}$.

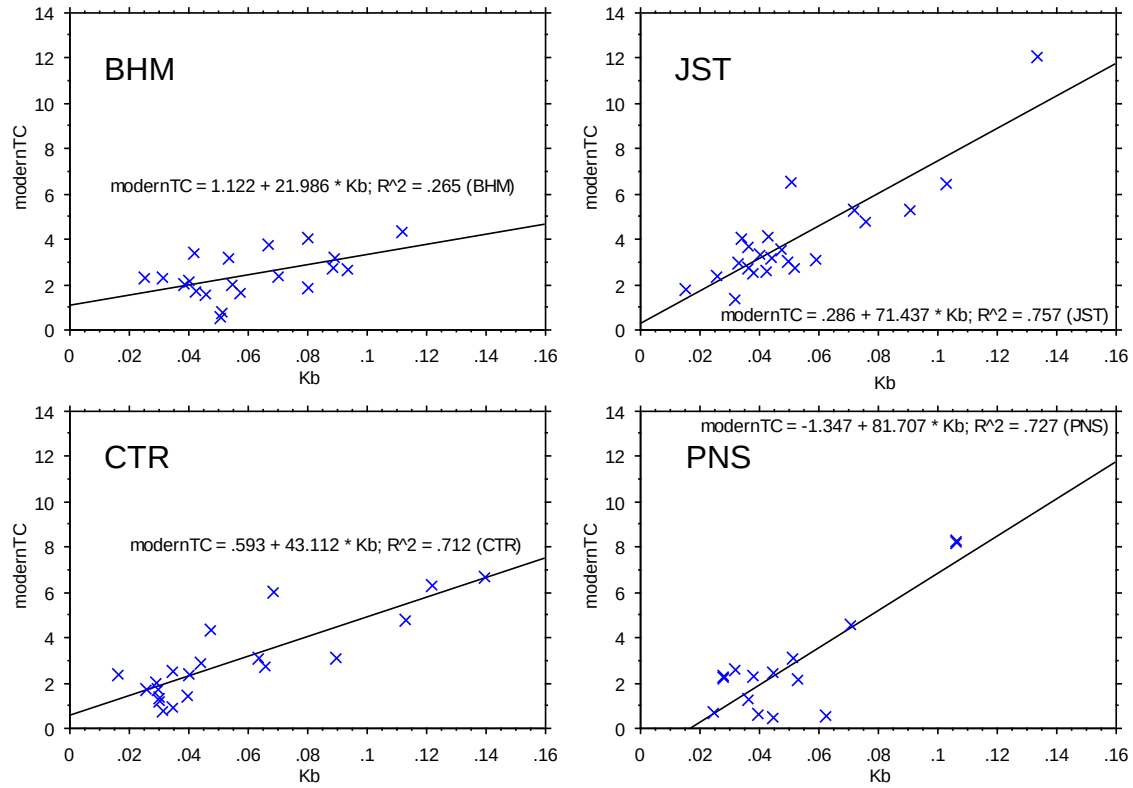


Figure S9. Modern total carbon (TC) (Blanchard et al., 2011) vs. biomass-burning K (Kb) for daily-average samples collected from the months of October through March (2004 and 2005).

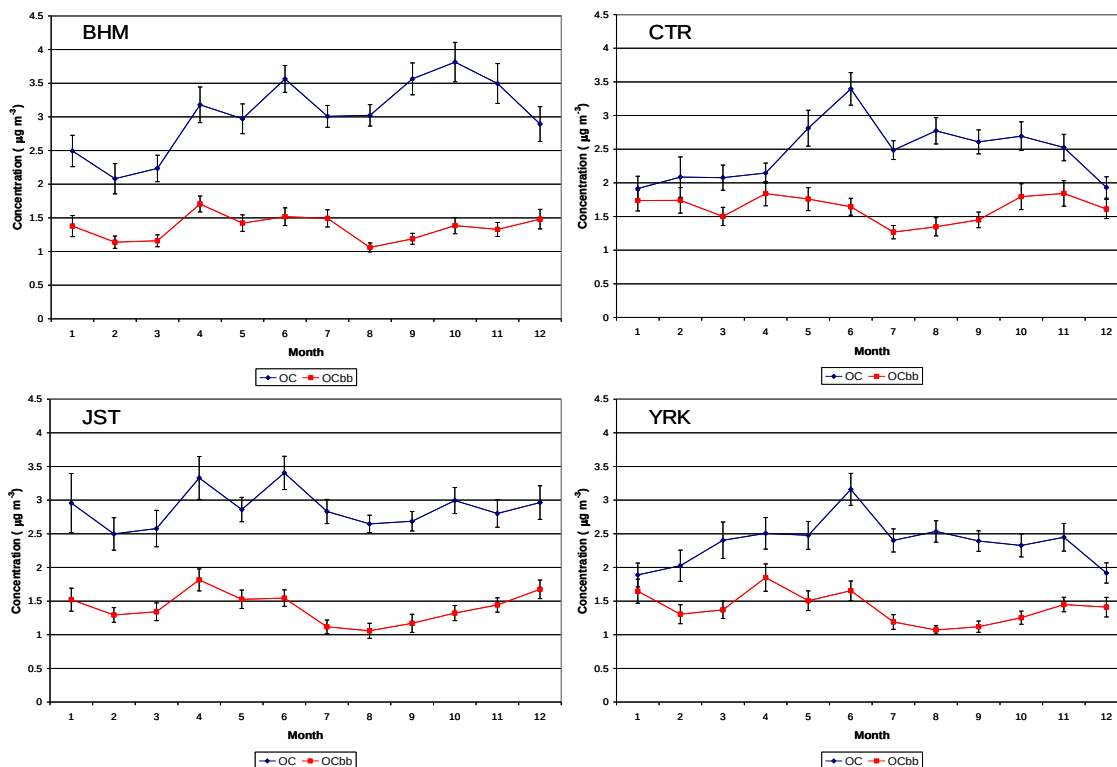


Figure S10. Monthly-average OC and OCbb at inland SEARCH sites, 1999-2013. Uncertainties are one standard error of the means.

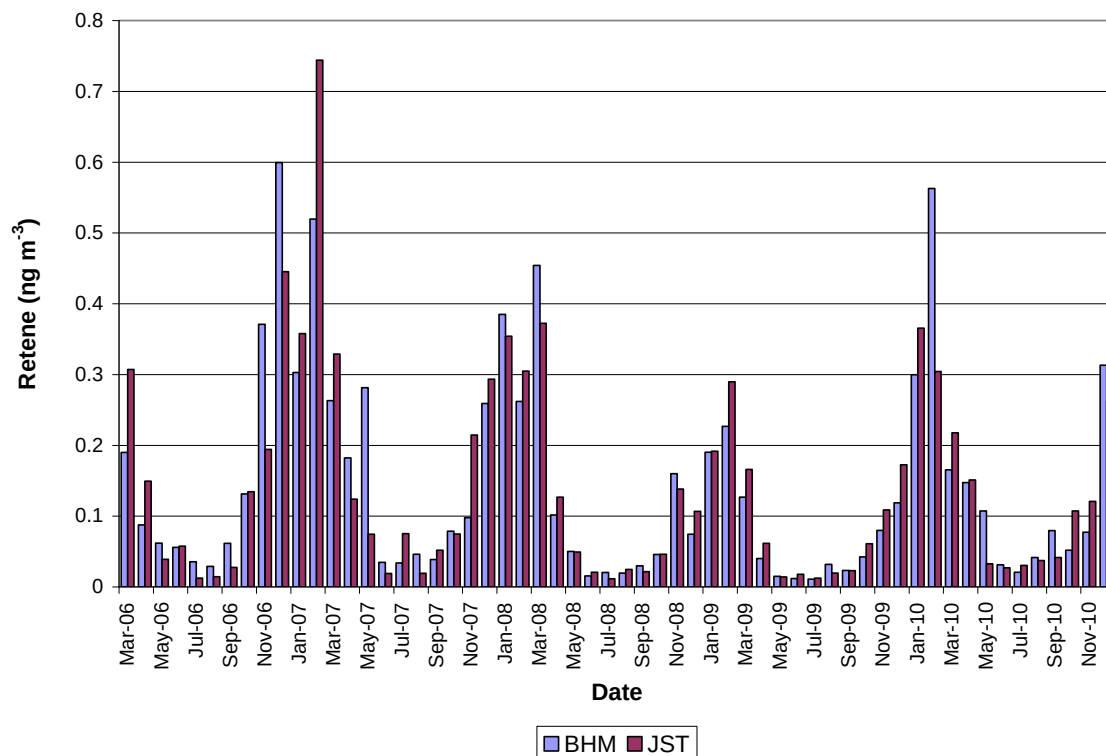


Figure S11. Monthly-average $\text{PM}_{2.5}$ retene concentrations at urban sites BHM and JST (Blanchard et al., 2014).

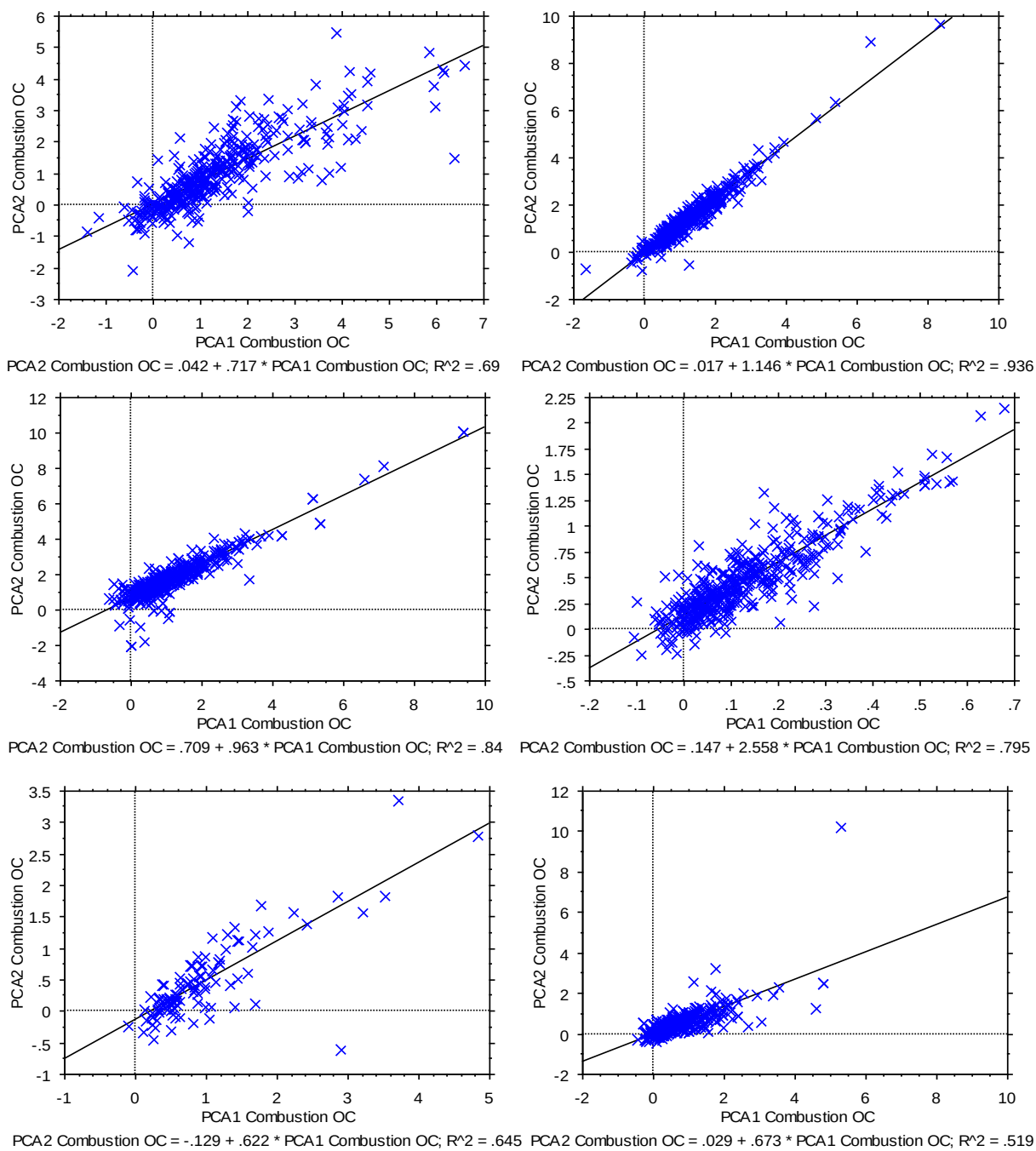
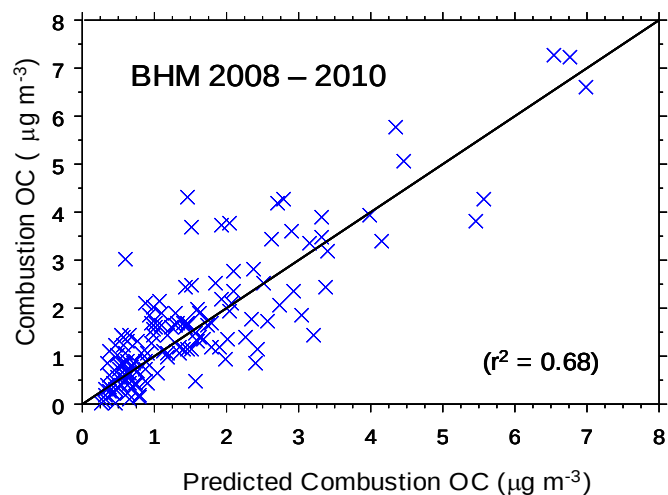


Figure S12. Comparison of PCA1 and PCA2 combustion-derived OC. Units are $\mu\text{g m}^{-3}$.



$$Y = 0.17 + 0.069 \cdot \text{NO} + 0.011 \cdot \text{PAHs} + 0.49 \cdot \text{iso/anteiso alkanes}$$

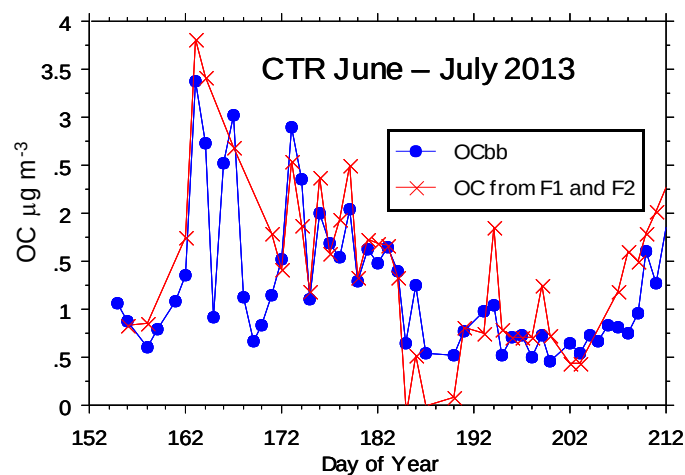


Figure S13. Comparison of daily combustion OC with indicators of mobile-source emissions at BHM (top) and with computed biomass-burning OC at CTR (bottom). For CTR, OC from F1 and F2 is the sum of combustion and crustal OC.

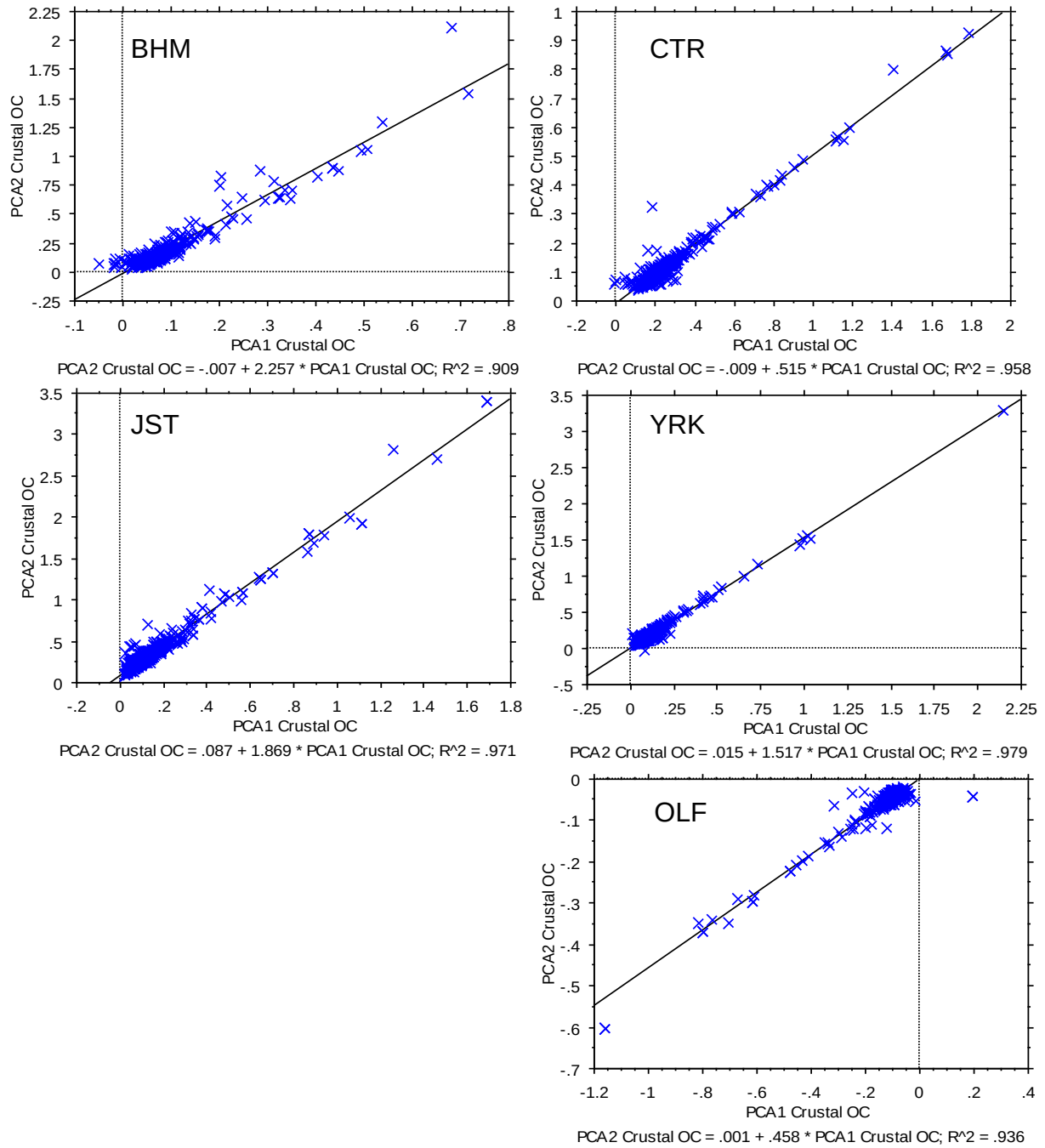


Figure S14. Comparison of PCA1 and PCA2 crustal OC. Units are $\mu\text{g m}^{-3}$.

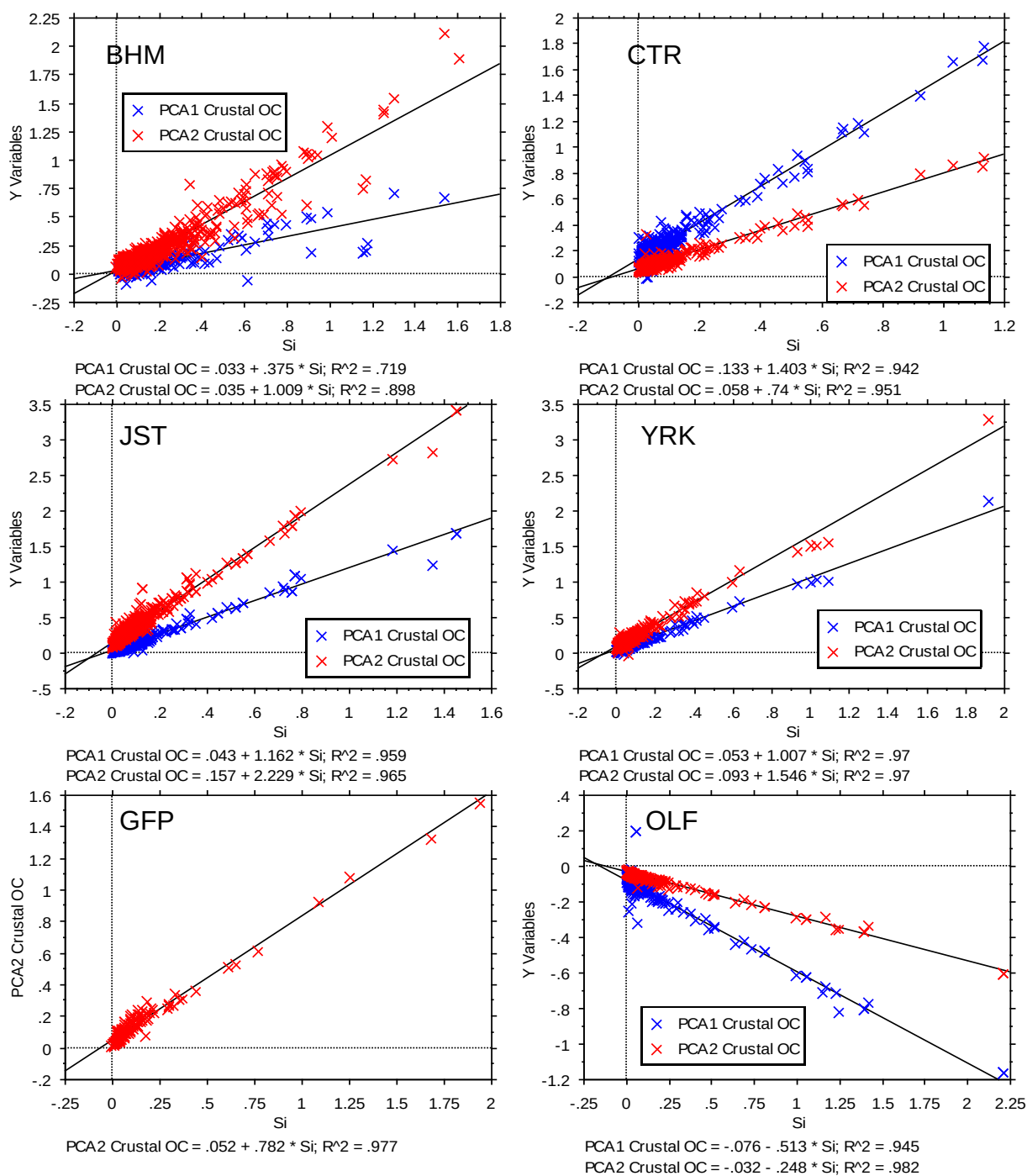


Figure S15. PCA1 and PCA2 crustal OC vs. silicon. Units are $\mu\text{g m}^{-3}$.

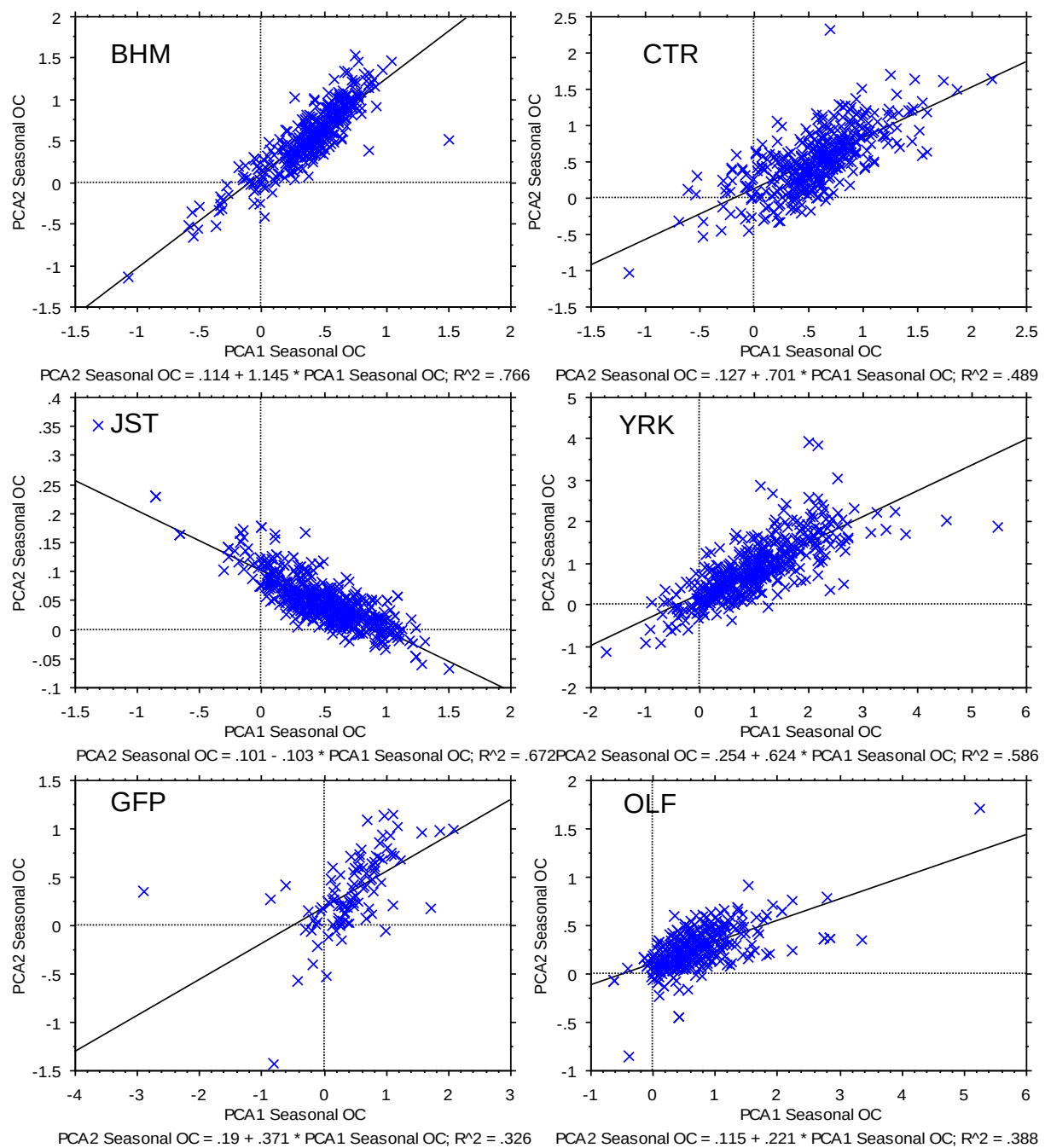


Figure S16. Comparison of PCA1 and PCA2 seasonal-component OC. Units are $\mu\text{g m}^{-3}$.

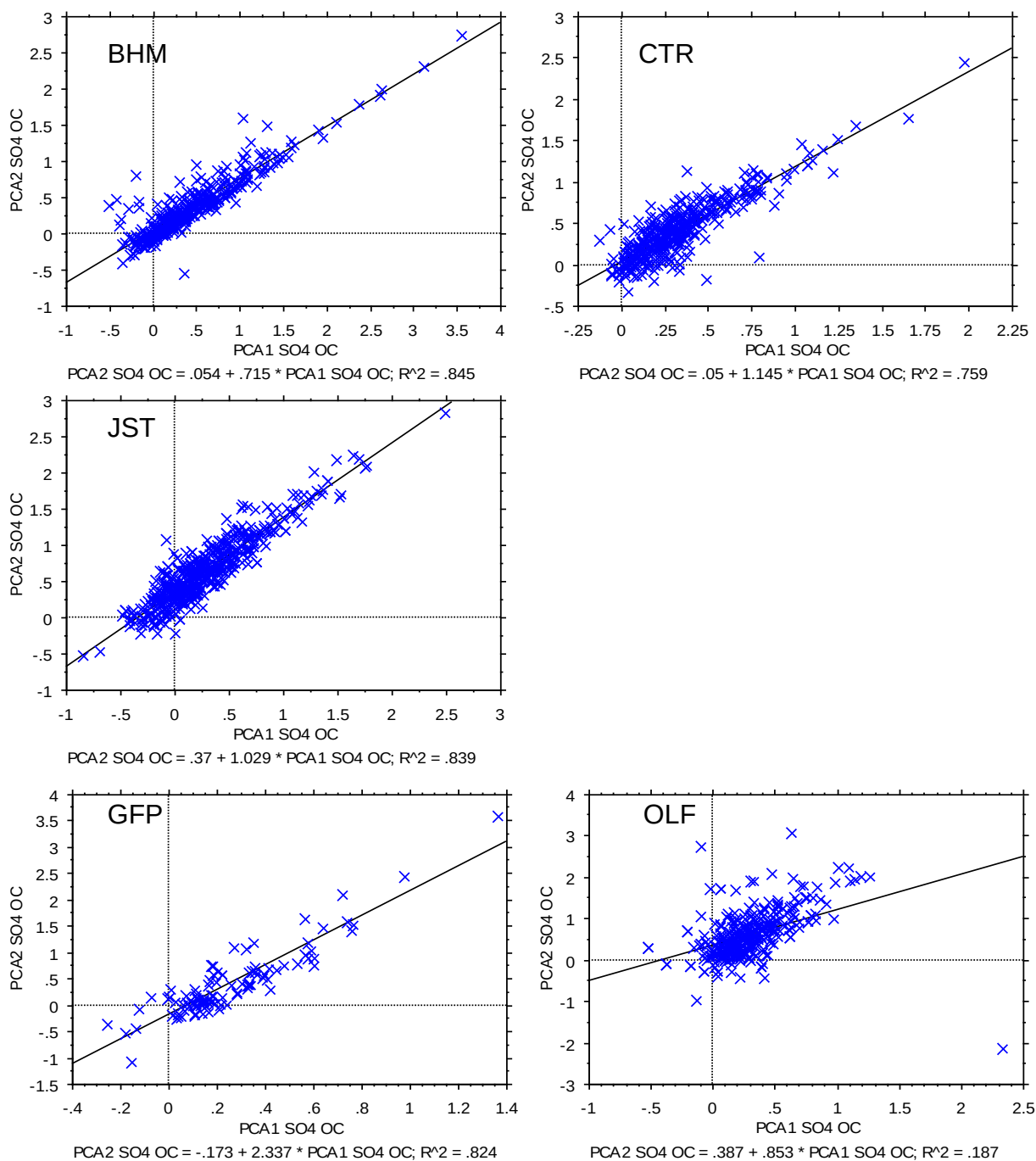


Figure S17. Comparison of PCA1 and PCA2 SO₄ OC. Units are $\mu\text{g m}^{-3}$.

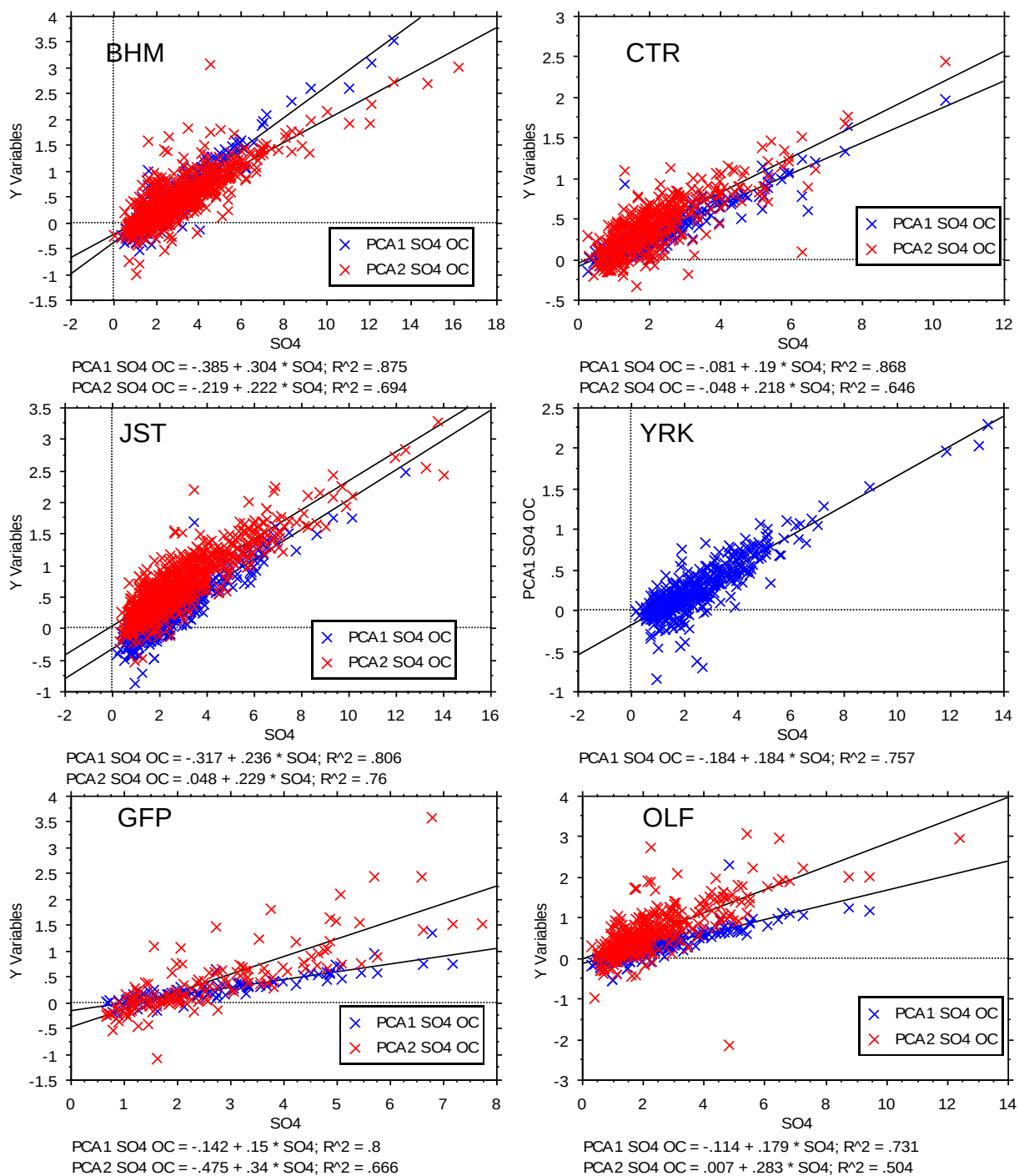


Figure S18. PCA1 and PCA2 SO₄ OC vs. SO₄. PCA2 did not separate seasonal and SO₄ components at YRK. Units are $\mu\text{g m}^{-3}$.

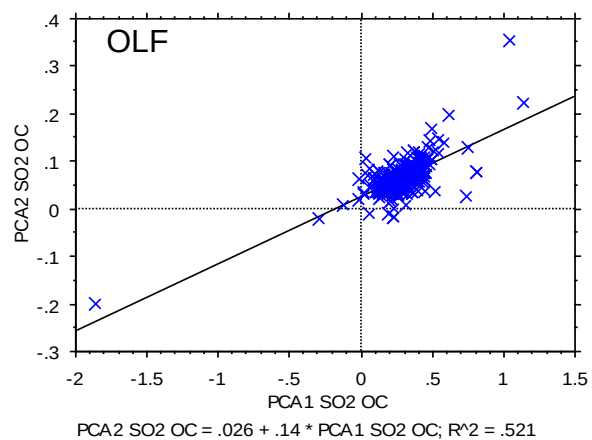
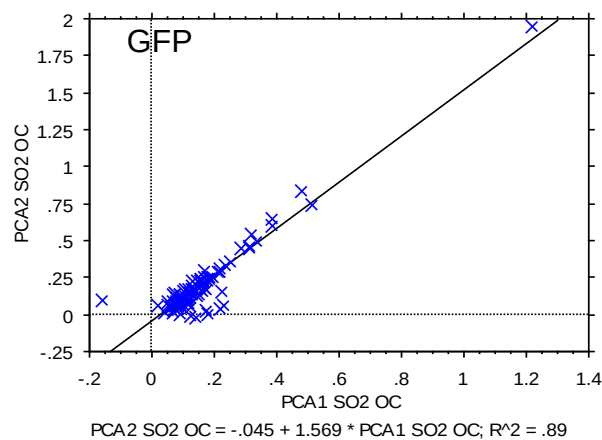
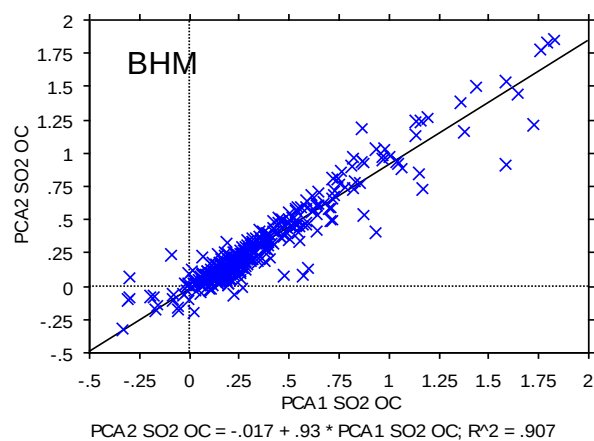


Figure S19. Comparison of PCA1 and PCA2 SO₂ OC. Units are $\mu\text{g m}^{-3}$.

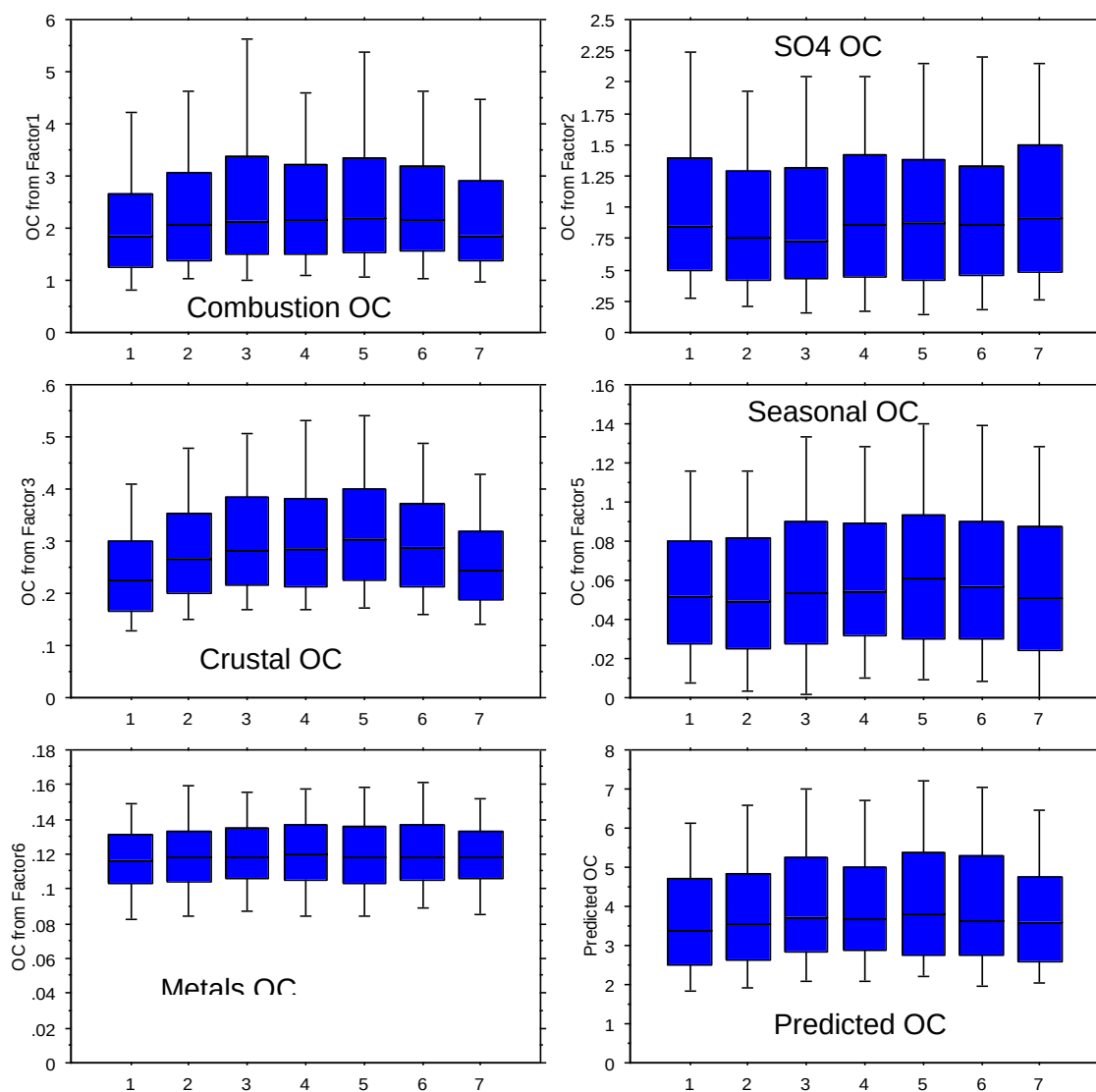


Figure S20. OC associated with PCA2 factors at JST vs. day of week, 1999 – 2013. Day of week begins with Sunday (1). Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Units are $\mu\text{g m}^{-3}$.

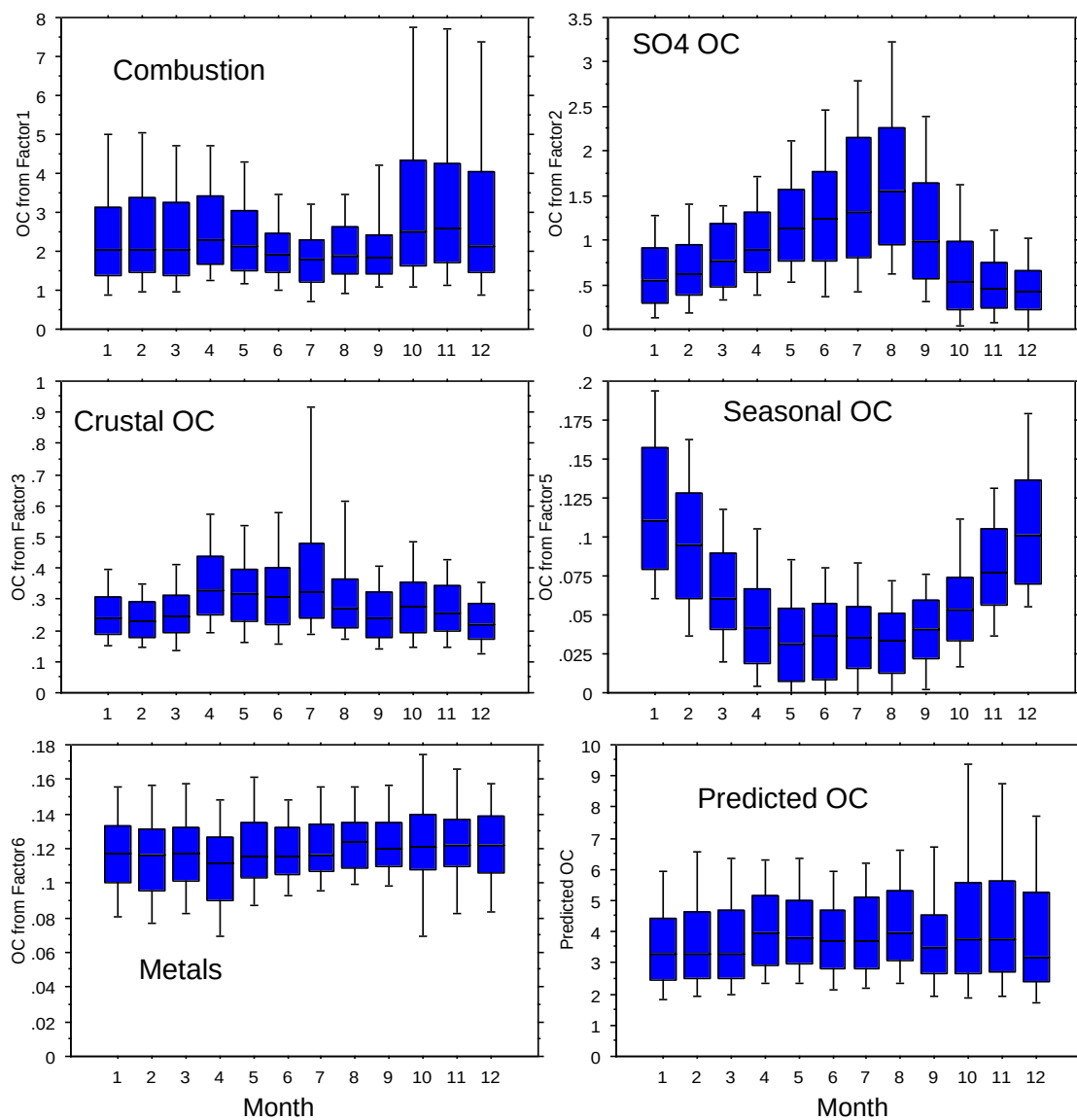


Figure S21. OC associated with PCA2 factors at JST vs. month, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Units are $\mu\text{g m}^{-3}$.

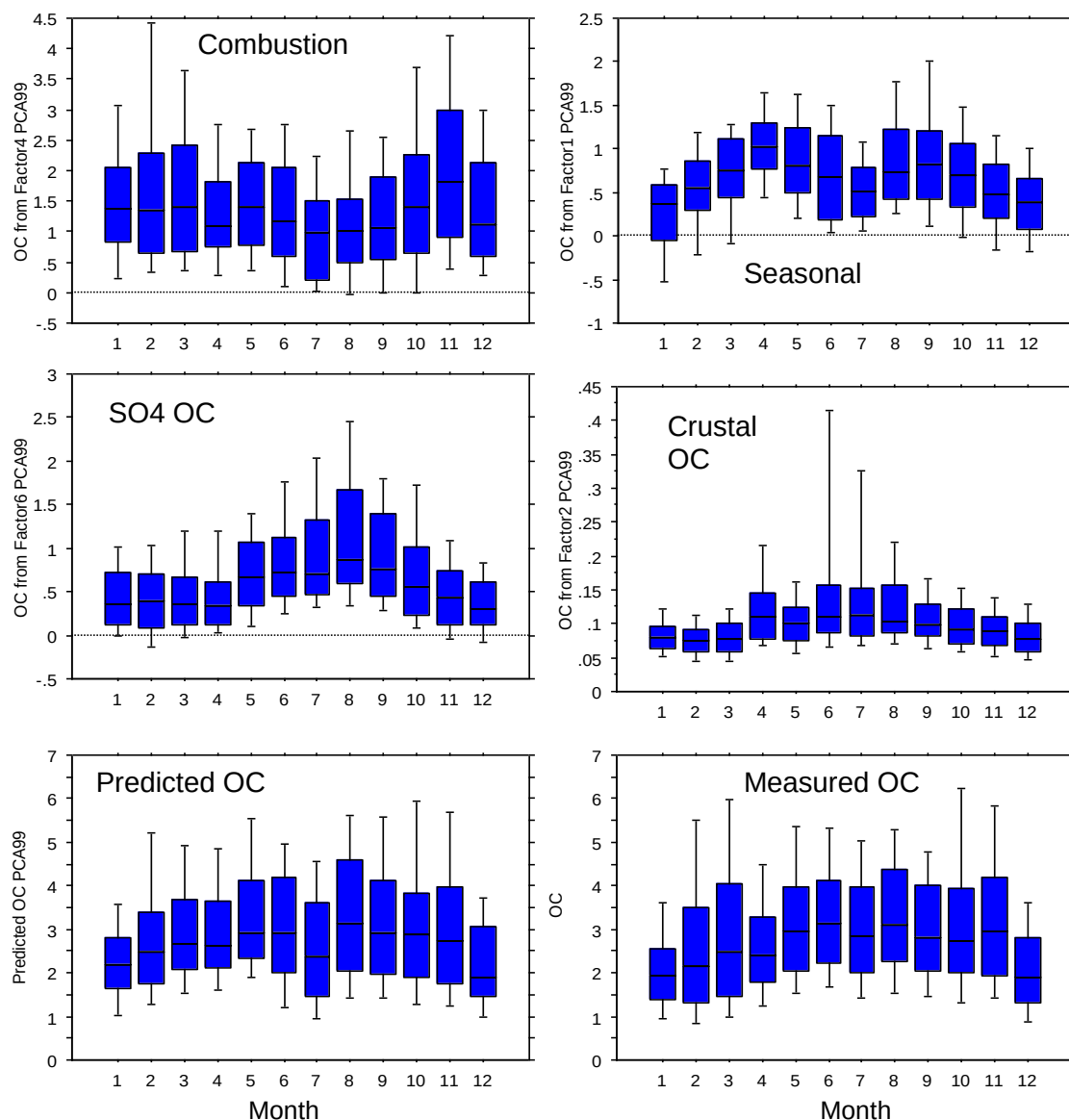


Figure S22. OC associated with PCA2 factors at CTR vs. month, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Units are $\mu\text{g m}^{-3}$.

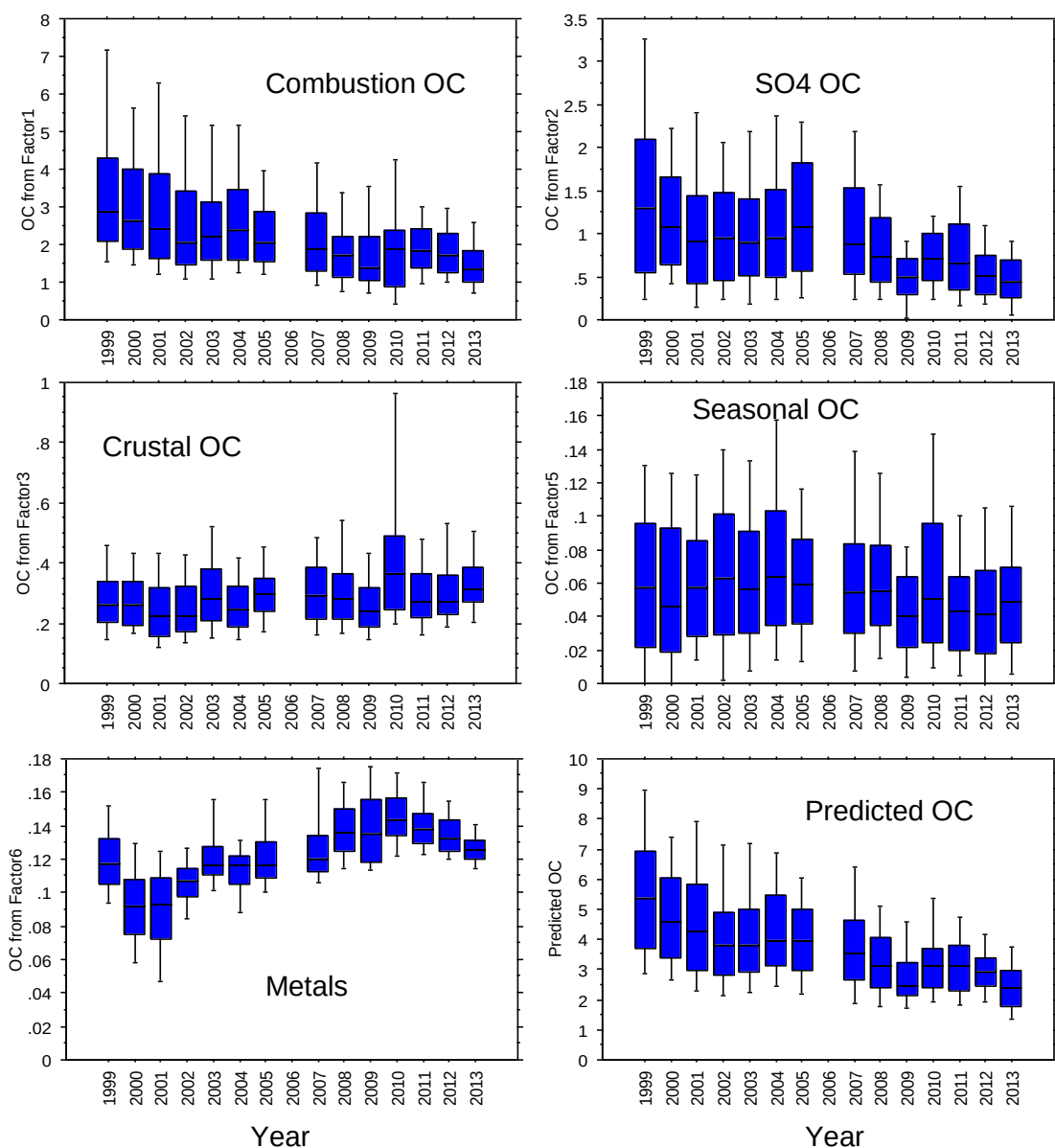


Figure S23. OC associated with PCA2 factors at JST vs. year, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. No predictions were made for 2006 due to missing CO and NO measurements. Units are $\mu\text{g m}^{-3}$.

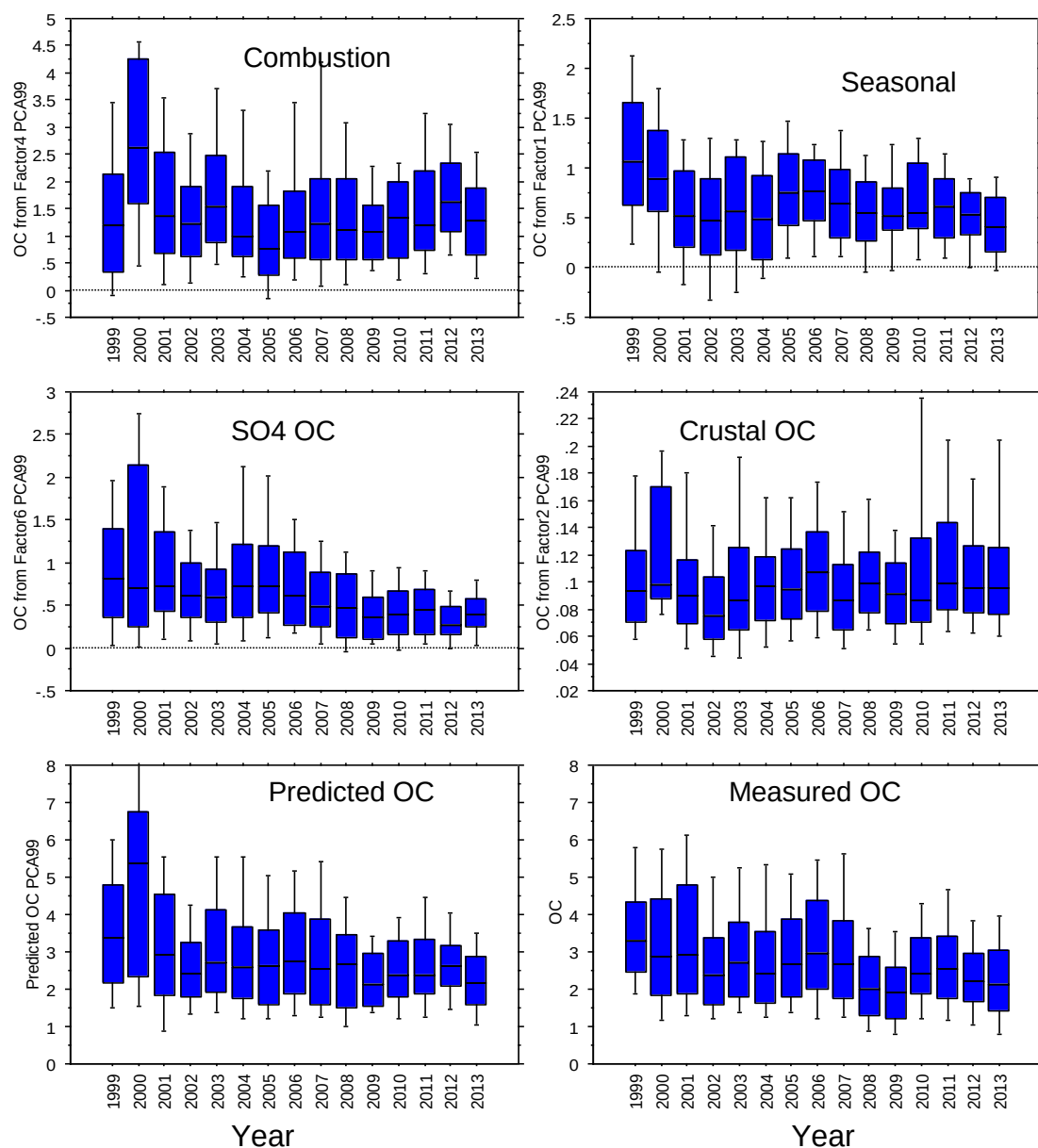


Figure S24. OC associated with PCA2 factors at CTR vs. year, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Years with more limited numbers of sampling days are 2000 (n=11), 2009 (n=31), and 2010 (n=38). Units are $\mu\text{g m}^{-3}$.

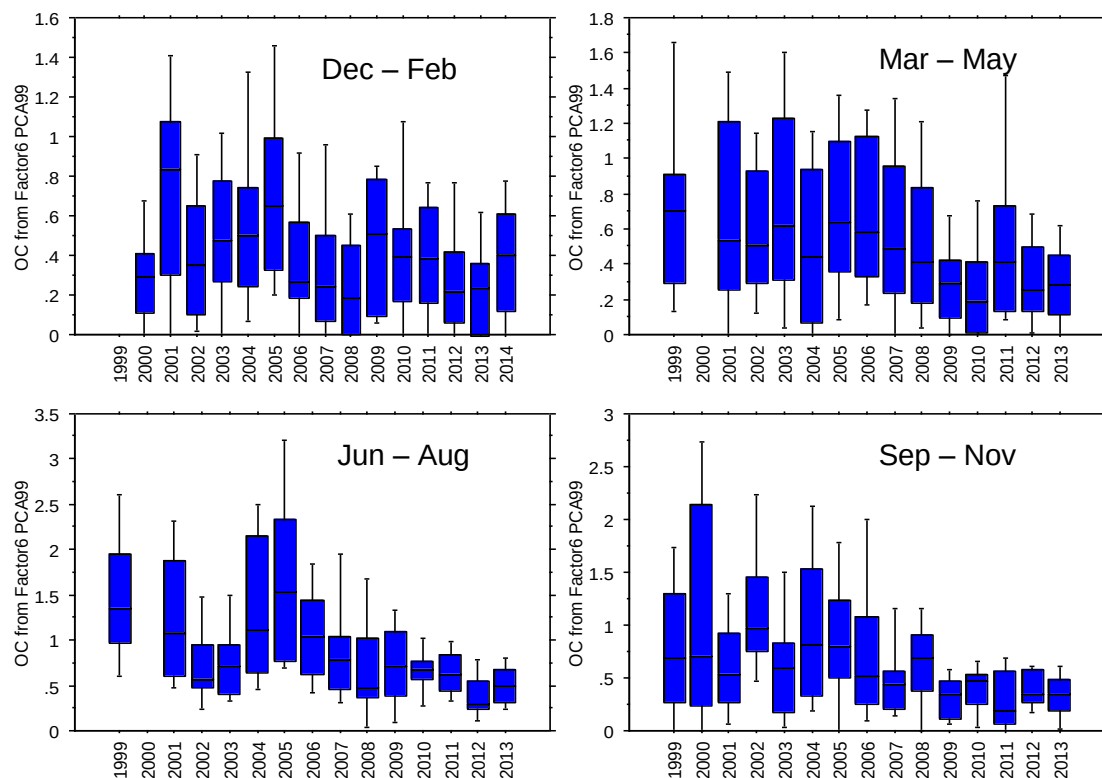


Figure S25. OC associated with PCA2 SO₄ factor at CTR vs. year, 1999 – 2013, shown by season. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Years with more limited numbers of sampling days are 2000 (n=11), 2009 (n=31), and 2010 (n=38). Year 2014 is represented by samples from December 2013. Units are $\mu\text{g m}^{-3}$.

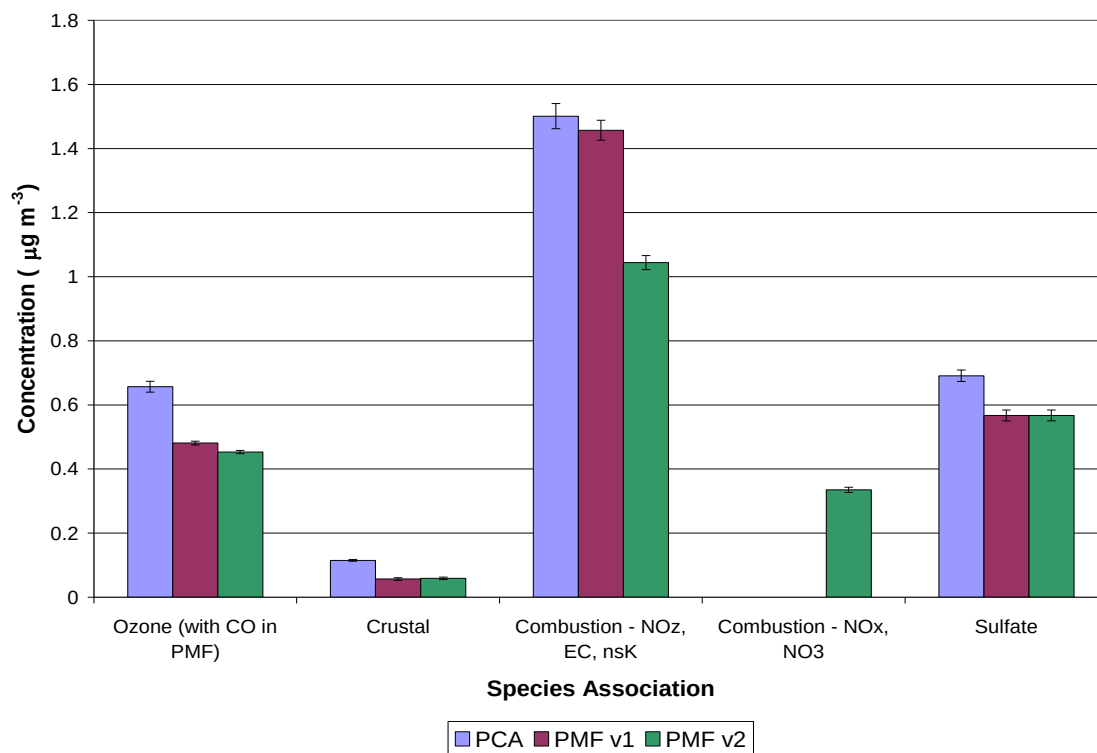


Figure S26. Comparison of CTR mean OC concentrations apportioned to PCA and PMF components. Uncertainties are one standard error of the means.

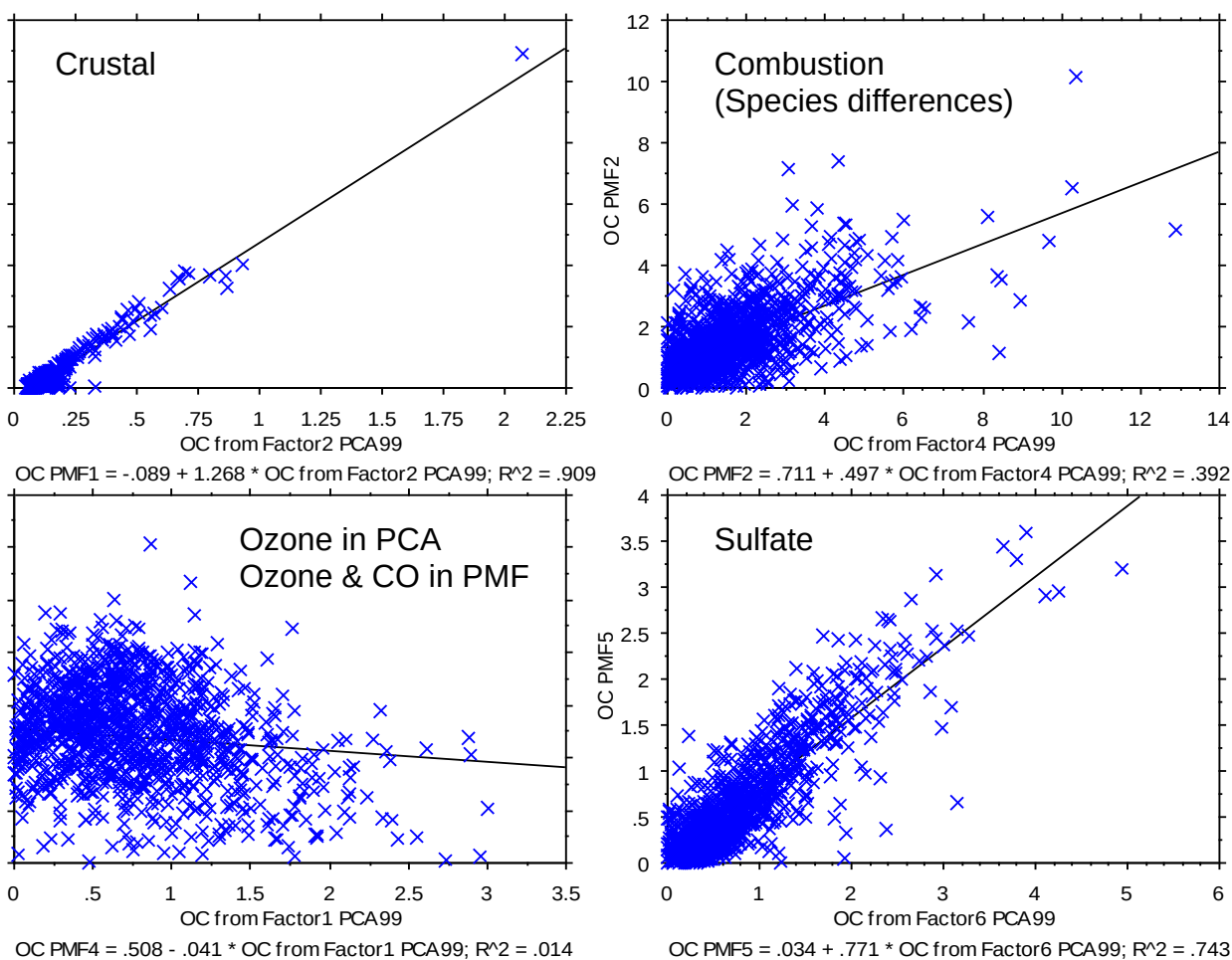


Figure S27. Comparison of CTR OC apportioned to PMF and PCA factors. PMF and PCA factor numbers are arbitrary and are matched by species associations. Units are $\mu\text{g m}^{-3}$.

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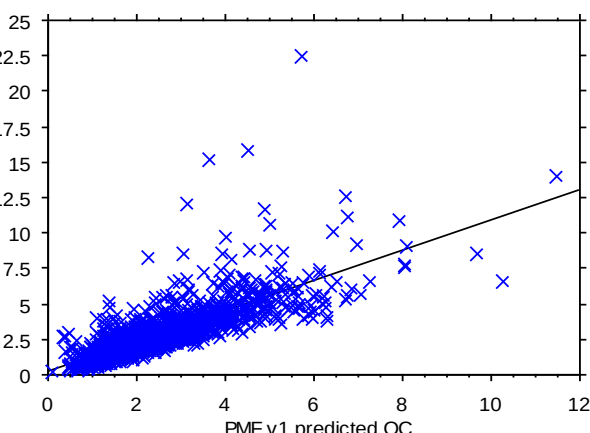
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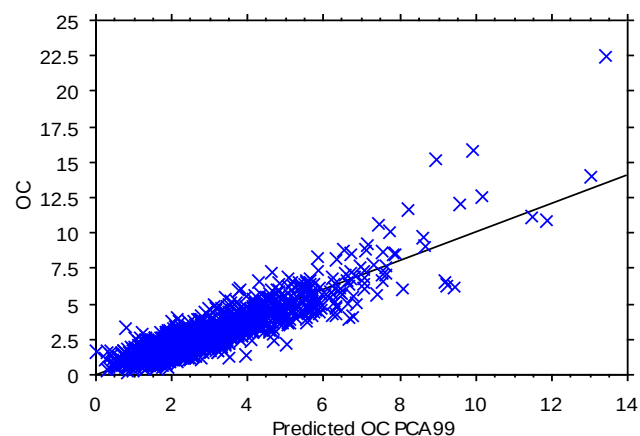
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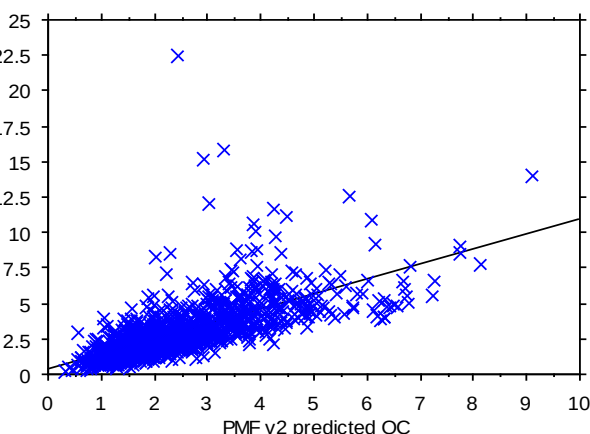
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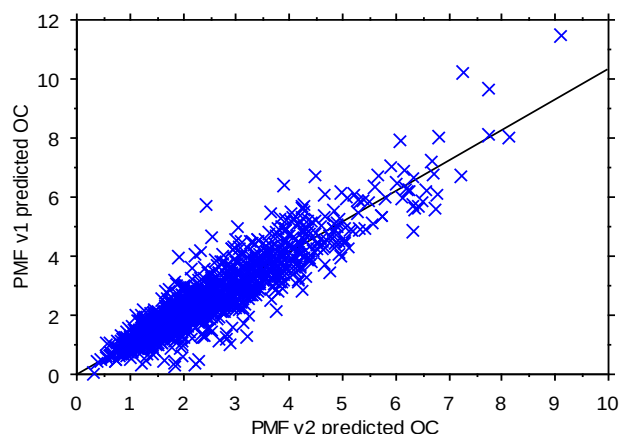
$$OC = .239 + 1.064 * PMF\ v1\ predicted\ OC; R^2 = .607$$



$$OC = -3.023E-6 + 1 * Predicted\ OC\ PCA99; R^2 = .8$$



$$OC = .347 + 1.06 * PMF\ v2\ predicted\ OC; R^2 = .482$$



$$PMF\ v1\ predicted\ OC = .019 + 1.03 * PMF\ v2\ predicted\ OC; R^2 = .85$$

Figures S28. Prediction of measured CTR OC by PMF and PCA. Both PMF applications underpredict the highest OC concentrations (upper left; lower left). The two PMF applications provide comparable predictions (lower right). Units are $\mu\text{g m}^{-3}$.