Supplement of Atmos. Chem. Phys., 16, 215–238, 2016
http://www.atmos-chem-phys.net/16/215/2016/
doi:10.5194/acp-16-215-2016-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

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

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Table S1. Aggregated estimates of composite OC and SOA (SOC) from various regional and local studies in the southeastern US using different methodologies. Unless otherwise stated, SOA (SOC) includes isoprene products and other products from reactions involving terpenoids and anthropogenic VOCs. Averages reported are shown for time periods listed. Investigators have used different terminologies with analytical methods (e.g., Figure S1), and analyses over different time periods, so that comparison of the OC percentages is necessarily qualitative to illustrate a range of SOC fractions, including natural components. A detailed comparison of methods, time periods, and analysis limitations is not attempted within the scope of the present study.

Investigators	**Time Period**	**Method**[a]	**Rural**		**Urban**		**Comments and Notes**
			SOC or SOA (μg m^{-3})	**%OC**	**SOC or SOA** (μg m^{-3})	**%OC**	
Lim and Turpin (2002)	Summer 99	EC Tracer	--	--	8.3	46	Regression by Deming method—ATL (JST)[e]
Saylor et al. (2006)	02	EC Tracer	1.1	30	1.8	32	Regression by York method- ATL, BHM; CTR,YRK
Zheng et al. (2006)	Sept 03-Jan 04	CMB-MM OA-POA	5.7	34	6.5	36	BHM, ATL; CTR SOC may include POA
Gao et al. (2006)	June 04	Filter MS-GC tracer; LCMS	0.3	9.1	0.2	6.3	JST, BHM; CTR total identified OA as SOA; mostly terpene derivatives
Weber et al. (2007)	June 04	Filter WSOC	--	--	2.8	58	ATL; (SOA=WSOC, estimated 70-80% biogenic)
Yu et al. (2007)	<06	Semi-empirical regional EC tracer	--	--	1.01[c]	35	semi-empirical continental-Southeast 7 sites GA, AL, SouthTN
Ding et al (2008)	04-05	CMB-MM; ^{14}C	0.6 (SOC_f) 2.5 (SOC_c)	78[g]	2.3 (SOC_f) 2.2 (SOC_c)	66[g]	SOC=fossil (SOC_f)+ contemporary (SOCc) BHM,ATL; CTR
Blanchard et al. (2008)	01-04	EC tracer; regression; mass balance; ^{14}C[b]	--	36-41	--	15-48	Annual 01-04. ATL, BHM; CTR; range depends on method used
Kleindienst et al. (2010)	05	Filter GCMS tracer; SOC mass fraction from lab. study	2.37[d]	42	2.05[d]	20	BHM, CTR (reported as SOA)
Chan et al. (2010)	Aug-Sept 08	Filter GCMS GCToFMS/tracers	0.1-1.4	(10.7 isoprene)	0.1-0.9	(7.4 isoprene)	ATL YRK; estimates from sum of isoprene products or from Kleindienst et

							al. tracer (day night separation)g
Zhang et al. (2010)	07	Filter WSOC	--	--	--	56	15 sites in Southeast (light absorbing WSOC=SOC)
Blanchard et al. (2013)	02-11	Integrated gas particle CMB	--	--	1.8	45	ATL composite OA = 4 ug/m3 exclude unaccounted for mass
Budusulistiorini et al (2013)	Summer 11	ACSM (AMS)	--	--	9	82 (33 isoprene)[f]	ATL. Mostly correlations between identified components of aerosol mass from PMF
Lin et al (2013)	Summer 10	Filter GC-EI-MS	--	(12-19 isoprene)[f]	--	--	YRK; OM% isoprene derivatives only; both low and High NOx contributions
Lewandowski et al. (2014)	May-Aug 05	Filter GCMS tracers; SOC mas fraction from lab study	2.37[d]	36	1.8[d]	17	ATL, BHM; CTR; differentiates biogenic SOC from anthropogenic SOC
Xu et al. (2015a, b)	June-July 13 (SOAS); ~ 1 yr (12-13-SCAPE)	HRToFMS and ACSM	4.5	89 (18 isoprene)[f]	9.1	69 (21 isoprene)[f]	ATL, CTR, (summer); PM1; OA—segregated with SOA sum of LO-OOA, MO-OOA ; seasonal isoprene OA only in summer; particle nitrate OA discussed
Liao et al. (2015)	May-June 12	(NOAA) PALMS	--	(2.2 IEPOXSO4)	--	--	Aircraft near ground values IEPOX-SO4 only
Hu et al (2015)	June-July 2013	AMS low NOx (Quant. PMF of AMS signal)	--	(17 IEPOX SO4)	--	--	Includes detils of IEPOX SO4 estimation, notes biomass burning ambiguity.
Kim et al. (2015)	Summer-fall 13	Various air-ground meas./ modeling	--	60	--	--	Integration of ground and aircraft obs.in SE. Values represent 1.5-3 km altitude; biogenic includes isoprene and terpenoid derivatives—anthropogenic SOA excluded
This study (2015)	00-13	Tracer/mass balance/PCA	2.9	39	3.9	26	CTR, ATL; SOC based on PCA1; SOC=

using SEARCH carbon and associated data	seasonal and SO4 contributions

[a]Methods include analytical techniques, air quality modeling and data analysis and interpretation. Instrumentation for analysis includes thermal differentiation for OC and BC, gas chromatogrphy-mass spectroscopy (GCMS, high resolution time of flight mass spectroscopy (HRToFMS), water extraction and liquid chromatography-mass spectroscopy, aerosol mass spectroscopy (AMS), carbon isotope analysis, particle analysis laser mass spectrometer (PALMS), and aerosol chemical speciation monitor (ACSM).
[b]Data set included SEARCH public archives.
[c]Southeastern region (15 monitoring sites for OC and EC) stated in urban category, but includes rural sites.
[d]Reported as $\mu gC/m^{-3}$; estimates of SOC mainly isoprene and monoterpene derivatives
[e]The SEARCH network sites included in studies were Jefferson Street, Atlanta, GA (JST or ATL), Centreville, AL (CTR), Yorkville, GA (YRK) and Birmingham, AL (BHM). A site locator map and description is found in Hidy et al. (2014).
[f]Parentheses isoprene derivative component of OC or OA (OM) only from AMS assignment.
[g]%OC is calculated as sum of fossil and contemporary SOC; values average over four seasons

Table S2. Primary air pollutant emissions within AL, GA, MS, and NW FL in 2013. Units are 1000 metric tons per year. $PM_{2.5}$ species were determined from NEI emissions of $PM_{2.5}$ mass using SPECIATE or from the EPA MOVES model (EC and OC from on-road diesel and gasoline). Zero values indicate emissions less than one-half the smallest reported significant figure (1 or 0.1 thousand metric tons). Source categories are defined in Blanchard et al. (2013).

			Speciated $PM_{2.5}$ Emissions								
Sector	CO	NO_x	EC	OC	K	Al	Ca	Fe	Si	SO_2	VOC
Agriculture	0	0	0.0	1.1	0.8	3.3	0.9	2.1	9.1	0	0
Area	294	11	3.9	16.4	2.4	0.0	0.0	0.0	0.1	0	246
Vegetation & soil	526	43	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0	5008
Commercial	5	7	0.1	0.1	0.0	0.0	0.0	0.0	0.0	4	0
Dust	0	0	0.3	6.3	1.6	4.5	6.6	4.0	14.5	0	0
EGU	25	98	0.1	0.2	0.0	0.2	0.2	0.1	0.4	247	2
Fires	1996	39	8.1	89.0	2.7	0.2	0.3	0.1	0.2	16	314
Industrial	155	162	1.3	4.8	0.7	0.8	0.4	0.4	2.3	110	132
Nonroad	686	147	5.2	2.4	0.0	0.0	0.0	0.0	0.0	3	107
Residential	35	10	0.3	2.4	0.0	0.0	0.0	0.0	0.0	0	6
On-road diesel	43	125	4.1	1.4	0.0	0.0	0.0	0.0	0.0	0	10
On-road gas	1129	133	0.3	1.3	0.0	0.0	0.0	0.0	0.0	1	60
Sum	4893	775	24	125	8	9	8	7	27	382	5886

Table S3. 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.

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-SO4	0.843	0.425	0.331	0.305	0.690	0.211	0.127	0.057	0.514
BHM	EC-SO4	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-SO4	0.772	0.492	0.350	0.293	0.360	0.181	0.181	0.024	0.520
CTR	EC-SO4	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-SO4	0.730	0.181	0.158	0.086	0.554	0.166	0.112	0.057	0.361
GFP	EC-SO4	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-SO4	0.854	0.276	0.191	0.350	0.552	0.099	0.084	0.144	0.365
JST	EC-SO4	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-SO4	0.702	0.110	0.129	0.257	0.523	0.060	0.066	0.016	0.402
OAK	EC-SO4	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-SO4	0.721	0.361	0.316	0.321	0.651	0.238	0.103	0.100	0.559
OLF	EC-SO4	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-SO4	0.568	0.006	0.030	0.089	0.700	0.088	0.043	0.024	0.569
PNS	EC-SO4	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-SO4	0.800	0.619	0.451	0.454	0.568	0.304	0.215	0.104	0.502
YRK	EC-SO4	0.764	0.414	0.293	0.453	0.782	0.253	0.097	0.096	0.391

Table S4. PCA1 and PCA2 orthogonal factors for SEARCH BHM daily data. PCA1: n = 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

Table S5. PCA1 and PCA2 orthogonal factors for SEARCH CTR daily data. PCA1: n = 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

Table S6. PCA1 and PCA2 orthogonal factors for SEARCH GFP daily data. PCA1: n = 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

Table S7. PCA1 and PCA2 orthogonal factors for SEARCH JST daily data. PCA1: n = 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	

Table S8. PCA1 and PCA2 orthogonal factors for SEARCH OAK daily data. PCA1: n = 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

Table S9. PCA1 and PCA2 orthogonal factors for SEARCH OLF daily data. PCA1: n = 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

Table S10. PCA1 and PCA2 orthogonal factors for SEARCH PNS daily data. PCA1: n = 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

Table S11. PCA1 and PCA2 orthogonal factors for SEARCH YRK daily data. PCA1: n = 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

Table S12. 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 μg m^{-3}. Factors are normalized (mean 0, variance 1), so units are μg m^{-3} factor-unit^{-1} for ΔOC/Δ 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
Mean OC	2.904 ± 0.040	2.422 ± 0.029	2.129 ± 0.078	3.783 ± 0.052	2.545 ± 0.102	2.274 ± 0.039	2.734 ± 0.107	2.840 ± 0.040
ΔOC/ΔCombustion	1.429 ± 0.040	1.028 ± 0.031	0.918 ± 0.077	1.703 ± 0.048	0.784 ± 0.101	0.971 ± 0.038	2.011 ± 0.128	0.213 ± 0.035
ΔOC/ΔCrustal	0.100 ± 0.043	0.201 ± 0.028	NS	0.140 ± 0.023	NS	-0.116 ± 0.024	NS	0.121 ± 0.021
ΔOC/ΔSeasonal	0.371 ± 0.048	0.455 ± 0.031	0.607 ± 0.069	0.406 ± 0.038	0.424 ± 0.076	0.641 ± 0.033	0.342 ± 0.109	1.270 ± 0.037
ΔOC/ΔSO_2	0.413 ± 0.046	NS	0.161 ± 0.076	0.087 ± 0.038	0.461 ± 0.087	0.284 ± 0.053	NS	NS
ΔOC/ΔSO_4	0.570 ± 0.042	0.263 ± 0.028	0.349 ± 0.093	0.735 ± 0.056	0.594 ± 0.102	0.409 ± 0.047	0.672 ± 0.137	0.634 ± 0.049
ΔOC/ΔMetals	0.182 ± 0.039	NS	NS	NA	0.250 ± 0.095	NS	NA	0.432 ± 0.033
ΔOC/ΔSalt	0.159 ± 0.039	NS	-0.549 ± 0.095	0.669 ± 0.044	-0.268 ± 0.073	-0.122 ± 0.027	-0.532 ± 0.092	0.212 ± 0.032
ΔOC/ΔOther[a]	0.132 ± 0.051	NA	0.569 ± 0.074	0.134 ± 0.031	NA	NA	NA	0.131 ± 0.022

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

Table S13. Statistically significant regression results for multiple regression of OC on PCA2 factor scores. Factor composition is defined in Tables S2 through S9. Units for mean OC are μg m^{-3}. Factor scores are normalized (mean 0, variance 1), so the units are μg m^{-3} factor-unit^{-1} for ΔOC/Δ factor. NS = not significant; NA = not applicable (PCA 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
Mean OC	3.938 ± 0.034	2.990 ± 0.023	2.296 ± 0.038	4.035 ± 0.024	2.625 ± 0.041	2.302 ± 0.025	2.668 ± 0.040	3.000 ± 0.023
ΔOC/ΔCombustion	2.097 ± 0.036	1.442 ± 0.024	0.920 ± 0.049	1.882 ± 0.023	1.641 ± 0.047	0.687 ± 0.023	1.685 ± 0.042	0.626 ± 0.021
ΔOC/ΔCrustal	0.205 ± 0.035	0.105 ± 0.024	0.176 ± 0.036	0.253 ± 0.024	NS	-0.054 ± 0.027	NS	0.180 ± 0.023
ΔOC/ΔSeasonal	0.492 ± 0.034	0.555 ± 0.022	0.436 ± 0.038	0.051 ± 0.023	0.596 ± 0.043	0.241 ± 0.025	0.519 ± 0.042	1.193 ± 0.022
ΔOC/ΔSO_2	0.420 ± 0.033	NS	0.274 ± 0.045	NS	NS	0.084 ± 0.023	0.146 ± 0.037	0.045 ± 0.021
ΔOC/ΔSO_4	0.841 ± 0.039	0.635 ± 0.023	0.776 ± 0.042	0.790 ± 0.023	0.656 ± 0.044	0.937 ± 0.026	NS	NS
ΔOC/ΔMetals	0.635 ± 0.042	NS	0.315 ± 0.032	0.103 ± 0.040	NS	0.218 ± 0.026	NS	0.389 ± 0.022
ΔOC/ΔSalt	NA	NA	NA	NA	NA	NA	NA	NA
ΔOC/ΔOther[a]	NA	NA	NA	NA	NA	NA	0.375 ± 0.039	0.237 ± 0.020

Table S14. Mean OC associated with components identified by PCA1 (2008 – 2013) and PCA2 (1999 – 2013). NS = not statistically significant, NA = not applicable (component 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%

a. Negative due to inverse associations of OC with crustal and salt components at (Tables S7 and S13).
b. JST PCA2 seasonal OC is associated with NO_3; JST PCA2 SO_4 component includes OC associated with O_3 (Table 2).
c. PNS and YRK PCA2 seasonal components include OC associated with SO_4 (Table 2).

Table S15. PCA3 orthogonal factors for SEARCH JST daily data. PCA3, 1999 – 2008: n = 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

Table S16. PCA4 (including NH_3) and PCA5 (excluding NH_3; including daily average O_3) orthogonal factors for SEARCH CTR daily data, 2004 - 2013. PCA4: n = 724, 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	

Table S17. Comparison of PCA1 CTR source apportionment to Xu et al. (2015a, b).

	OC from each PCA factor (% of mean OC)			Unc (%)		AMS OC (%)	AMS OA (%)
PCA Factor	2008-13	2013	SOAS		AMS Factor[a]	SOAS	SOAS
Combustion	52	53	38	18	MO-	34	39
					BBOA	11	10
Sulfate	11	13	13	6	Isop	20	18
Seasonal	14	22	23	15	LO-	35	32
Crustal	23	13	20	4			
Fitted sum	100	100	94				
Mean OA[a]	NA	NA	NA				5.0
Mean OC[a]	2.41	2.26	2.63			2.31	
Mean OC in PCA subset[a]	2.43	2.32	2.61			NA	
OM/OC	1.58	1.66	1.36			2.16	
N days for mean OC	606	156	40				
N days in PCA subset	383	105	29				

a. MO-OOA (MO-), biomass burning OA (BBOA), isoprene OA (Isop), and LO-OOA (LO-)

b. µg m^{-3}

Table S18. Comparison of PCA1 JST source apportionment to Xu et al. (2015a, b).[a]

	OC from each PCA factor (% of mean OC)				Unc (%)		AMS OC (%)		AMS OA (%)	
PCA Factor	2008-13	2012	MJ 2012	ND 2012		AMS Factor[b]	MJ 2012	ND 2012	MJ 2012	ND 2012
Combustion	38	41	29	57	12	HOA	14	25	10	19
						MO-	22	26	27	31
Salt[c]	23	21	20	7	11	BBOA	10	10	10	9
Other[d]	4	4	2	12	2	COA	14	23	11	20
Sulfate	10	6	9	1	7	Isop	19		21	
SO_2	3	1	1	1	1					
Seasonal	17	18	25	11	15	LO-	21	17	21	19
Crustal	6	5	3	2	4					
Fitted sum	100	95	89	91						
Mean OA[e]	NA	NA	NA	NA					9.1	7.9
Mean OC[e]	2.88	2.98	3.36	3.65			4.70	5.45		
Mean OC in PCA subset[e]	3.78	2.90	3.36	3.65			NA	NA		
OM/OC	1.51	1.37	1.34	1.51			1.93	1.40		
N days for mean OC	904	114	8	8						
N days in PCA subset	516	109	8	8						

a. Sample periods are May 10 - Jun 2, 2012 (MJ2012) and Nov 6 - Dec 4, 2012 (ND 2012)

b. MO-OOA (MO-), biomass burning OA (BBOA), isoprene OA (Isop), and LO-OOA (LO-)

c. Associated with K, Mg, Cl

d. Associated with Na

e. $\mu g\ m^{-3}$

Table S19. Comparison of YRK PCA1 source apportionment to Xu et al. (2015a, b).[a]

	OC from each PCA factor (% of mean OC)						AMS OC (%)		AMS OA (%)	
PCA Factor	2008-13	2012	JJ 2012	DJ 2012	Unc (%)	AMS Factor[b]	JJ 2012	DJ 2012	JJ 2012	DJ 2012
Combustion	5	4	2	14	14	MO-	25	42	30	49
Metals[c]	21	20	11	27	2					
Salt[d]	7	5	2	11	7	BBOA		35		30
Other[e]	6	7	7	8	5					
Sulfate	11	6	5	8	11	Isop	38		36	
Seasonal	42	46	47	27	5	LO-	37	23	34	22
Crustal	6	6	11	6	4					
Fitted sum	97	95	84	102						
Mean OA[f]	NA	NA	NA	NA					11.2	3.23
Mean OC[f]	2.33	2.36	3.06	1.78			5.66	1.73		
Mean OC in PCA subset[f]	2.40	2.35	2.97	1.78			NA	NA		
OM/OC	1.78	1.77	1.8	1.56			1.98	1.31		
N days for mean OC	585	119	9	11						
N days in subset	426	97	7	10						

a. Sample periods are June 26 - July 20, 2012 (JJ2012) and December 5, 2012 - January 10, 2013 (DJ2012)

b. MO-OOA (MO-), biomass burning OA (BBOA), isoprene OA (Isop), and LO-OOA (LO-)

c. Associated with Cu

d. Associated with Na, Cl, Mg, K

e. Associated with Zn

f. $\mu g\ m^{-3}$

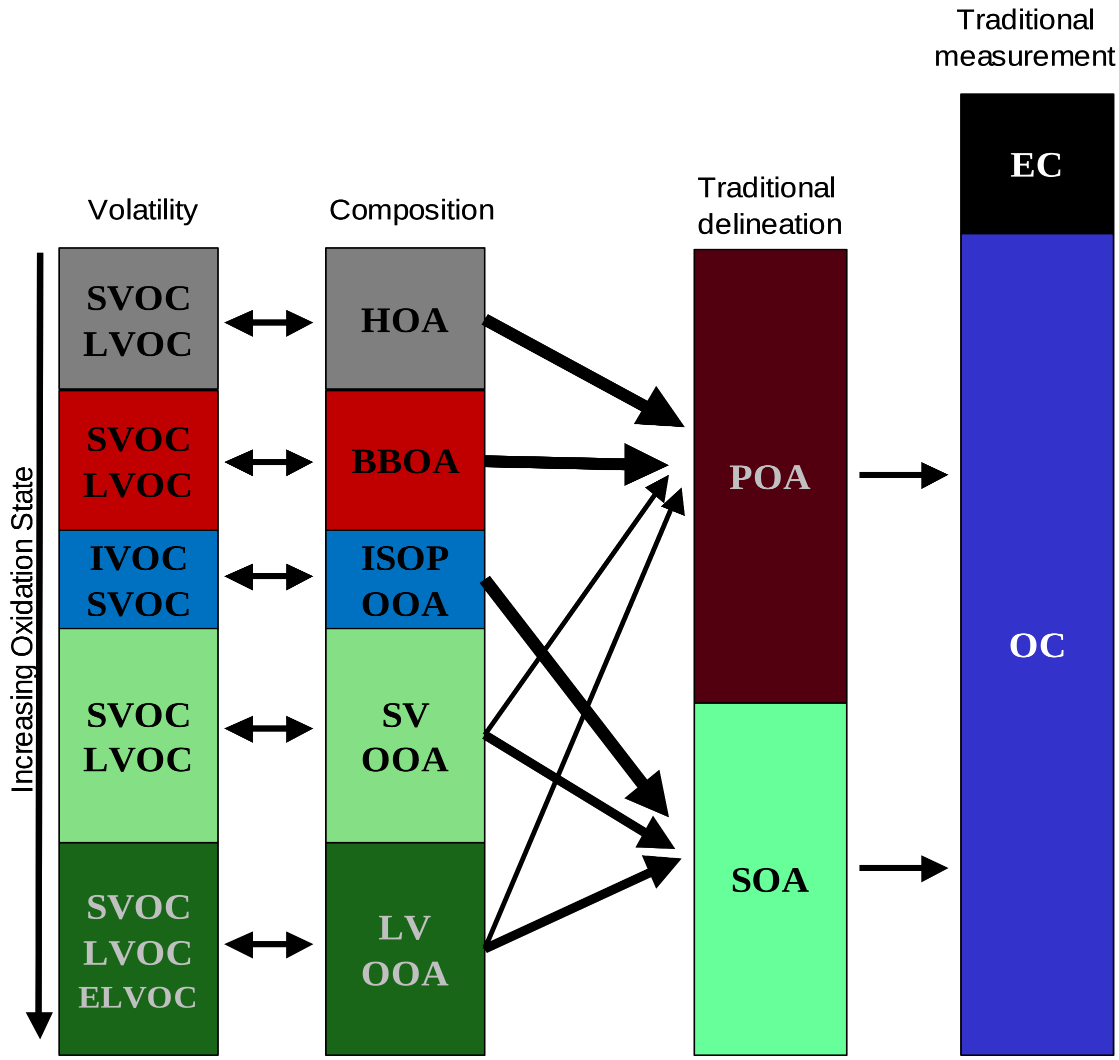


Figure S1. Graphical depiction of various categorizations of OA. Traditional measurements and delineation: EC, OC, POA, SOA as defined in the text. Composition-based categories: HOA, hydrocarbon-like OA; BBOA, biomass burning OA; ISOP OOA, isoprene-derived oxidized OA; SVOOA, semi-volatile oxidized OA (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 O_3 and SO_4, suggestive of more-oxidized OA.

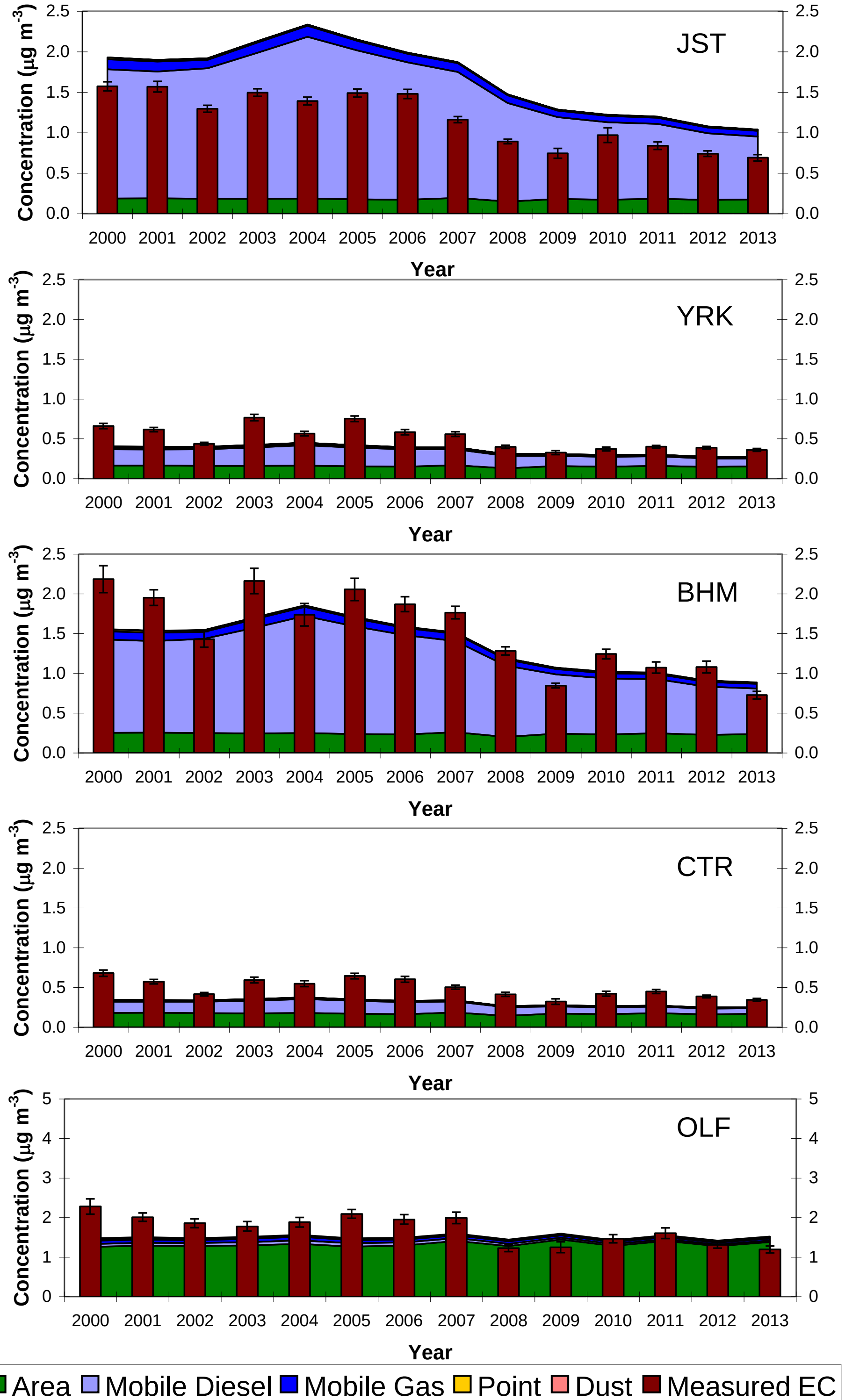


Figure S2. Apportionment of EC trends from chemical mass balance (CMB) receptor model predictions compared with observed mean annual EC concentrations at SEARCH sites. Model predictions were extended to 2012 and 2013 by using model parameters previously fit to data from 2000 – 2011 (Blanchard et al., 2013) along with regional emissions from 2012 and 2013 (Hidy et al., 2014).

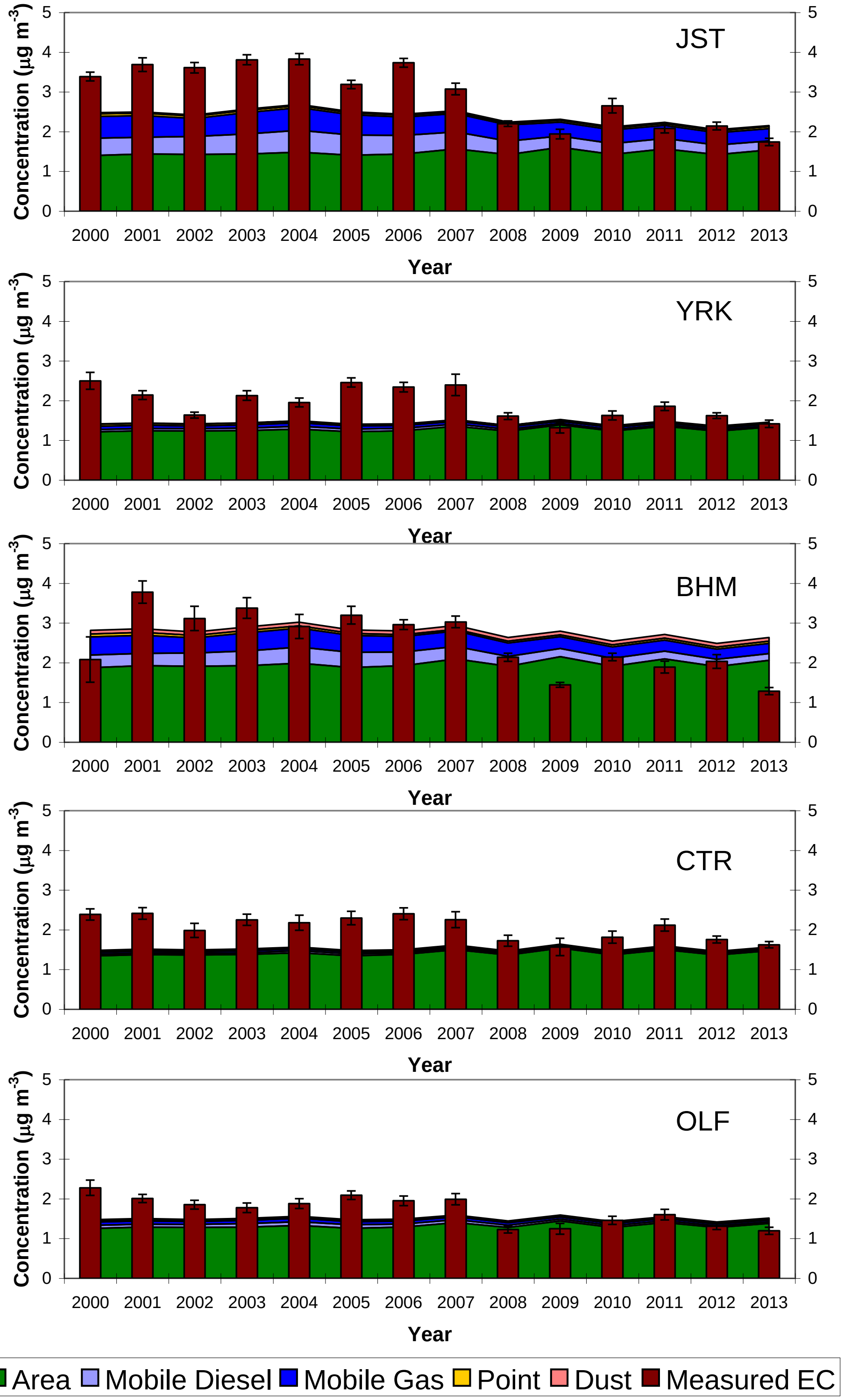


Figure S3. Apportionment of OC trends from chemical mass balance (CMB) receptor model predictions compared with observed mean annual OC concentrations at SEARCH sites. Model predictions were extended to 2012 and 2013 by using model parameters previously fit to data from 2000 – 2011 (Blanchard et al., 2013) along with regional emissions from 2012 and 2013 (Hidy et al., 2014).

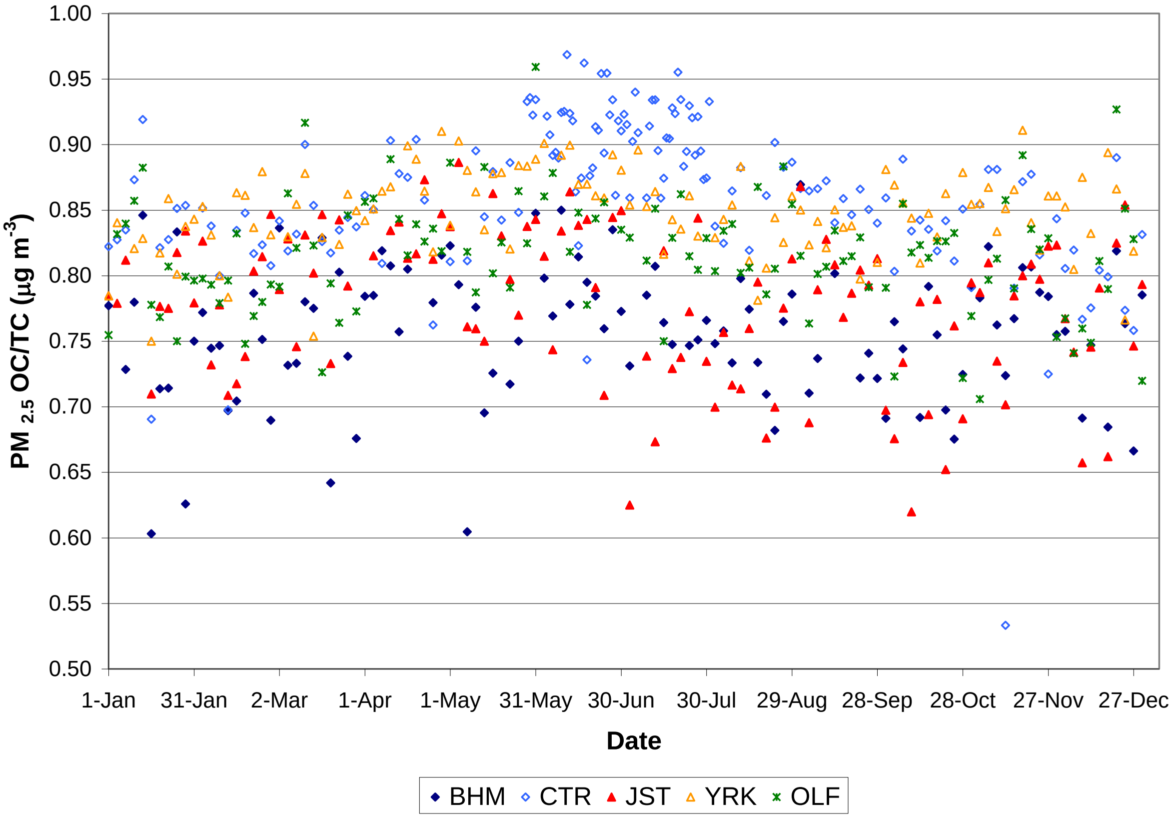
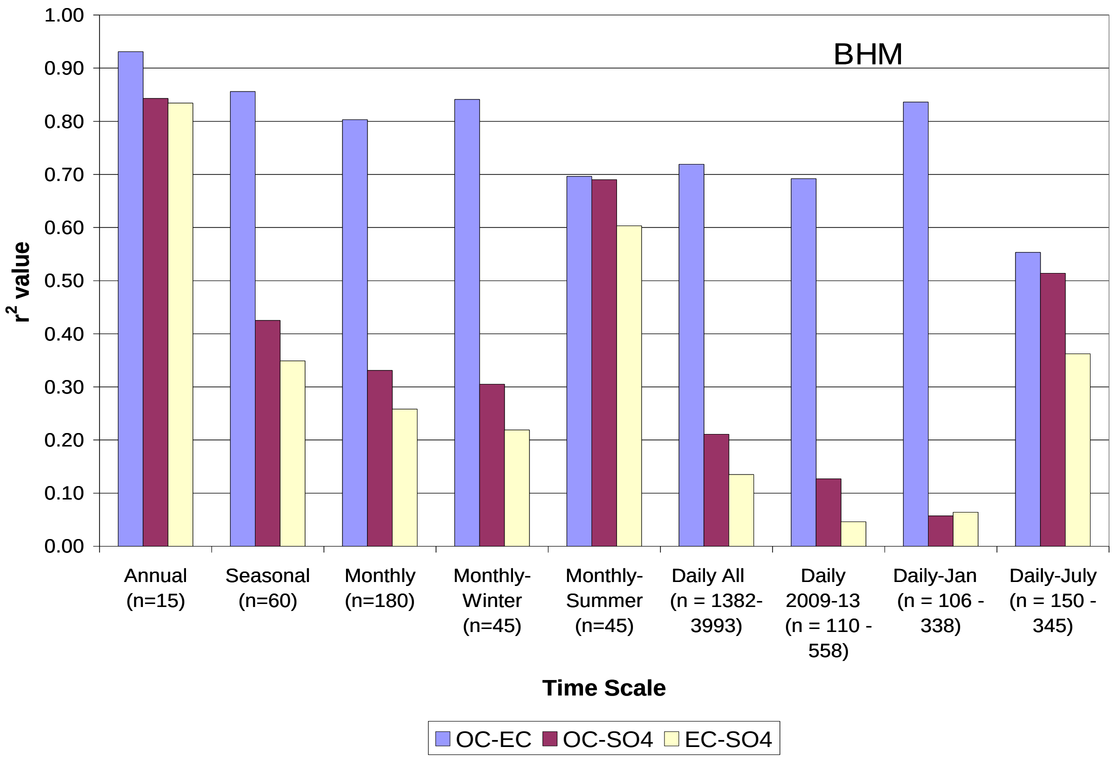


Figure S4. Ratio of OC/TC in daily-average $PM_{2.5}$ filter samples collected during 2013 vs. date. The highest OC/TC ratios occur at CTR during the SOAS campaign.



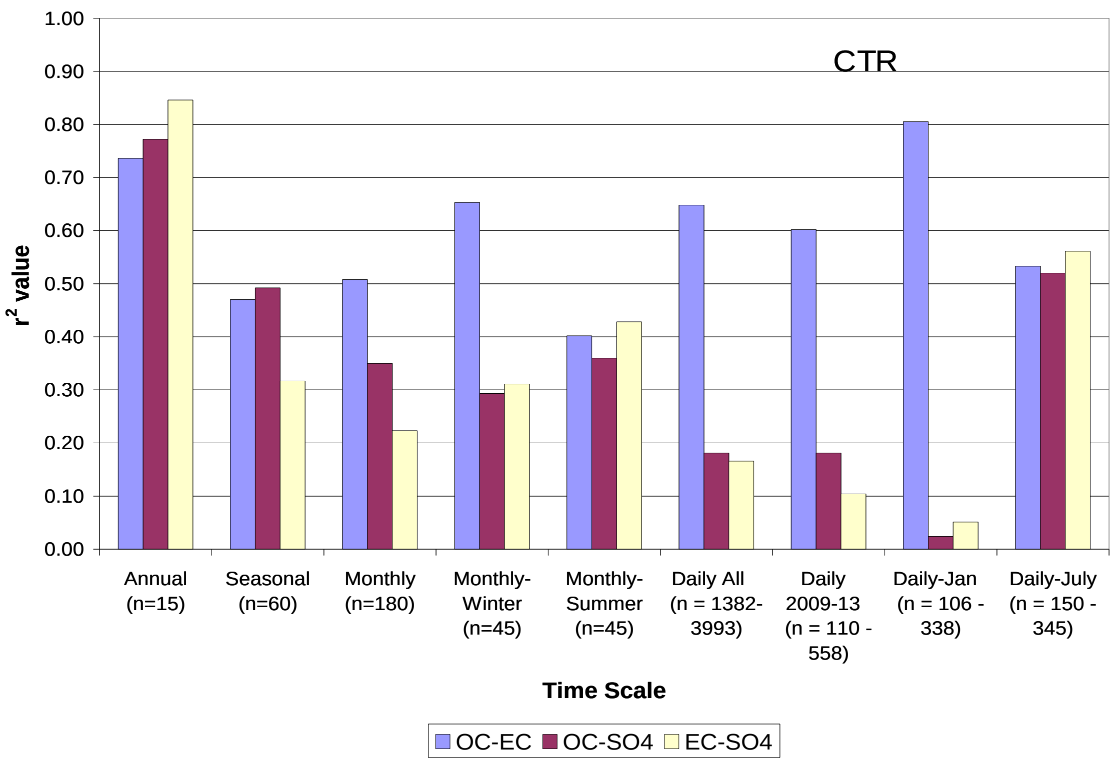


Figure S5. Correlations (r^2) among mean OC, EC, and SO_4 based on annual, seasonal, and monthly averaging periods and on daily measurements. Summer is defined as June through August, winter is December through February, autumn is September through November, and spring is March through May.

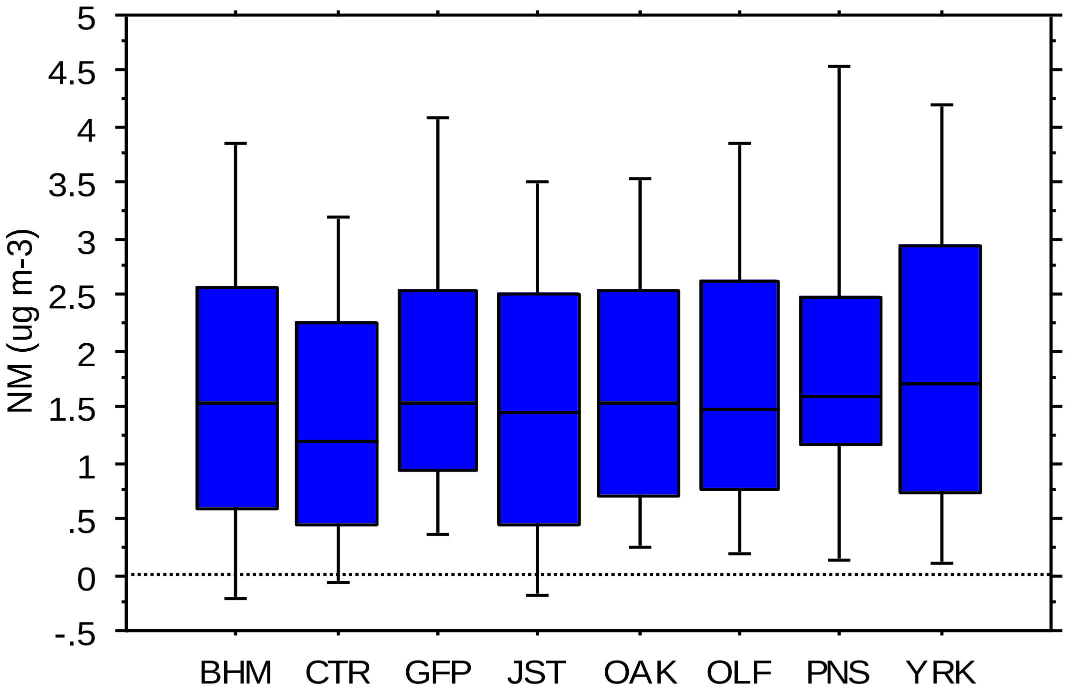


Figure S6. Distributions of non-measured (NM) $PM_{2.5}$ computed from 2009 – 2013 daily data as described in the text. Distributions indicate the 10^{th}, 25^{th}, 50^{th}, 75^{th}, and 90^{th} percentiles. No measurements were made at PNS after 2009, at OAK after 2010, and at GFP after 2012 (GFP EC and OC data were not available after 2010).

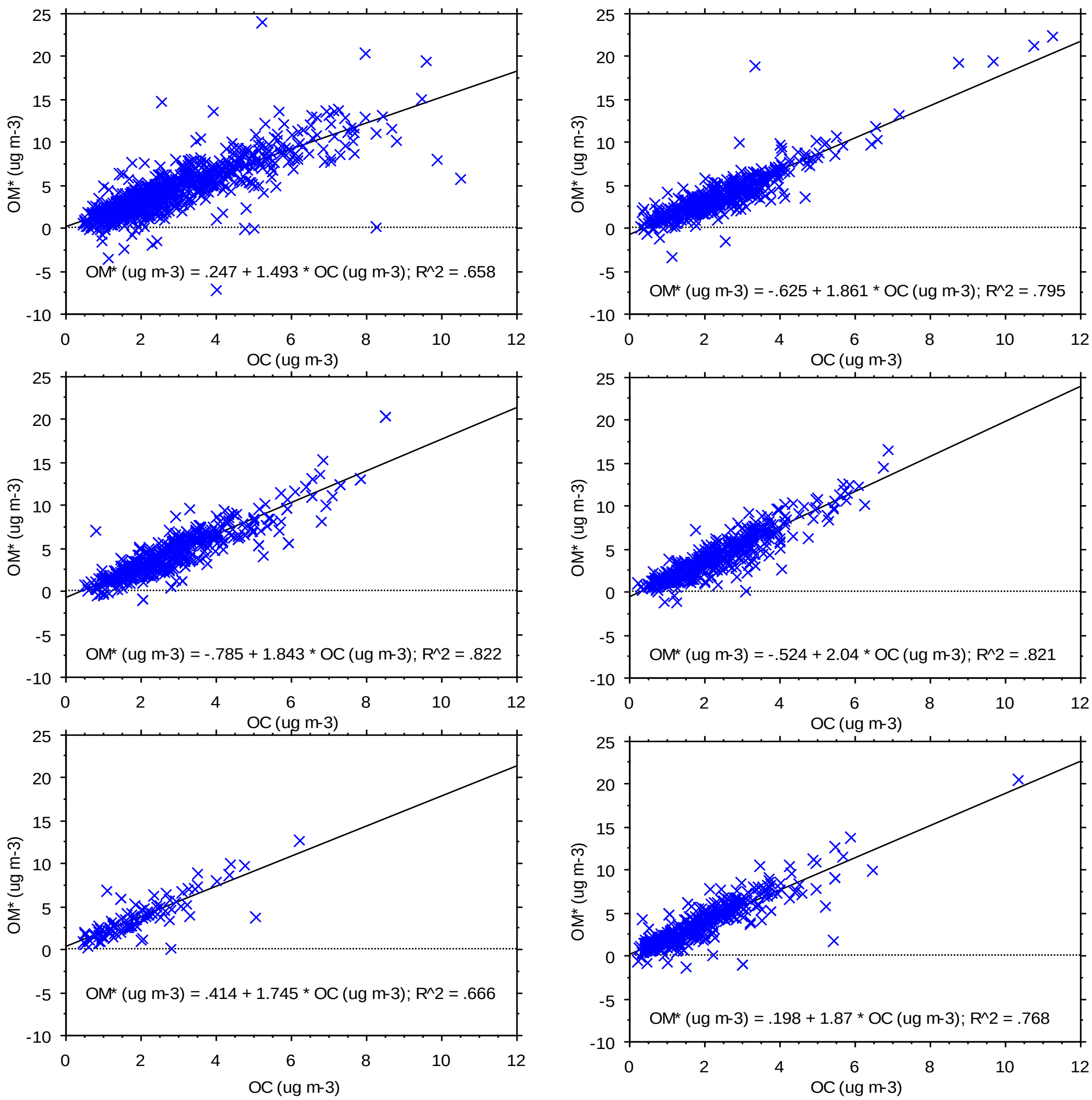


Figure S7. OM* vs. OC at SEARCH sites, 2009 – 2013. OM* is the sum of measured OC and the computed difference of $PM_{2.5}$ mass minus the sum of measured species concentrations. OM* is an upper bound for OM (see text).

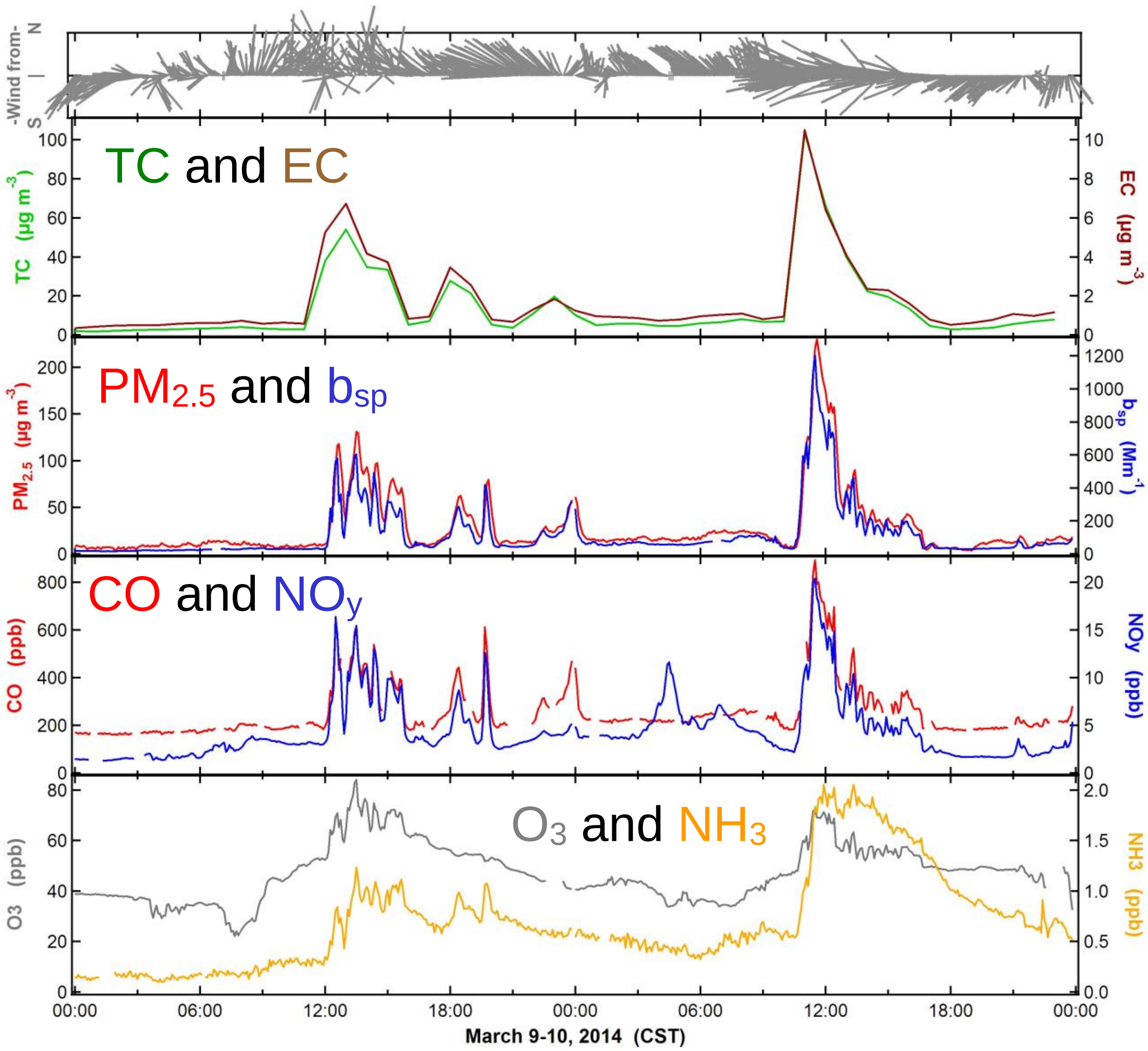


Figure S8. Observed biomass burning impacts at CTR on March 9 – 10, 2014. The burn was located ~10 km northwest of CTR on a ~1000 acre parcel. EC and TC data are hourly; all other measurements are 5-minute resolution. The surface wind directions potentially trace the plume but other factors (e.g., nocturnal inversion, fire intensity) also affect observed concentrations. NW winds occurred briefly on March 9 with weaker NW winds later in the day. The winds on March 10 show a persistent NW-SW direction followed by a shift to SW. The TC levels in the plume are very high relative to baseline: ~60 – 100 µg m^{-3} vs < ~ 5 µg m^{-3}. OC/EC ratios for the peak periods are about 9:1 to 11:1 (similar to the fire emission ratio in Table S2). The midday O_3 concentration peaks on the two days parallel the NO_y concentrations, but it is unclear if this pattern is directly related to the smoke plume or to more general effects from multiple mission sources in the region. The association of the NH_3 peak with the smoke plume is not explained, but could relate to fire chemistry.

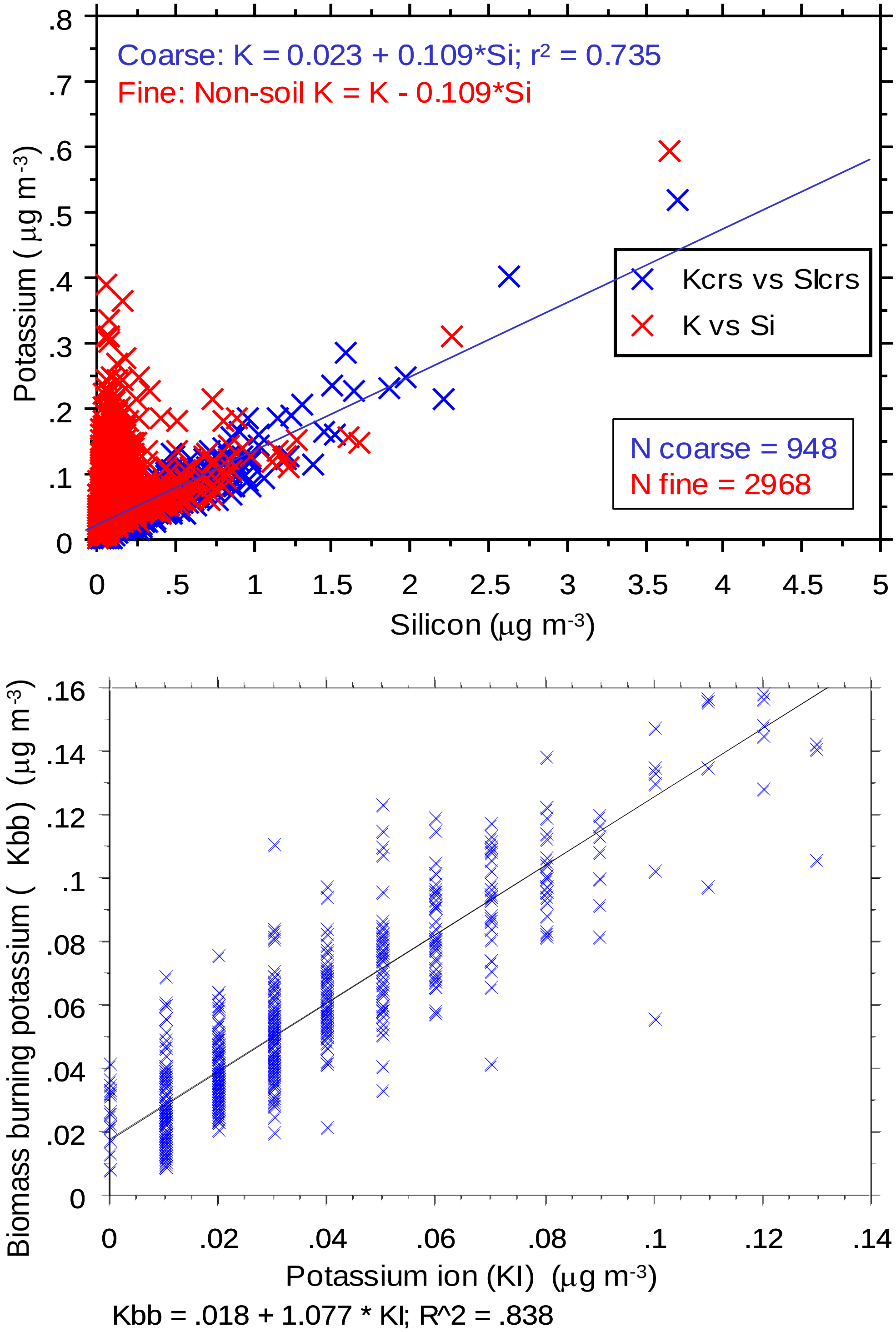


Figure S9. PM_{coarse} and $PM_{2.5}$ K vs. Si at CTR (top) and computed non-soil K (inferred as biomass burning K) vs. water soluble K (K ion) (bottom).

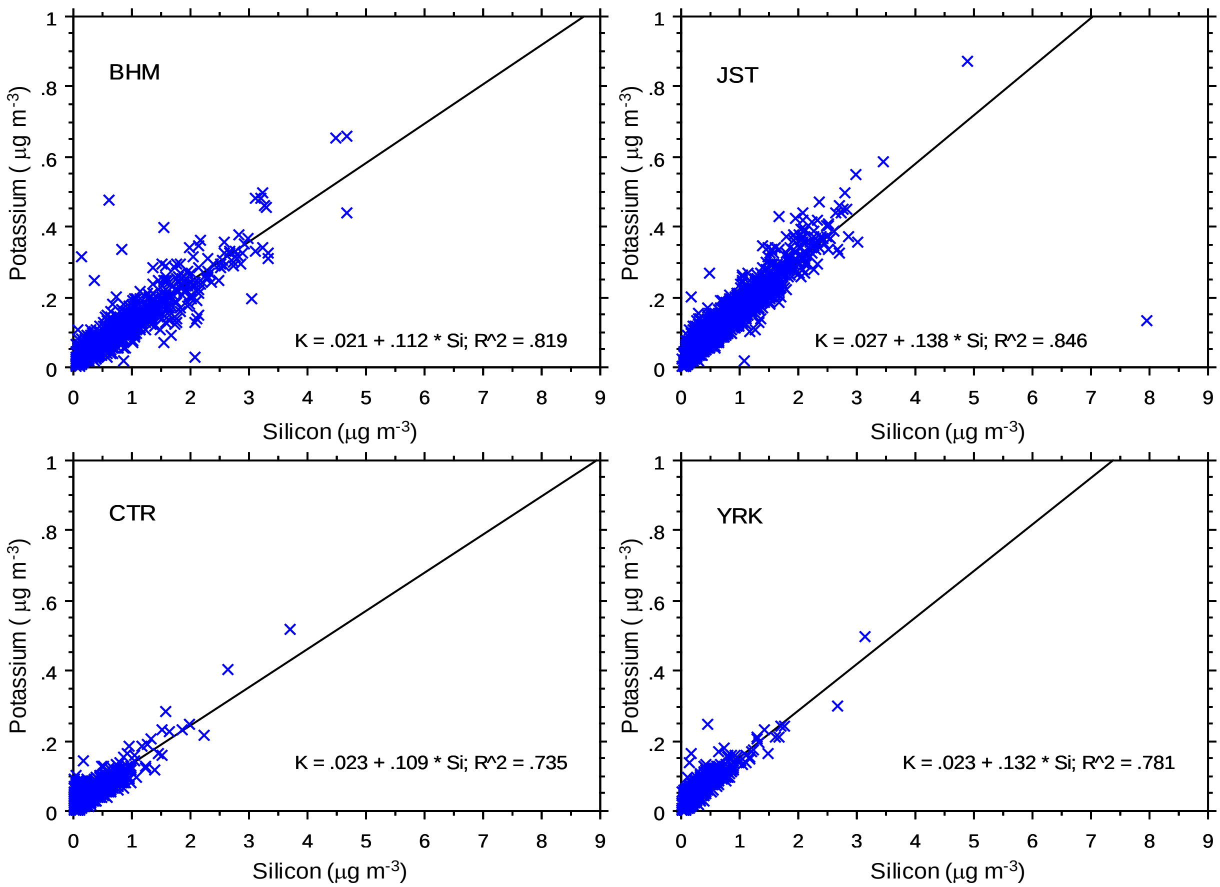


Figure S10. $PM_{10-2.5}$ K vs. Si.

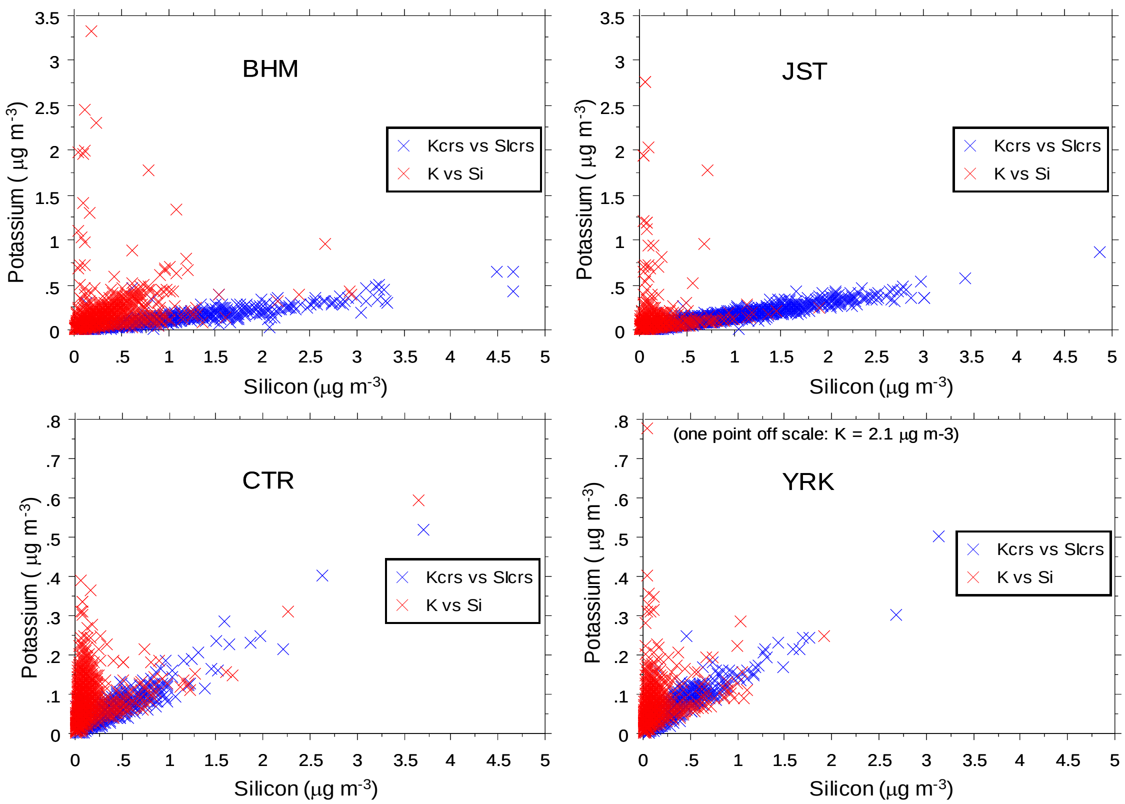


Figure S11. PM_{crs} ($PM_{10-2.5}$) K vs. Si and $PM_{2.5}$ K vs. Si. Units are µg m^{-3}.

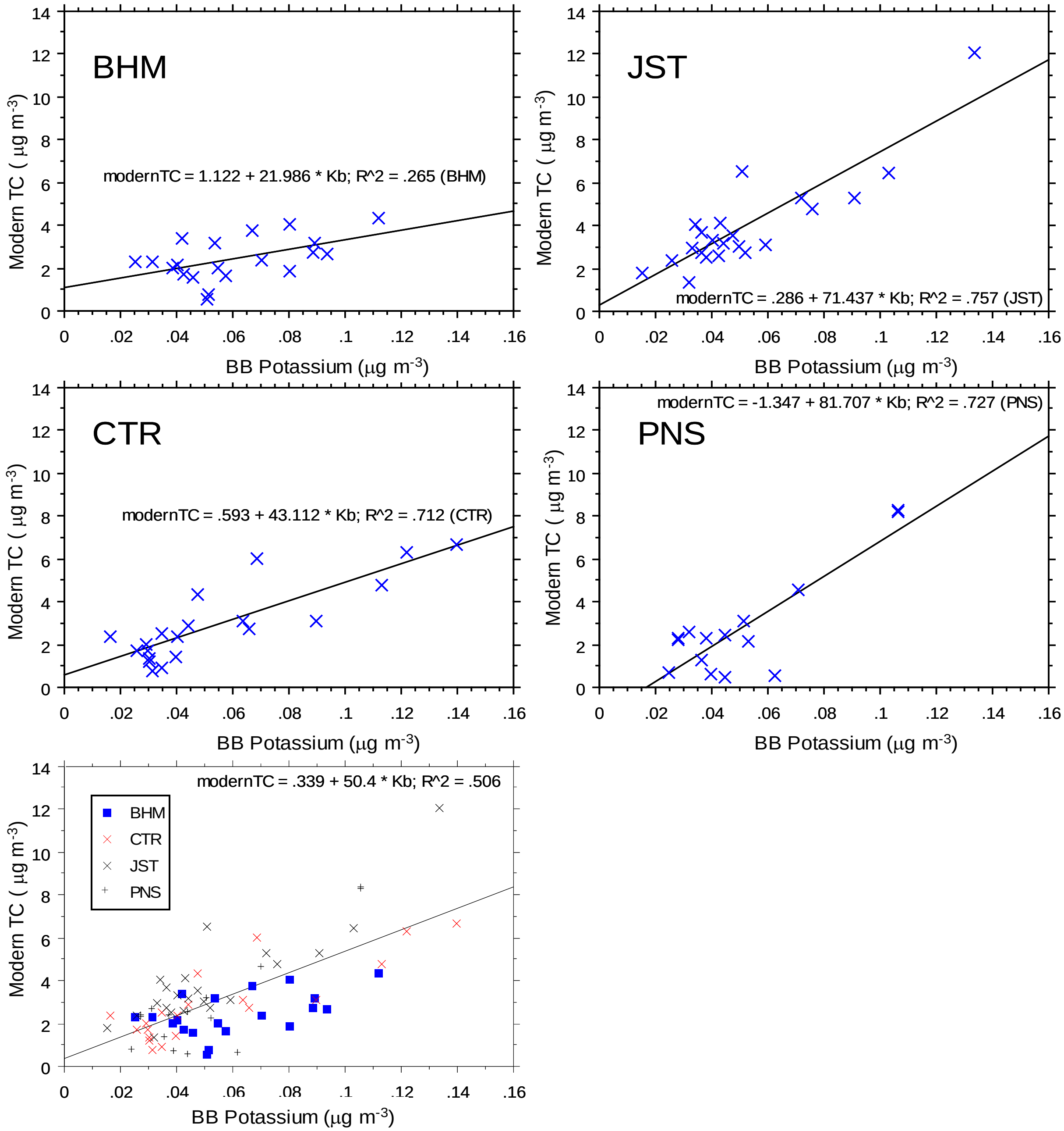


Figure S12. Modern total carbon (TC) (Blanchard et al., 2011) vs. biomass burning (BB) potassium (Kbb) for daily-average samples collected from the months of October through May (2004 and 2005).

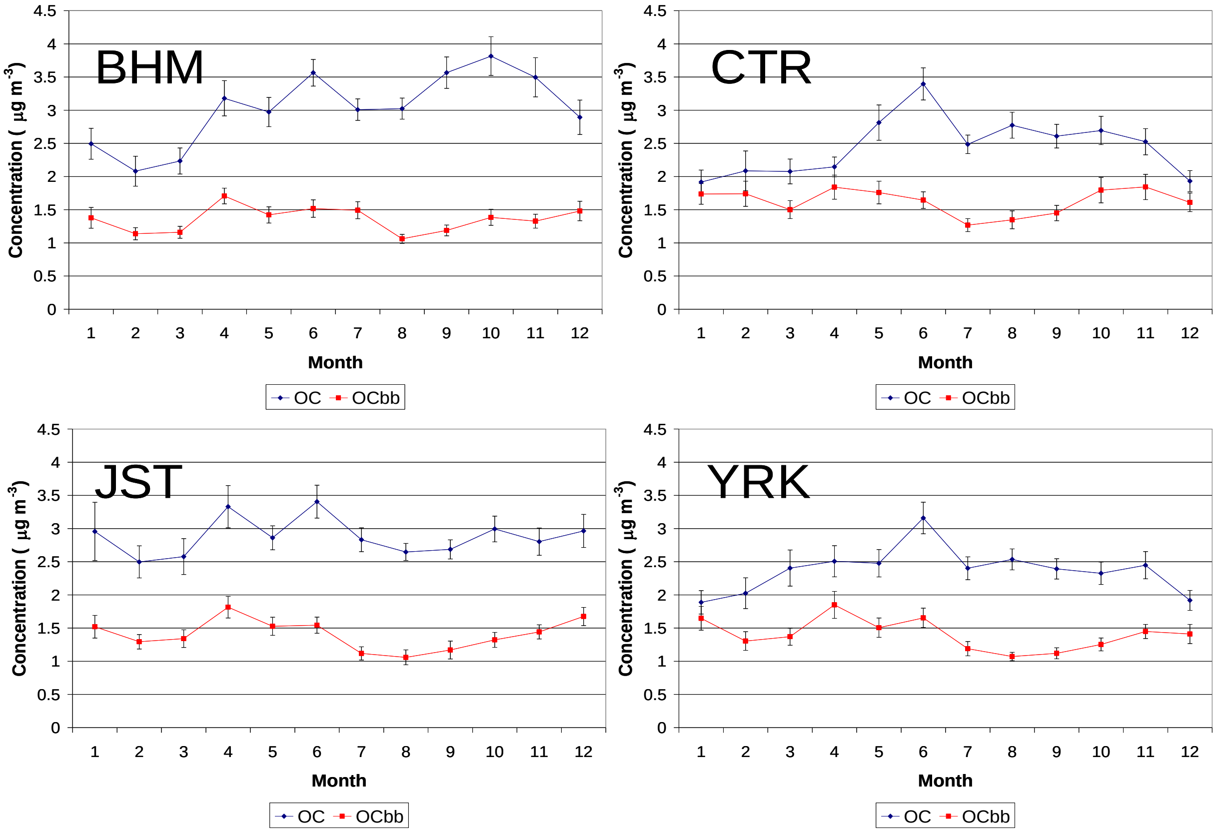


Figure S13. Monthly-average OC and OCbb at inland SEARCH sites, 1999-2013. Uncertainties are one standard error of the means.

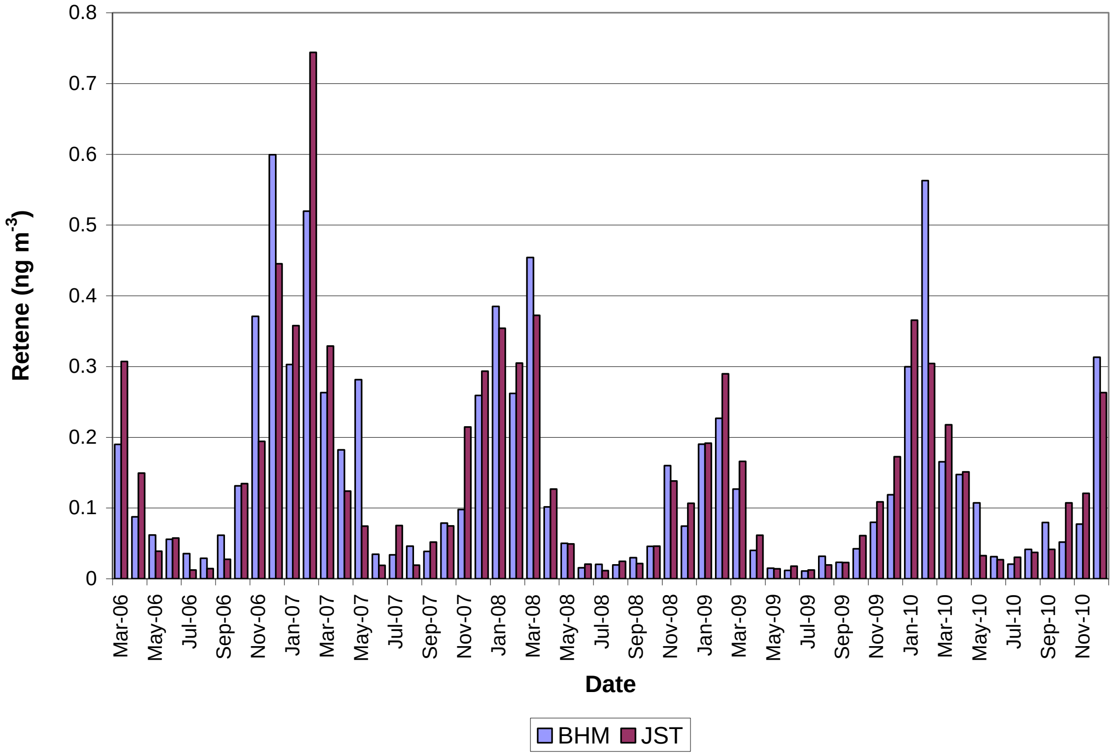


Figure S14. Monthly-average $PM_{2.5}$ retene concentrations at urban sites BHM and JST (Blanchard et al., 2014).

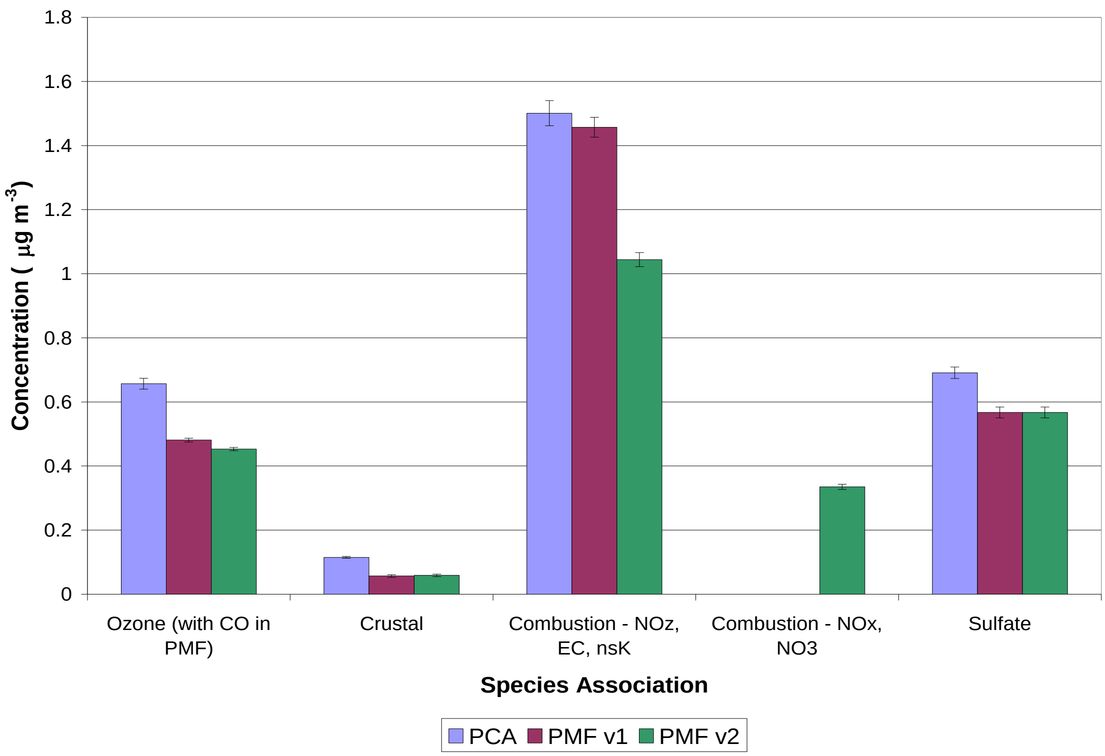


Figure S15. Comparison of CTR mean OC concentrations apportioned to PCA and PMF components. Uncertainties are one standard error of the means.

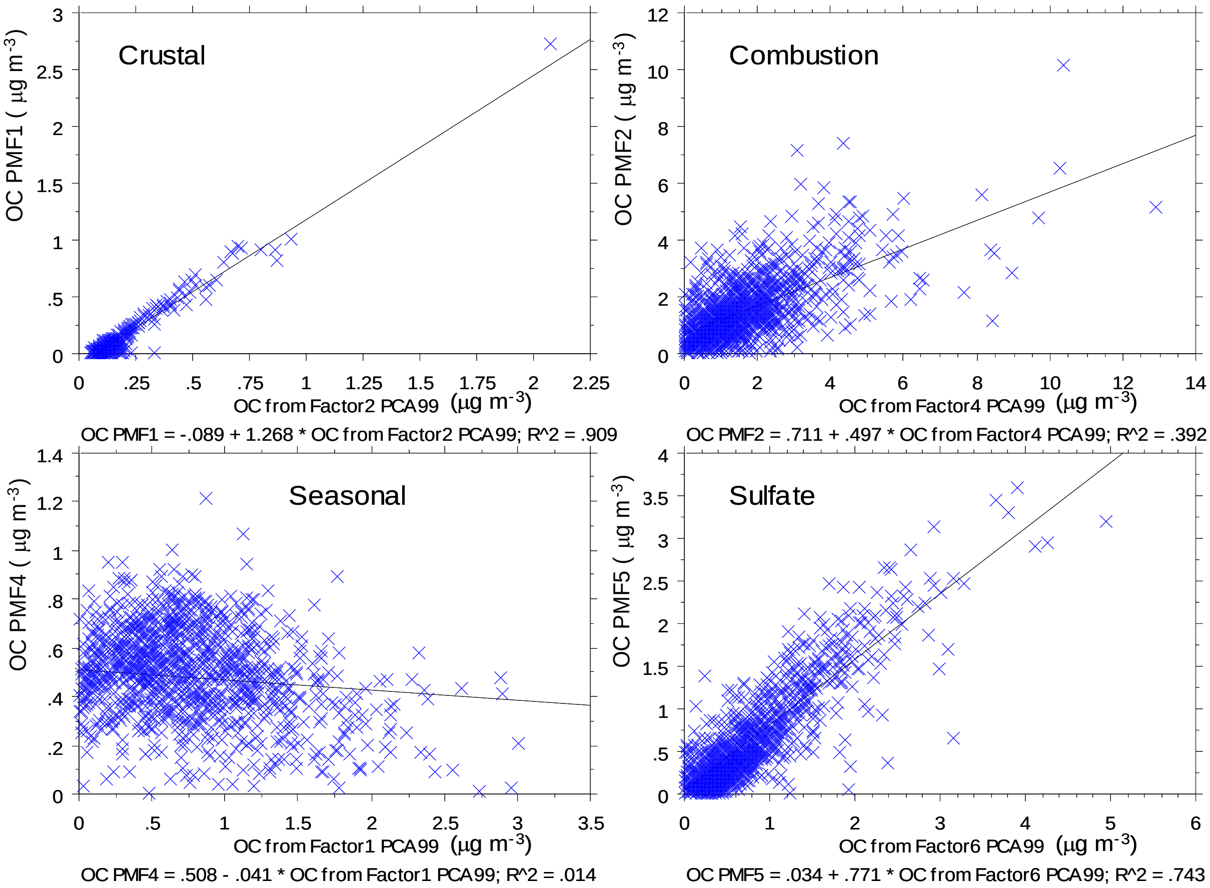
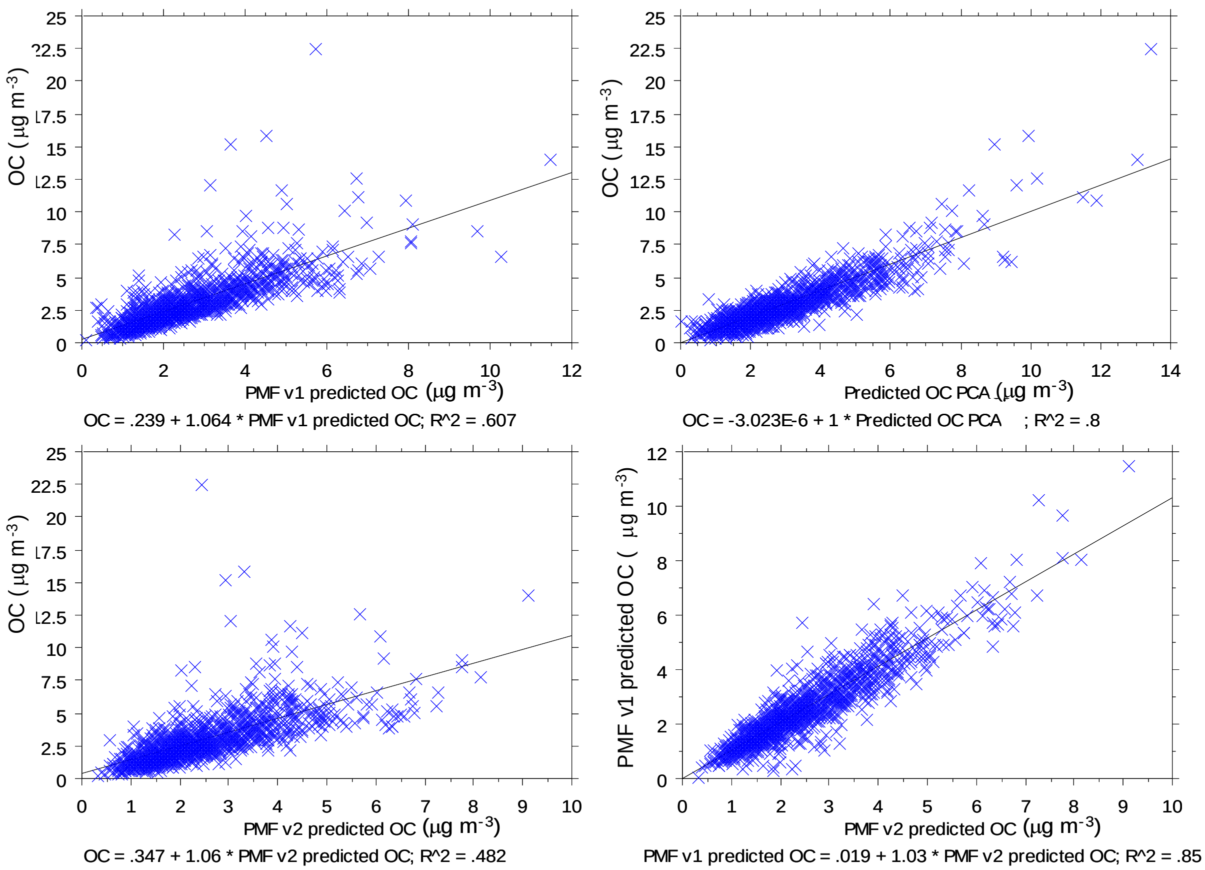


Figure S16. Comparison of CTR OC apportioned to PMF and PCA factors. PMF and PCA factor numbers are arbitrary and are matched by species associations. PCA99 refers to PCA2 beginning in 1999.



Figures S17. 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).

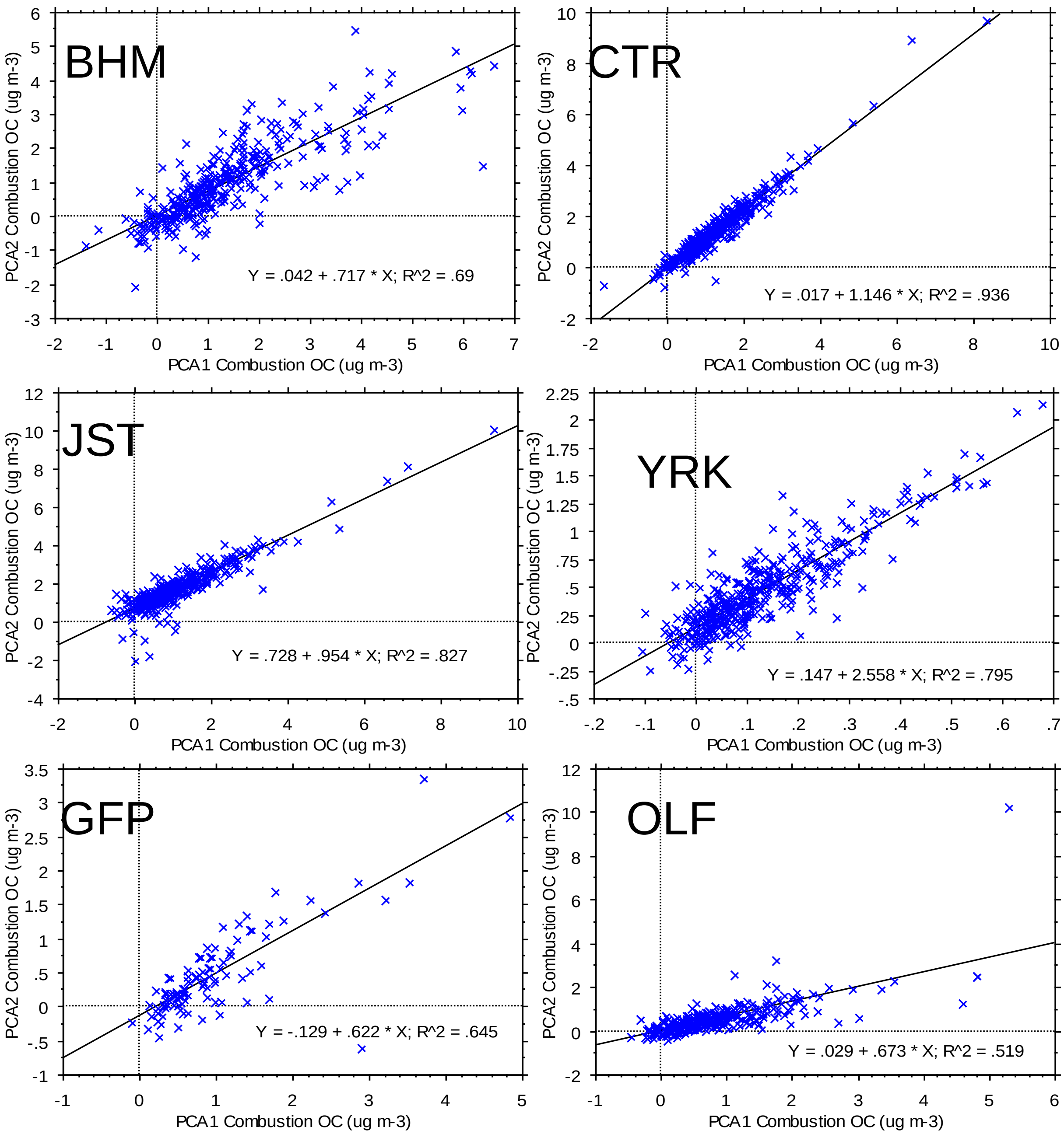


Figure S18. Comparison of PCA1 and PCA2 combustion-derived OC.

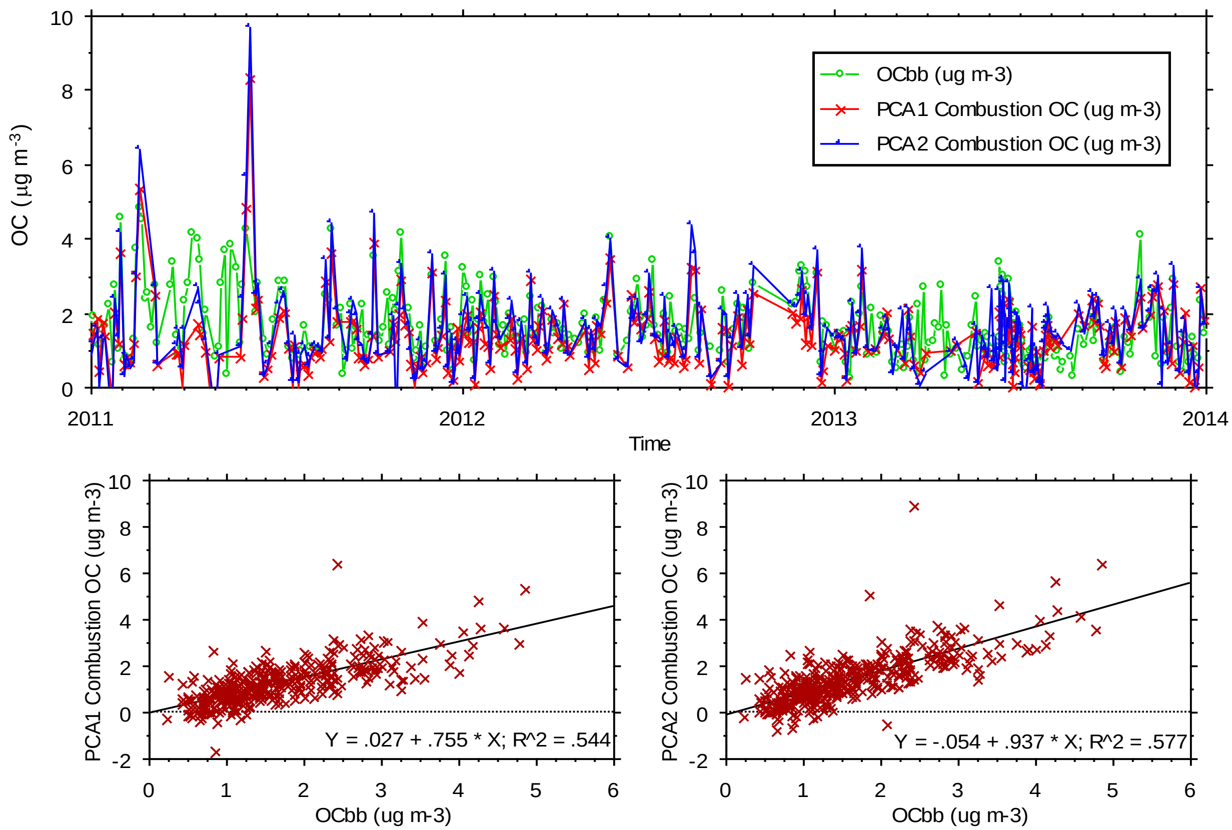


Figure S19a. Comparison of OCbb, PCA1 combustion OC, and PCA2 combustion OC at CTR. The scatter plots show all data points in 2008 – 2013. OCbb correlates significantly ($p < 0.0001$) with both PCA1 and PCA2 combustion OC. Whereas OCbb is computed from Kbb, PCA1 and PCA2 combustion OC concentrations are determined by concentrations of CO and EC in addition to K ion (PCA1) or Kbb (PCA2). On average, OCbb is ~10 – 20% higher than PCA1 or PCA2 combustion OC.

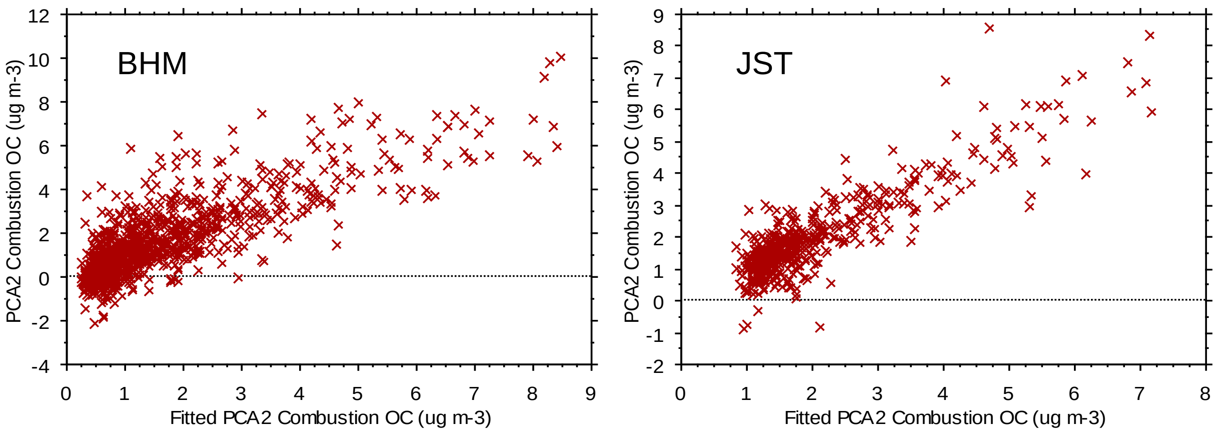


Figure S19b. PCA2 combustion OC vs predicted combustion OC as predicted by multiple regression on species not included in the PCA. (Left) BHM, predictor species 1-hour daily maximum NO and condensed-phase sums of n-alkanes, iso- and anteiso-alkanes, hopanes, and polycyclic aromatic hydrocarbons (PAHs) ($R^2 = 0.67$, $p < 0.0001$ for all species). (Right) JST, predictor species 1-hour daily maximum NO, condensed-phase sum of n-alkanes, gas-phase sum of aromatics ($R^2 = 0.78$, $p < 0.0001$ for all species). Gas-phase species data were not available for BHM. The correlations indicate that PCA2 combustion OC is associated with fresh emissions (NO) and with non-oxidized organic compounds.

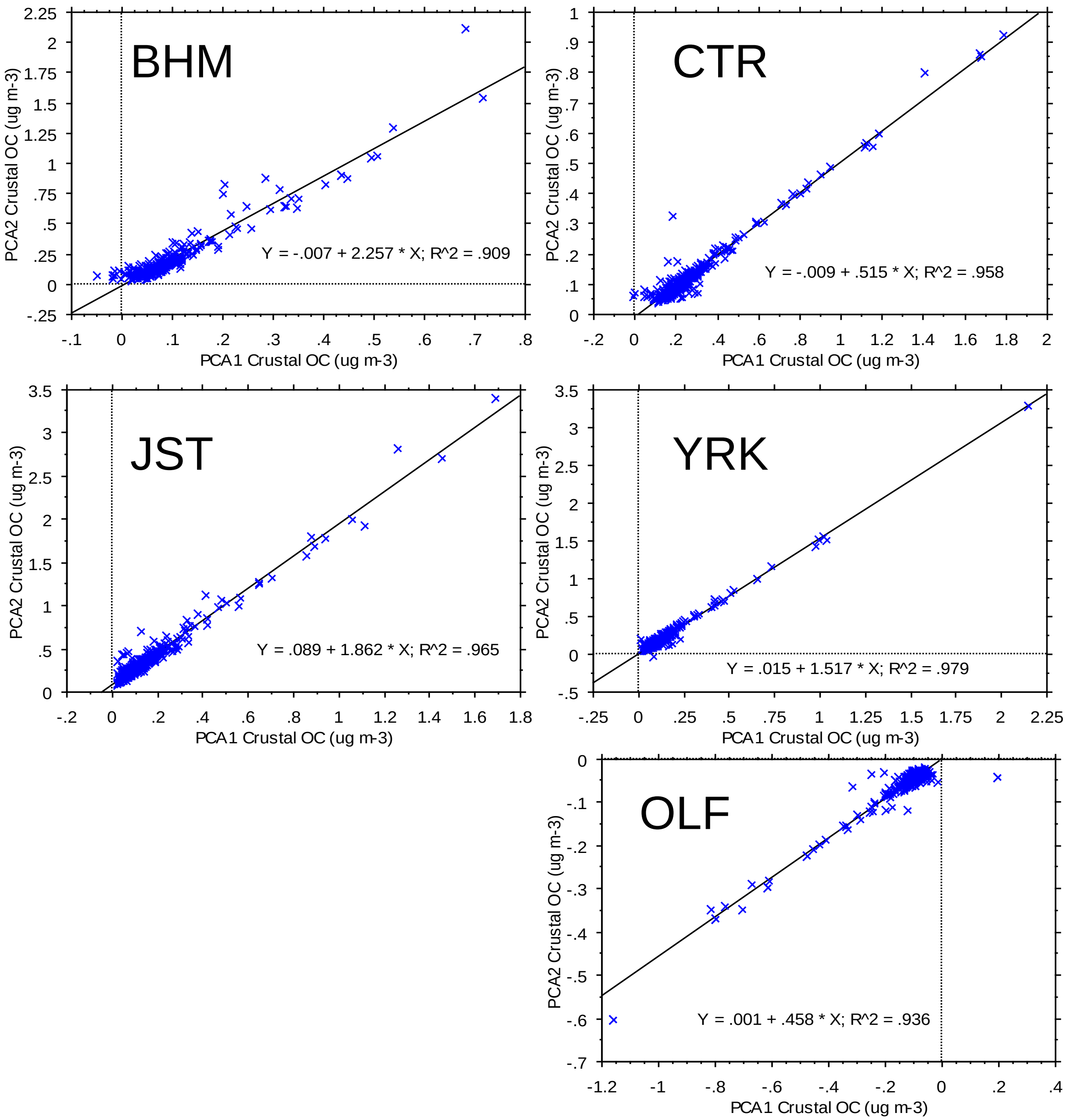


Figure S20. Comparison of PCA1 and PCA2 crustal OC.

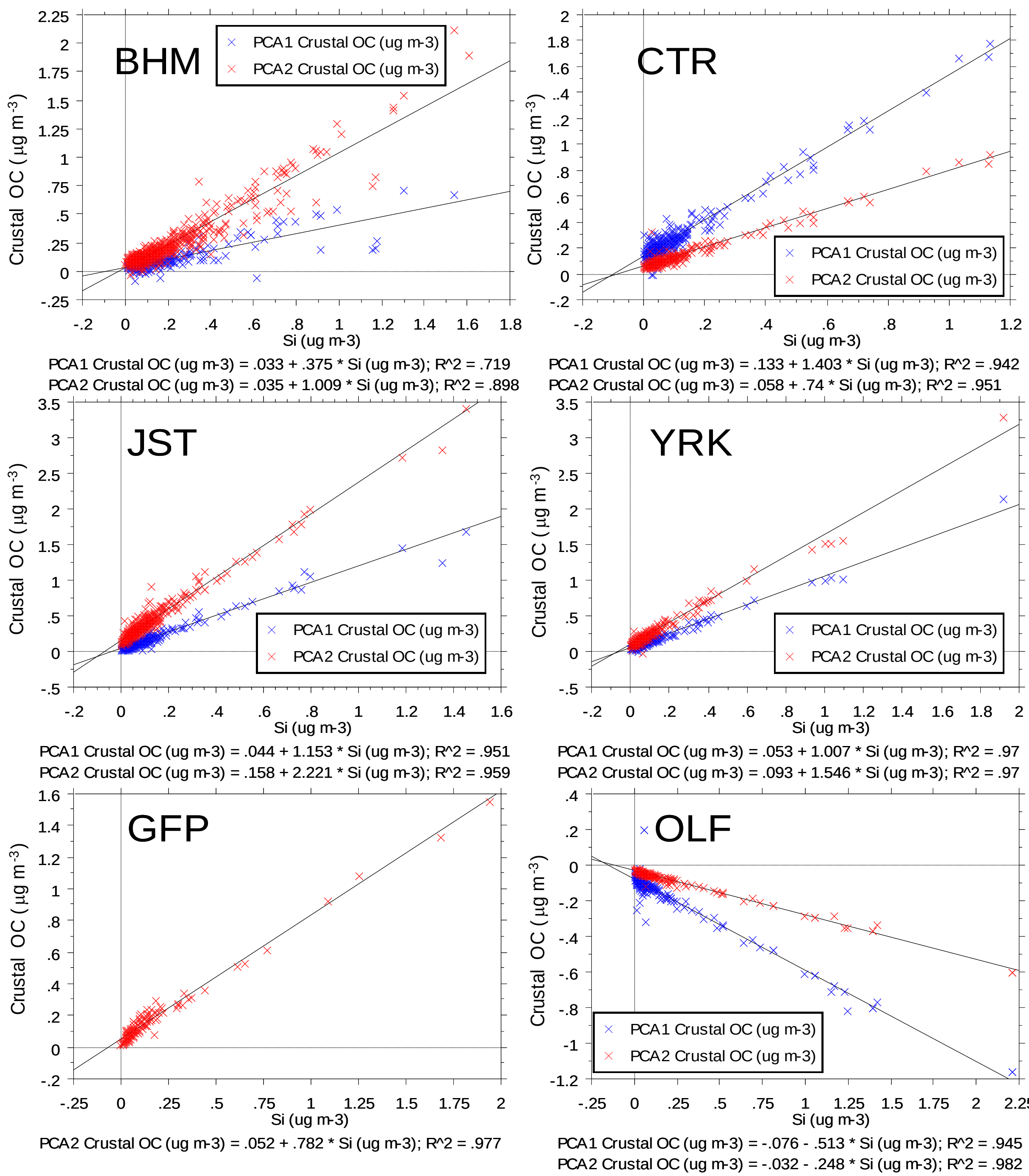


Figure S21. PCA1 and PCA2 crustal OC vs. silicon.

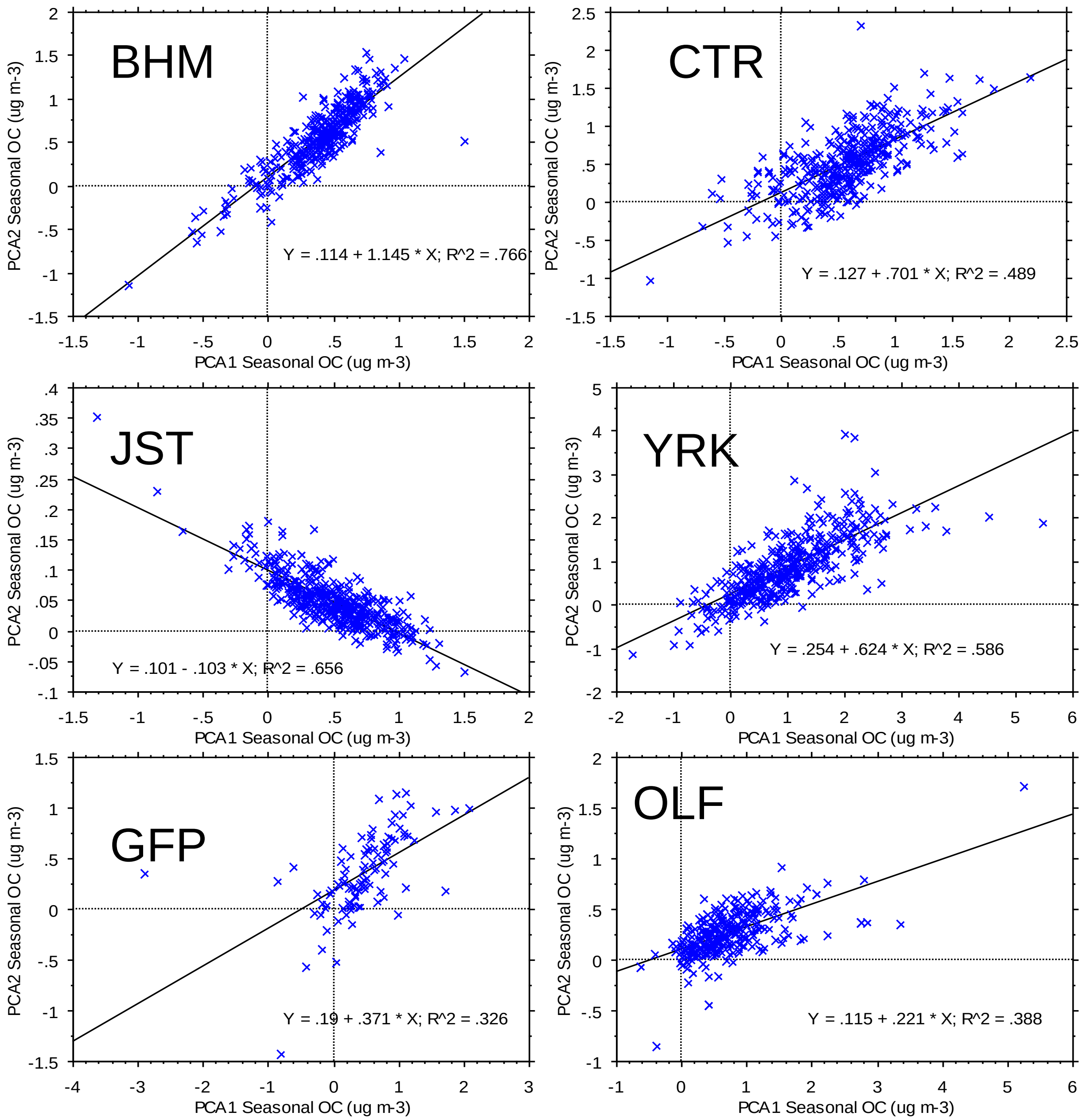


Figure S22. Comparison of PCA1 and PCA2 seasonal-component OC.

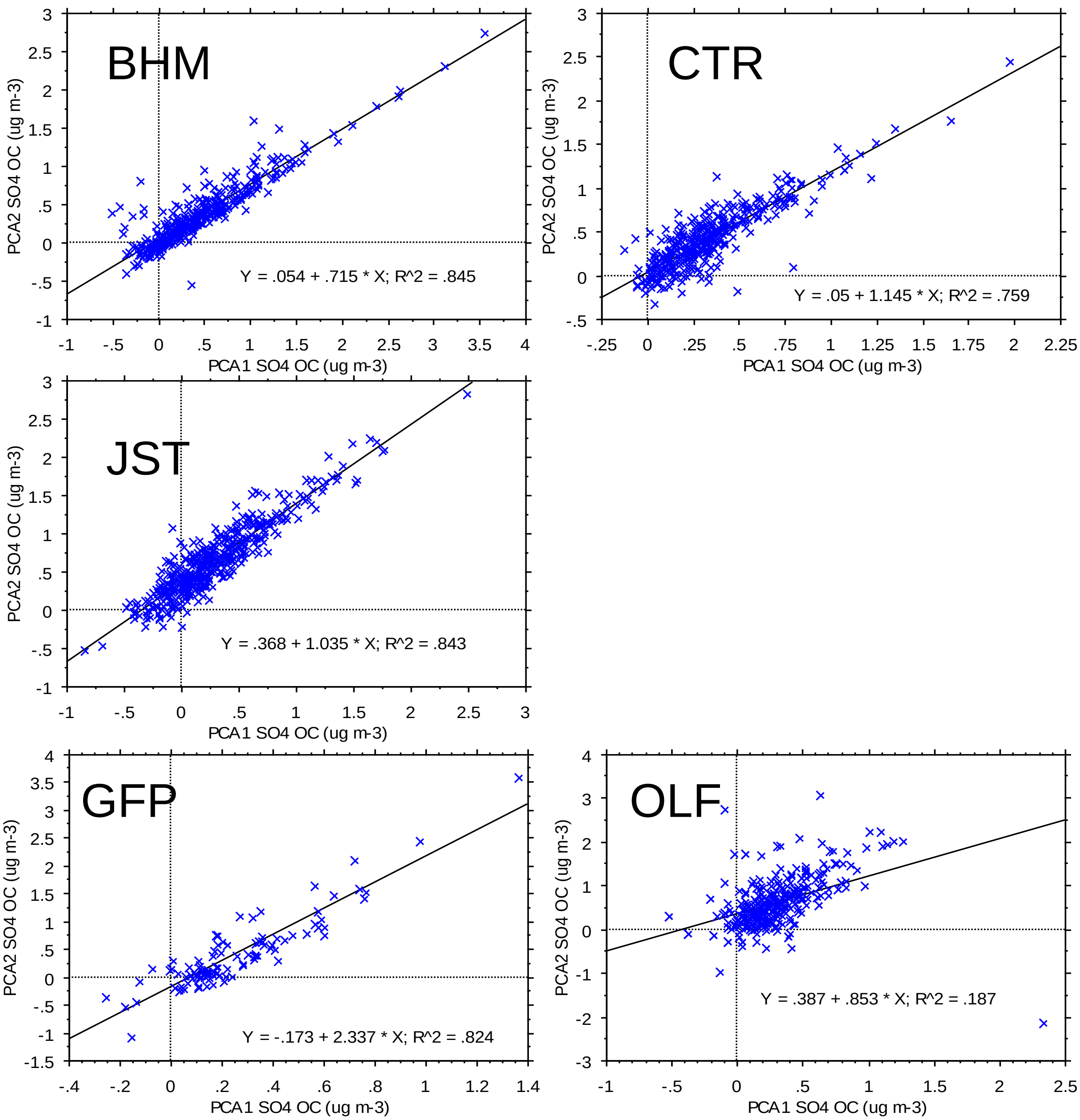


Figure S23. Comparison of PCA1 and PCA2 SO_4 OC.

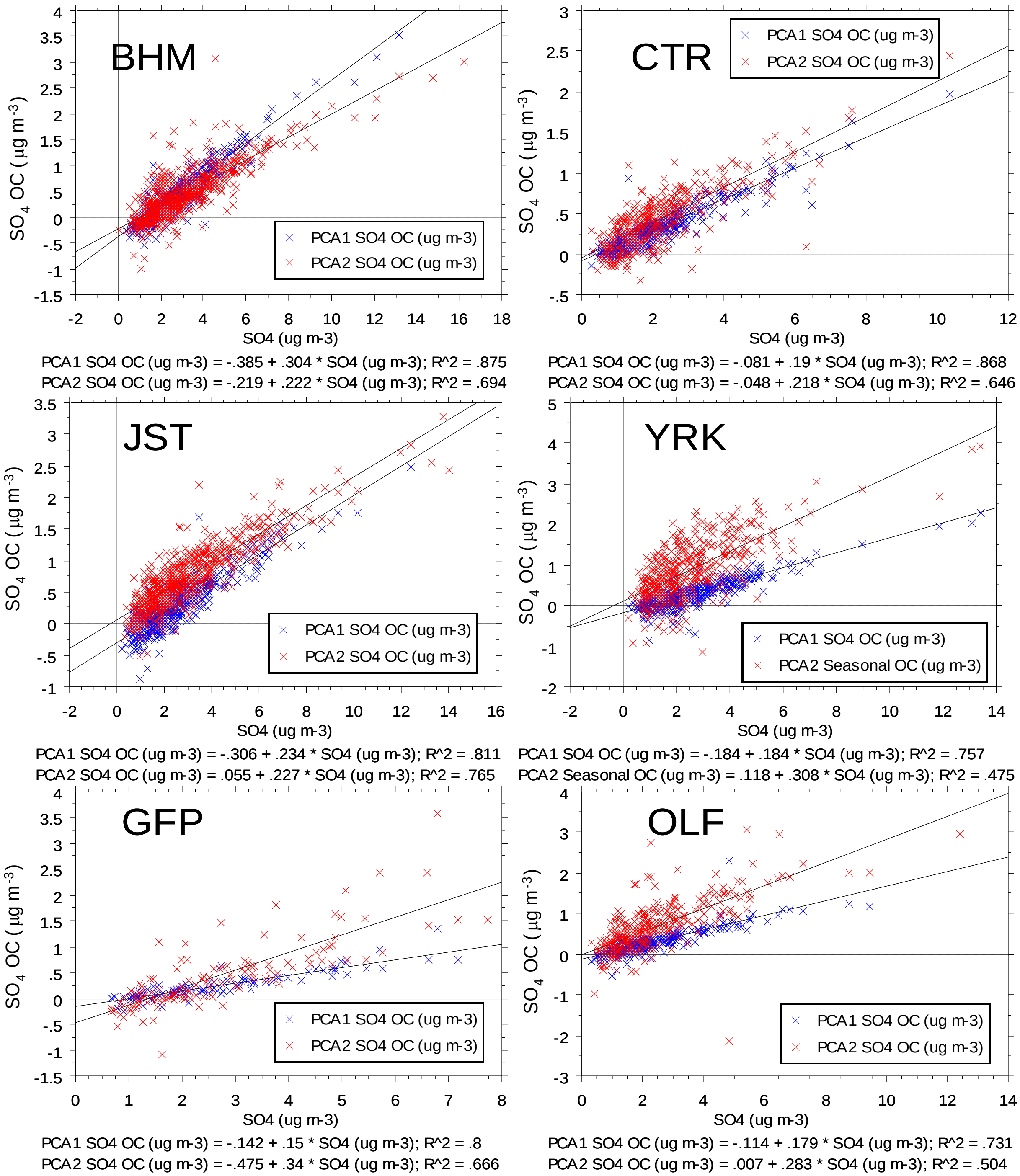


Figure S24. PCA1 and PCA2 SO_4 OC vs. SO_4. PCA2 did not separate seasonal and SO_4 components at YRK.

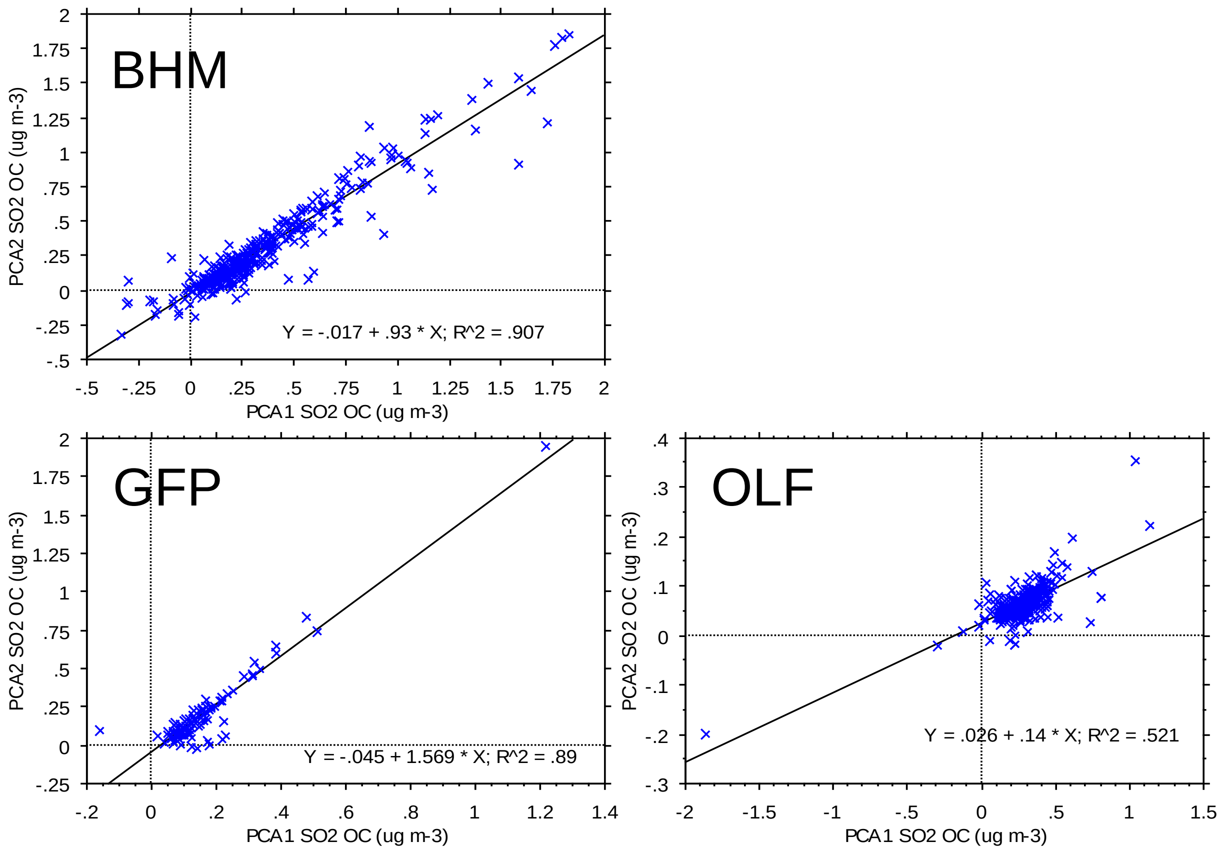


Figure S25. Comparison of PCA1 and PCA2 SO_2 OC.

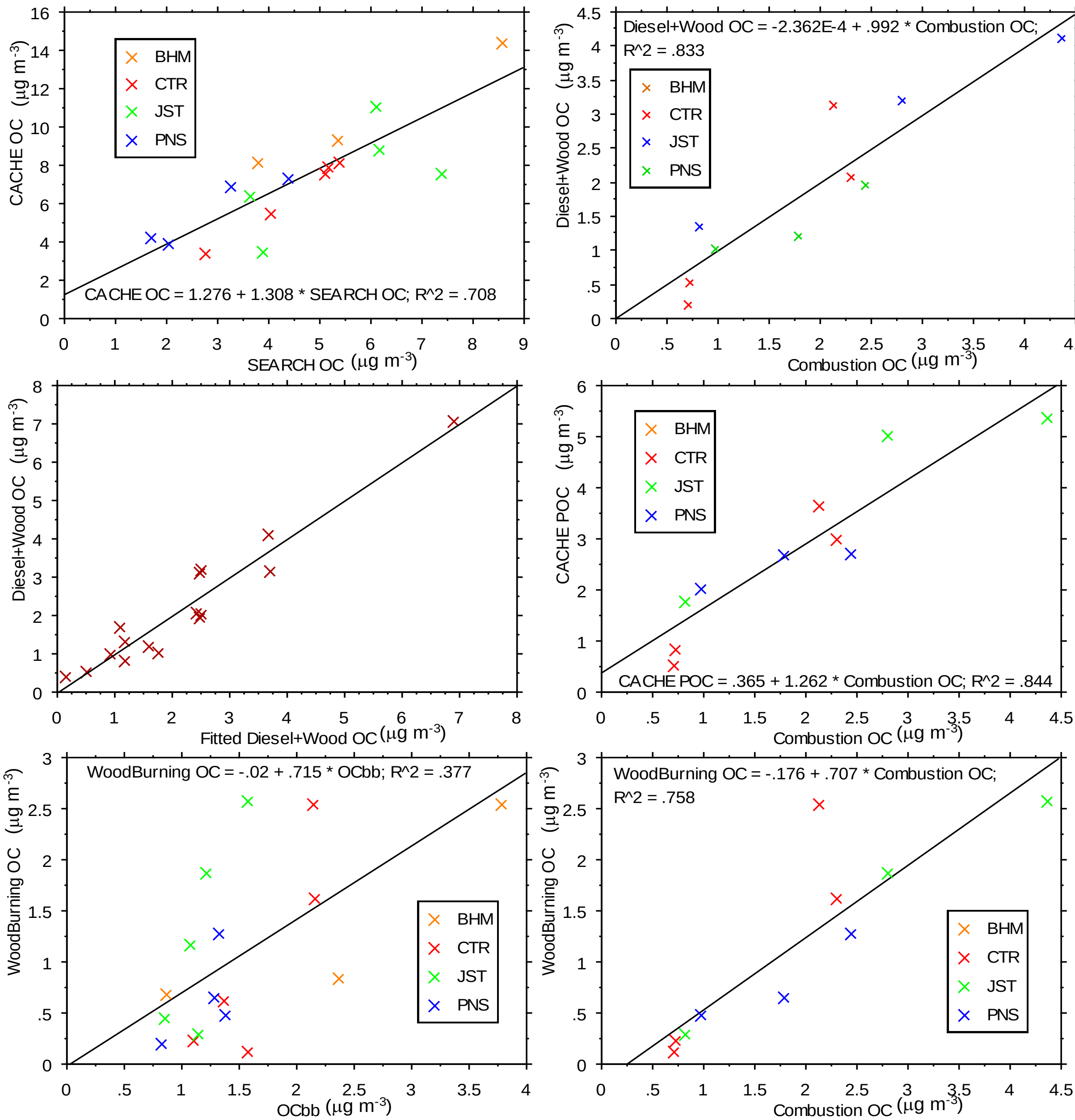


Figure S26. CACHE sampler OC source apportionments (Kleindienst et al., 2010) vs. SEARCH PCM sampler OC measurements and computed source contributions. SEARCH combustion OC was determined using PCA2 and is OC that is associated with CO, EC, Kbb, and NO_x (Section 3.5). SEARCH OCbb was determined from Kbb (Section 3.4). CACHE POC is the sum of seven source categories described by Kleindienst et al. (2010).

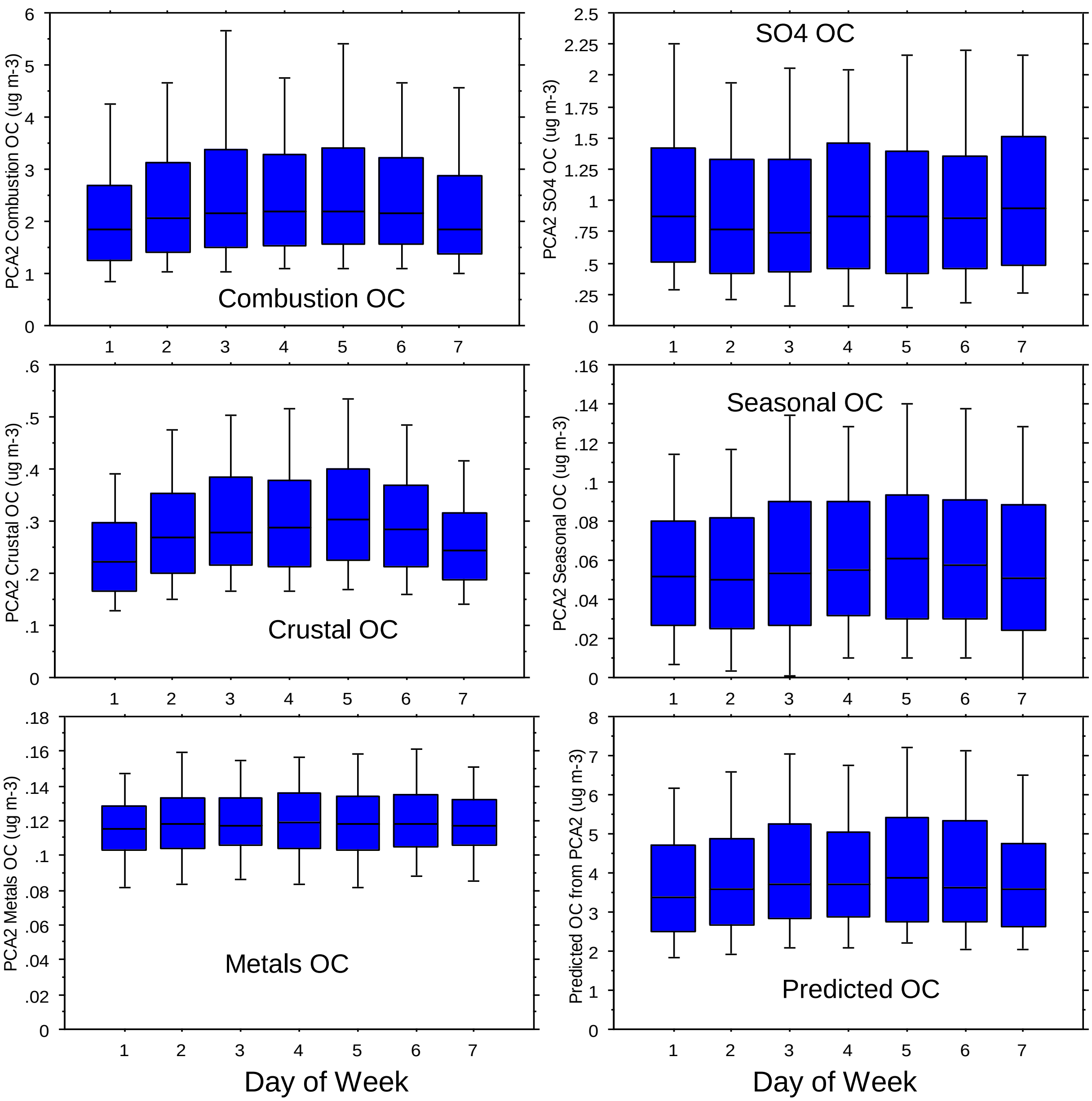


Figure S27. OC associated with PCA2 factors at JST vs. day of week, 1999 – 2013. Day of week begins with Sunday (1). Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Units are μg m^{-3}.

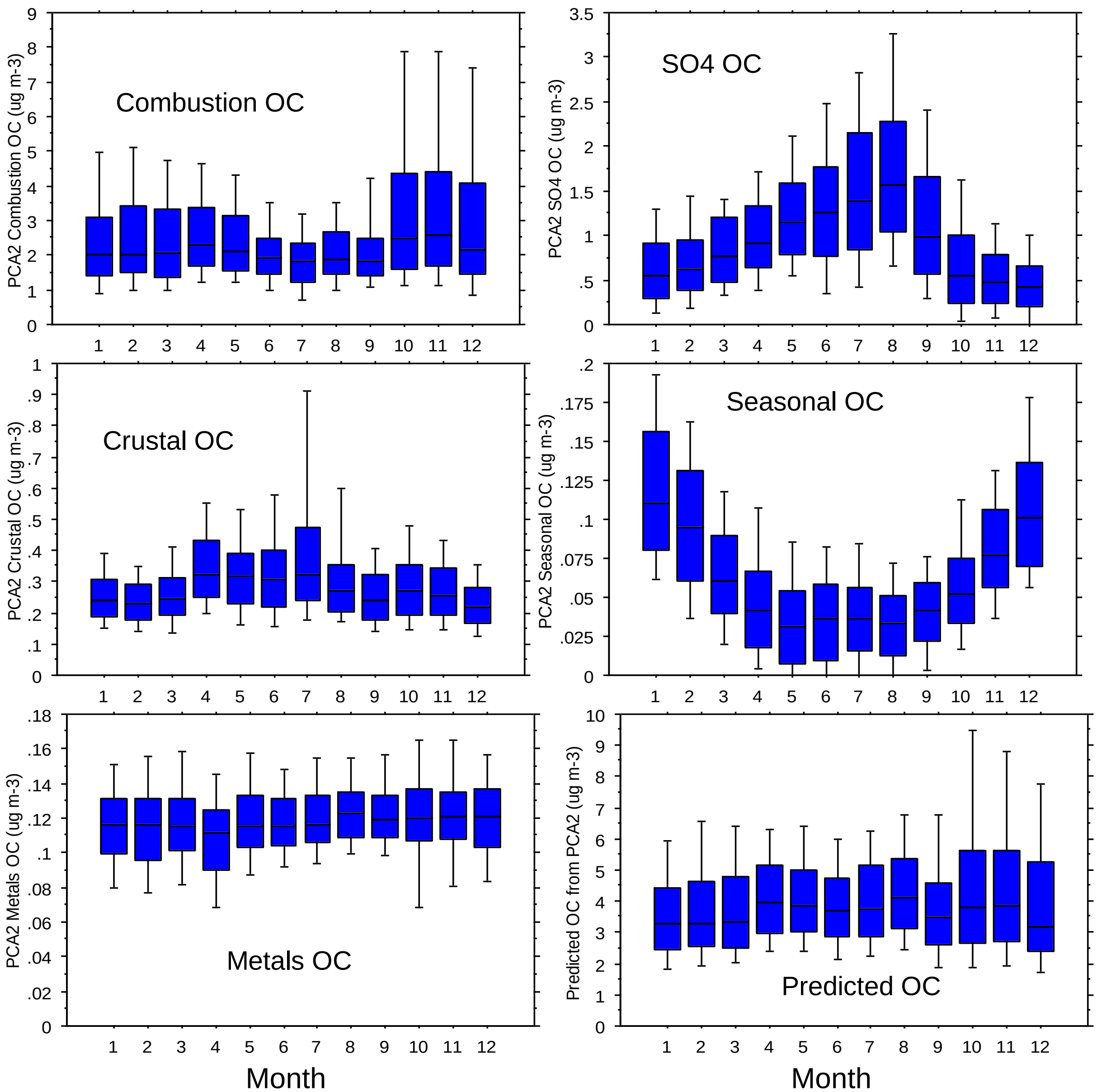


Figure S28. OC associated with PCA2 factors at JST vs. month, 1999 – 2013. Distributions indicate the 10^{th}, 25^{th}, 50^{th}, 75^{th}, and 90^{th} percentiles.

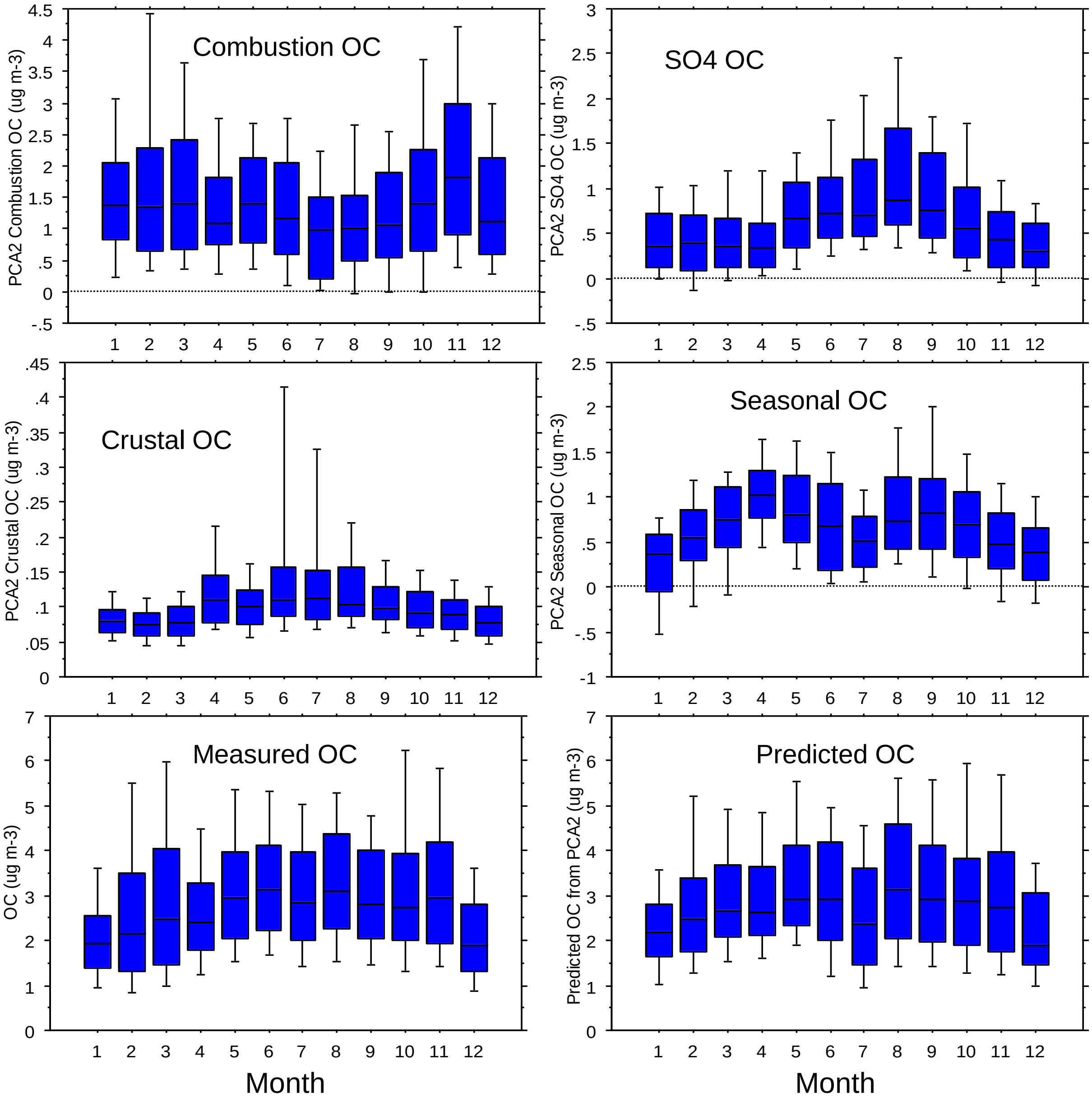


Figure S29. OC associated with PCA2 factors at CTR vs. month, 1999 – 2013. Distributions indicate the 10^{th}, 25^{th}, 50^{th}, 75^{th}, and 90^{th} percentiles.

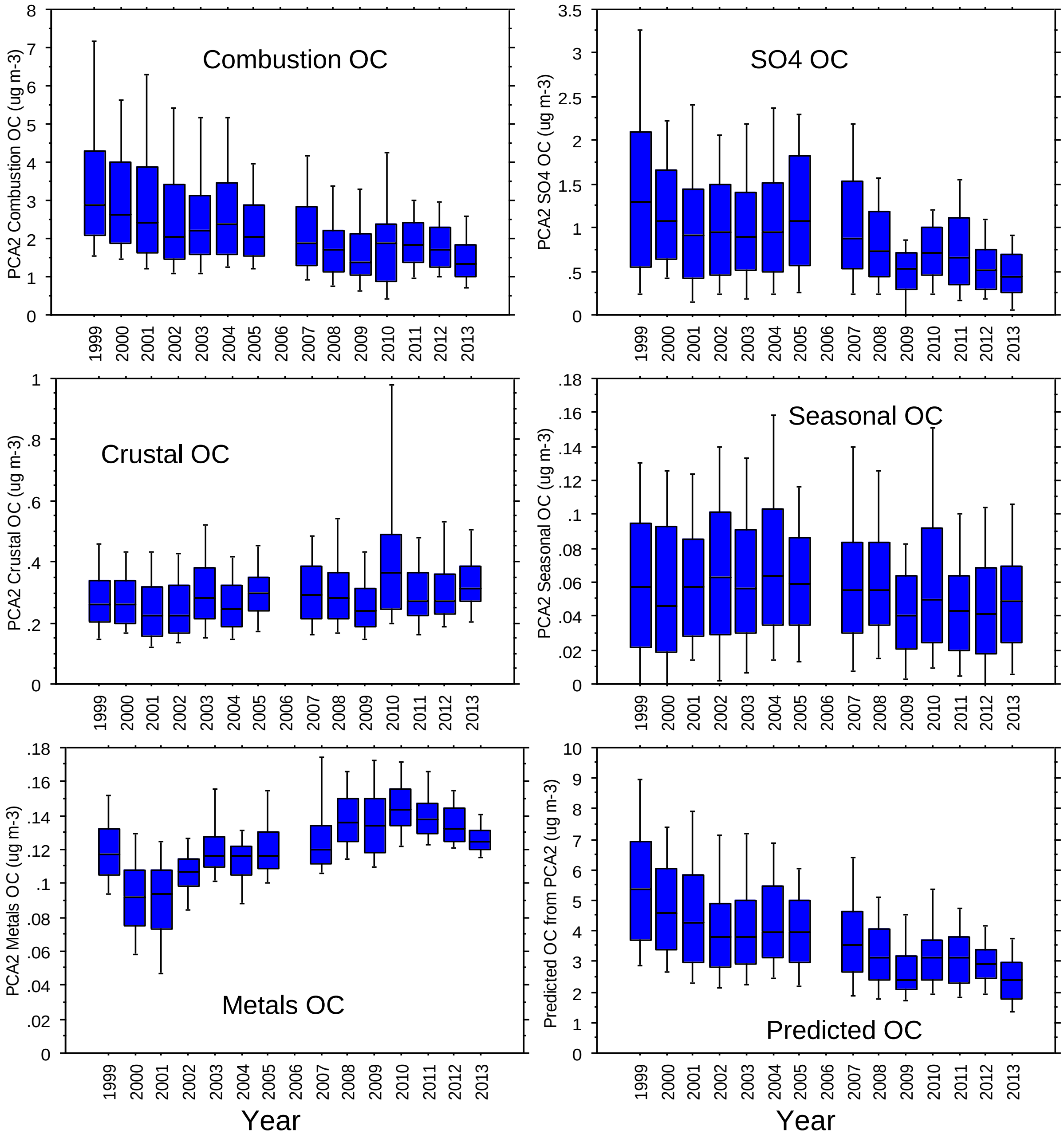


Figure S30. OC associated with PCA2 factors at JST vs. year, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. No predictions were made for 2006 due to missing CO and NO measurements.

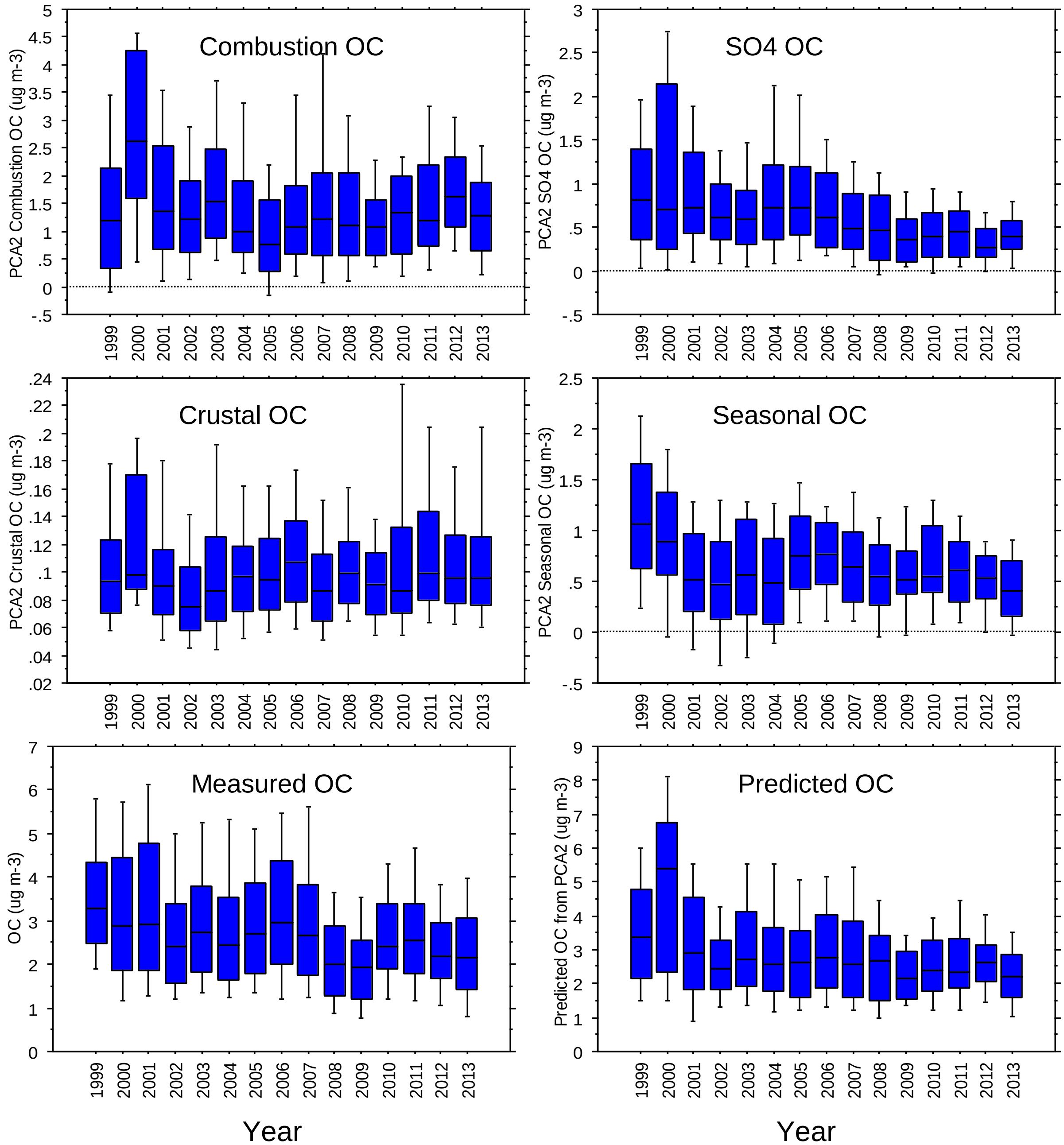


Figure S31. OC associated with PCA2 factors at CTR vs. year, 1999 – 2013. Distributions indicate the 10th, 25th, 50th, 75th, and 90th percentiles. Years with fewer sampling days are 2000 (n=11), 2009 (n=31), and 2010 (n=38).

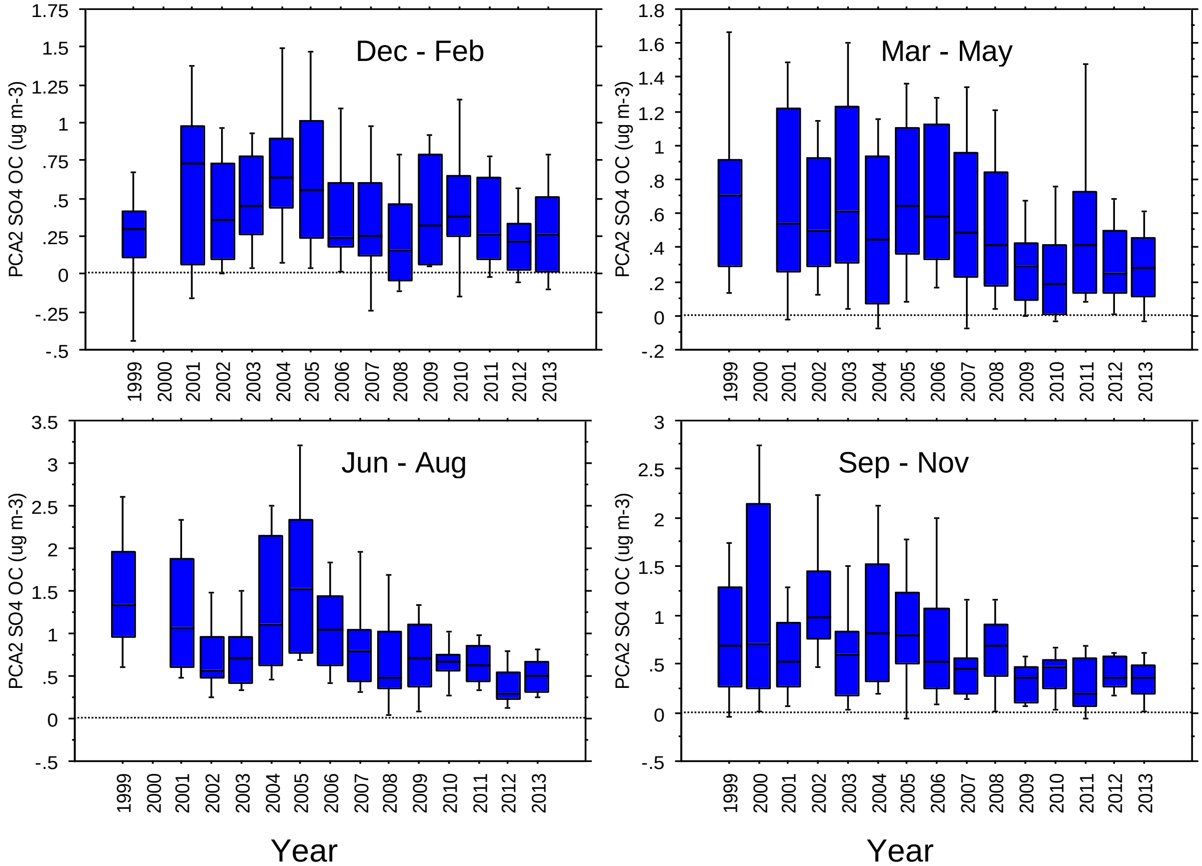


Figure S32. OC associated with PCA2 SO_4 factor at CTR vs. year, 1999 – 2013, shown by season. Distributions indicate the 10^{th}, 25^{th}, 50^{th}, 75^{th}, and 90^{th} percentiles. Years with fewer sampling days are 2000 (n=11), 2009 (n=31), and 2010 (n=38).

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