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Supplement of

Fine particulate matter source apportionment using a hybrid chemical transport and receptor model approach

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Note S1 Using a newer version of CMAQ

We are developing a hybrid method that aims to "correct" source-oriented-modeling-based (or "SM") $PM_{2.5}$ source apportionment results by using regularly available measurements (e.g., routine observations from monitoring networks). The hybrid method as expressed in our Eq. (12) can be applied to "correct" source apportionment results obtained from CMAQ with DDM as in this manuscript, CMAQ using other source apportionment approaches (including brute force methods), or other CTM-based methods. We are not developing a method that has to be attached to a specific CTM framework. Therefore the CTM-based results to be "corrected" can be from applications of a new CTM model with a recent emissions inventory or a "seasoned" but still widely used CTM with an extensively evaluated emissions inventory as in our case study. We used CMAQ because it is a widely used model, and v4.5 was chosen because it was the only version available that had DDM at the time the initial calculations were conducted. Since that time, a more recent version of CMAQ, v4.7.1, became available with DDM, and CMAQ v5.0.2 is expected to be released with DDM soon.

While CMAQ v4.6, v4.7, v4.7.1, v5.0 and v5.0.1 included theoretical improvements over v4.5 in many ways, the improvements in the ability to simulate $PM_{2.5}$ and its components haven't been substantial (Simon et al. 2012, Appel et. al. 2013), as suggested by the model performance evaluation of CMAQv5.0.1 in its application to the southeastern US (http://semap.ce.gatech.edu/node/1835). Additionally, of these newer CMAQ versions, DDM-3D is only available in v4.7.1. The implementation of DDM-3D and higher order DDM-3D for $PM_{2.5}$ to CMAQ 5.0.2 is still ongoing.

We have rerun the model using v4.7.1 and re-assessed the impact on the prescribed burn sensitivities (which were chosen since they were one of the sources most adjusted and also contribute to SOA). The correlation (R^2) in the calculated DDM sensitivities scaled by $PM_{2.5}$ concentration between the two versions is 0.94 (see Fig. S4), which is larger than the correlation in simulated $PM_{2.5}$ (0.86), and is much larger than the correlation between the simulated and observed $PM_{2.5}$ obtained by using contemporary CTMs (e.g., Figure 8 of Simon et al. 2012 shows that most of the correlations were between approximately 0.2-0.5). Given that the sensitivities and simulated concentrations are both quite similar, and much more correlated than between the modeled concentrations and observations, the use of a different version in this application makes little difference. For these reasons, we do not believe that using v4.7.1 instead of v4.5 would have substantially impacted our results.

Note S2 Correction of organic and elemental carbon measurements at CSN sites

Monthly, sampler type specific additive positive artifact *a* as well as multiplicative negative artifact *m* (see Malm et al. (2011) for details) are used. The majority of the CSN sites use Met One, Andersen and URG samplers. The measurements from other types of samplers such as various R&P samplers were removed from analysis due to no appropriate *a* and *m* available for them.

Note S3 Ignoring higher order sensitivities

One could include higher order sensitivities in approximating source impact results, especially for sectors that contribute to nitrate and ammonium. In fact our hybrid

method, as designed, can add source impact portions estimated from using higher order sensitivities when they are available.

The first order DDM sensitivities usually don't exactly add up to the predicted total $PM_{2.5}$ concentrations due to ignoring the higher order sensitivities. However, among the 932 cases we have tested with our hybrid method, only 125 cases (13.4%) had a larger than 30% difference. We have done the analysis of our hybrid method application both with and without these cases (out of one-moth long measurements at 164 CSN sites), and found that the changes are small.

Both Burr and Zhang (2011) and Koo et al. (2009) have found that the sum of BFM estimated source contributions will not equal the simulated base case concentrations either. On the other hand, Koo et al. (2009) have shown that first order DDM gave reasonably good predictions for impacts of SOA precursors, primary aerosols and on-road mobile source emissions. Their results in Figures 4, 6 and 7 also show that first-order DDM compared reasonably well with BFM for 100% reductions at apportioning sulfate, nitrate and ammonium to sources emitting SO_2, NO_x and NH_3 in winter times, with the nitrate estimation being slightly worse. They have also shown that first-order DDM performs well in determining the impact of sources that have indirect effects, such as the motor vehicle emissions that include multiple pollutants.

First order DDM sensitivities best approximate a small perturbation in a linear system. Note that our first-order DDM sensitivities were calculated for all emitted compounds from the sources, and this process utilized the DDM's proven advantage of capturing indirect effects. Also, we have split the total emissions into 33 sectors, which made the emissions of most sectors constitute a small fraction of total emissions. This

further diminishes the disadvantage of first-order DDM not capturing well the impacts from large changes of emissions. The hybrid method is designed to improve on an imperfect CTM-based source impact estimation method (no method is perfect on source apportionment), though we only showed the improvement with a DDM-sensitivity-based source impact estimation method here.

Note S4 Regrouping the hybrid results

First, we split the secondary contributions from primary contributions for each of the 33 sources, using source specific composition profiles. Then we merged the primary portions of 33 sources into the 13 reduced sources. Finally we merged the secondary portions correspondingly into ammonium sulfates, ammonium nitrate, and secondary organic carbon. This regrouping made the hybrid method results directly comparable to those of RM methods.

Note S5 RM model applications for the comparison

The Atlanta CSN site (130890002) is located in South Dekalb in Atlanta, near an interstate highway, not far away (~10 km) from the Hartsfield-Jackson Atlanta International Airport, one of the "busiest" airports in the world. The CMB-LGO method was conducted with the 8 source profiles (5 primary and 3 secondary) used by Balachandran et al. (2012). The Chicago site (170310076) is located in Lawndale, a residential community on the southwest side of Chicago. The CMB results that were resolved for 8 sources (no split between gasoline and diesel vehicles) from Rizzo and Scheff (2007)used measurement data composites from this site and another nearby CSN

site at Springfield, with twice as many samples coming from this site than the other site. The Detroit site (261630001) is located in Allen Park, a populated area southwest of the Detroit industrial center. Gildemeister et al. (2007) used PMF method at this site and revolved 8 sources (factors), including metal processing and road salts. The Los Angeles site (060658001) is located in Rubidoux of Riverside County in a suburban residential area, downwind of downtown Los Angeles. Pham et al. (2008) used CMB with 10 source profiles including profiles for cooking process, residual oil burning and sea salt. The OC measurements were not used in the CMB modeling because they were believed to be highly biased. The New York site (360050083) is located in the middle of the Bronx, a heavily populated urban area. Coutant et al. (2003) used PMF at this site and resolved 7 sources, including fuel oil burning. The Pittsburgh site (420030008) is located in Lawrenceville, an urban residential site, downwind from the Pittsburgh downtown. Maranche (2006) used PMF at this site and resolved 11 sources (factors). We chose to aggregate the 11 sources to 7 sources due to the non-distinctiveness of some of the original 11 sources.

Table S1. Performance evaluation of MM5-generated meteorological parameter fields compared to the Techniques Development Laboratory (TDL) surface observations.

Wind speed (m/s)			Wind direction (deg)			Air temperature (K)			Humidity (g/kg)		
Mean OBS	Bias	RMSE	Mean OBS	Bias	Gross error	Mean OBS	Bias	RMSE	Mean OBS	Bias	Gross error
3.72	-0.01	2.02	261.25	2.56	25.98	270.63	-0.79	3.23	3.33	0.01	0.52

*RMSE: Root mean square error.

Table S2. Source classification codes (SCCs) grouped in the 33 source categories.

Category	SCCs included (with leading digits)	Source Inventory	Emission Uncertainty	$\sigma_{\ln R_j}$
AGRIBURN	280150	Fire inventory	±5	1.792
AIRCRAFT	2275	Non-road inventory	±0.5	0.405
BIOGENIC	27010, 27012	BEIS3 calculation	±0.5	0.405
COALCMB	101001, 101002, 101003, 102001, 102002, 102003, 103001, 103002, 103003, 10500102, 10500202, 390001, 390002, 390003, 2101001, 2101002, 2101003, 2102001, 2102002, 2103001, 2103002, 2103003, 2104001, 2104001, 2104002, 2104003, 2199001, 2199002, 2199003, 2390001, 2390002, 2390003	Point and area inventories	±0.1	0.095
DIESELCMB	20200401, 20400302, 20400402, 27000, 2800	Point inventory	±0.3	0.262
DUST	2275085000, 2294000000, 2294000001, 2294000002, 2294005000, 2294005002, 2294010000, 2294010001, 2294010002, 2294015000, 2294015001, 2294015002, 2296000000, 2296005000, 2296010000, 2311000000, 2311000010, 2311000040, 2311000050, 2311000060, 2311000070, 2311010000, 2311010010, 2311010040, 2311010050, 2311010060, 2311010070, 2311020000, 2311020010, 2311020040, 2311020050, 2311020060, 2311020070, 2311030000, 2311030010, 2311030040, 2311030050, 2311030060, 2311030070, 2311040000, 2801000000, 2801000001, 2801000002, 2801000003, 2801000004, 2801000005, 2801000006, 2801000007, 2801000008, 30300519, 30300831, 30300832, 30300833, 30300834, 30302321, 30302322, 30500290, 30501024, 30501030, 30501031, 30501039, 30501045, 30501046, 30501048, 30501049, 30501050, 30501090, 30501640, 30502011, 30502504, 30531090, 31100101, 31100102, 31100103, 31100205, 31100206, 50100401	Dust-area and point inventories. Including road dust, industrial dust and livestock dust etc.	±10	2.398
FUELOILCMB	101004, 101005, 102005, 103004, 103005, 10500105, 10500205, 201001, 202001, 202005, 203001, 20400303, 20400403, 20400408, 30190001, 30190002, 30190011, 30190012, 30190021, 30190022, 30290001, 30290002, 30390001, 30390002, 30390011, 30390012, 30390021, 30390022, 30490001, 30490002, 30490011, 30490012, 30490021, 30490022, 30490031, 30490032, 30590001, 30590002, 30590011, 30590012, 30590021, 30590022, 30600101, 30600103, 30600111, 30600901, 30600902, 30790001, 30790002, 30790011, 30790012, 30790021, 30790022, 30890001, 30890002, 30890011, 30890012, 30890021, 30890022, 30990001, 30990002, 30990011, 30990012, 31000401, 31000401, 31000402, 31000403, 31000411, 31000412, 31000413, 31390001, 31390002, 390004, 390005, 39900501, 39990001, 39990011, 39990012, 399900221, 39990022, 2101004, 2101005, 2102004, 2102005, 2103004, 2103005, 2104004, 2104005, 2199004, 2199005, 2399004, 2399005	Point and area inventories	±1.0	0.693
LIVESTOCK	2805, 2806, 2807, 302021, 302020	Area and point inventories	±0.3	0.262
LPGCMB	101010, 102010, 103010, 10500110, 10500210, 202010, 203010, 20400409, 27300, 30290005, 30600107, 30890004, 390010, 399010, 2101007, 2102007, 2103007, 2104007, 2199007, 2399007	Point and area inventories	±0.3	0.262
LWASTEBURN	50100202, 502002, 50300201, 2610	Non-fire inventories	±5	1.792
METALPRODUCT	3030, 3038, 3040, 3048, 3090, 3098, 2303, 2304,2309,284101	Point and area inventories	±0.3	0.262
MEATCOOKING	302013, 302036, 30203803, 2302002, 2302003	Point and area inventories	±5	1.792
MEXCMB_M	2101, 2102, 2103, 2104	Mexican Inventory only	±0.5	0.405
MINERALPRODUCT	3050, 3051, 3053, 3058, 2305	Point inventory	±0.3	0.262

NAGASCMB	101006, 102006, 10201401, 103006, 10500106, 201002, 203002, 20400301, 30190003, 30190013, 30190023, 30290003, 30390013, 30490003, 30490013, 30490023, 30490033, 30500206, 30590003, 30590013, 30590023, 30600105, 30600903, 30609903, 30790003, 30790013, 30790023, 30890003, 30890013, 30890023, 30990003, 30990013, 30990023, 31000404, 31000414, 31390003, 390006, 39900601, 39990003, 39990013, 39990023, 40201001, 40290013, 40290023, 50190006, 50290006, 50390006, 2101006, 2102006, 2103006, 2104006, 2199006, 2399006	Point and area inventories	±0.5	0.405
NRDIESEL	2270, 2280002,2282020,2283002	Non-road inventory	±0.5	0.405
NRFUELOIL	2280003	Non-road inventory	±0.5	0.405
NRGASOLINE	2260,2265,2280004,2282005,2282010	Non-road inventory	±0.5	0.405
NRLPG	2267	Non-road inventory	±0.5	0.405
NRNAGAS	2268	Non-road inventory	±0.5	0.405
NROTHERS	2280001	Non-road inventory	±0.5	0.405
OPENFIRE	2610	Fire inventory	±5	1.792
ORDIESEL	2230	Mobile6 Calculation	±0.5	0.405
ORGASOLINE	2201	Mobile6 Calculation	±0.5	0.405
OTHERCMB	20200301, 202017, 203003, 20400401, 2600, 2650, 2750, 2760, 399017, 101007, 101008, 101011, 101012, 101013, 101014, 101015, 102007, 102008, 102011, 102012, 102013, 102014, 102015, 102016, 102017, 103008, 103012, 103013, 10500113, 10500213, 10500214, 201007, 201008, 201009, 201010, 201013, 20200402, 20200403, 202007, 202009, 202017, 203007, 203008, 203009, 204001, 204002, 20400304, 20400305, 20400399, 20400404, 20400405, 20400407, 20400499, 30190004, 30190014, 30190024, 30390004, 30390024, 30490004, 30490014, 30490024, 30490034, 30490035, 30600102, 30600104, 30600106, 30600108, 30600199, 30600904, 30600906, 30600999, 30790004, 30790024, 30890024, 31000405, 31000415, 390007, 390008, 390012, 390013, 399007, 399008, 399016, 39990004, 39990014, 39990024, 2101009, 2101010, 2101011, 2102009, 2102010, 2102011, 2104009, 2104010, 2104011, 2199009, 2199010, 2199011, 2390009, 2390010, 2390011, 2801520000, 2801520004, 2801520010, 28100250	Point and area inventories	±0.5	0.405
OTHERS	All leftover SCCs	Point and area inventories	±1.0	0.693
PRESCRBURN	2810005,2810015,2810020		±10	2.398
RAILROAD	2285	Non-road inventory	±0.5	0.405
SEASALT	274004	CMAQ Sea-salt Module	±0.5	0.405
SOLVENT	24, 2501, 2505, 2510, 2515, 2306, 2310, 306008, 3909, 4	Area and point inventory	±0.5	2.398
WILDFIRE	2810001	Fire inventory	10	0.405
WOODFUEL	101009, 102009, 103009, 10500209, 2101008,2102008, 2103008,2199008,2390008	Point and area inventories	±0.5	1.792
WOODSTOVE	2104008	Area inventory	±5	0.405

Table S3. Performance evaluation of CMAQ modeled concentrations (initial predictions) compared to the AQS, SLAMS, CSN, IMPROVE and SEARCH observations.

Pollutant	Mean of Observation	Number of Pairs	MNB (%)	MNE (%)
1-hr O_3 (with 40ppb cutoff)	44.3 ppb	33054	3.2	18.7
8-hr O_3 (with 40ppb cutoff)	44.2 ppb	22712	1.4	14.5
1-hr NO_2	9.0 ppb	247911	51.2	91.1
1-hr CO	678.9 ppb	259185	12.7	80.9
			FB (%)	FE (%)
1-hr PM2.5	10.5 μg m^{-3}	163743	34.2	70.9
24-hr PM2.5	11.5 μg m^{-3}	11871	27.8	54.4
24-hr Sulfate	1.6 μg m^{-3}	2865	35.1	54.4
24-hr Nitrate	1.9 μg m^{-3}	2733	-44.4	95.9
24-hr Ammonium	1.0 μg m^{-3}	2862	10.1	58.1
24-hr EC	0.4 μg m^{-3}	2828	23.0	71.0
24-hr OC (no corrections to CSN observations)	3.5 μg m^{-3}	2843	2.4	63.8

* The examined bias and error measures were calculated as follows:

$$MNB = \frac{1}{N}\sum_{i=1}^{N}\frac{\left(C_i^{sim} - C_i^{obs}\right)}{C_i^{obs}} \times 100\%, \quad MNE = \frac{1}{N}\sum_{i=1}^{N}\frac{\left|C_i^{sim} - C_i^{obs}\right|}{C_i^{obs}} \times 100\%, \quad FB = \frac{1}{N}\sum_{i=1}^{N}\frac{2\left(C_i^{sim} - C_i^{obs}\right)}{\left(C_i^{sim} + C_i^{obs}\right)} \times 100\%, \quad FE = \frac{1}{N}\sum_{i=1}^{N}\frac{2\left|C_i^{sim} - C_i^{obs}\right|}{\left(C_i^{sim} + C_i^{obs}\right)} \times 100\%.$$

Table S4. Species that measured at CSN monitoring sites

	Total mass	Major Components	Elements (35)
41 species	$PM_{2.5}$	EC, OC, Sulfate, Nitrate, Ammonium	Na, Mg, Al , Si, P, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Zr, Mo, Ag, Cd, In, Sn, Sb, Ba, La, Ce, Hg, Pb

Table S5. Composite $PM_{2.5}$ source composition profiles for the 33 categories

Species	AGRIBURN	AIRCRAFT	BIOGENIC	COALCMB	DIESELCM	DUST	FUELOILC
OC	3.88E-01	1.76E-01	0.00E+00	3.87E-02	1.76E-01	4.87E-02	1.14E-01
EC	1.09E-01	7.71E-01	0.00E+00	2.05E-02	7.71E-01	1.73E-03	4.91E-02
NH4	1.80E-02	0.00E+00	0.00E+00	2.42E-02	0.00E+00	8.62E-04	0.00E+00
NO3	3.50E-03	1.14E-03	0.00E+00	1.98E-03	1.14E-03	1.80E-03	0.00E+00
SO4	1.65E-02	2.95E-03	0.00E+00	1.22E-01	2.95E-03	5.98E-03	3.32E-01
NCOM	2.72E-01	4.39E-02	0.00E+00	1.55E-02	4.39E-02	1.95E-02	4.57E-02
MO	3.51E-03	1.39E-04	0.00E+00	1.54E-01	1.39E-04	2.99E-01	0.00E+00
H2O	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.64E-03	0.00E+00
Na	6.55E-03	0.00E+00	0.00E+00	1.16E-03	0.00E+00	1.13E-03	0.00E+00
Mg	8.06E-04	0.00E+00	0.00E+00	4.26E-03	0.00E+00	4.04E-03	0.00E+00
Al	3.00E-04	0.00E+00	0.00E+00	4.35E-02	0.00E+00	5.94E-02	0.00E+00
Si	1.50E-04	0.00E+00	0.00E+00	8.02E-02	0.00E+00	1.74E-01	3.47E-03
P	0.00E+00	0.00E+00	0.00E+00	2.48E-03	0.00E+00	9.58E-04	0.00E+00
S	6.50E-03	2.52E-03	0.00E+00	5.21E-02	2.52E-03	2.69E-03	8.32E-02
Cl	9.05E-02	2.05E-04	0.00E+00	5.99E-03	2.05E-04	1.40E-03	1.77E-03
K	7.04E-02	3.80E-05	0.00E+00	4.25E-03	3.80E-05	1.71E-02	1.74E-04
Ca	3.30E-04	5.83E-04	0.00E+00	3.93E-02	5.83E-04	5.38E-02	2.85E-04
Ti	1.00E-05	4.00E-06	0.00E+00	2.44E-03	4.00E-06	3.89E-03	3.64E-04
V	0.00E+00	6.00E-06	0.00E+00	1.05E-04	6.00E-06	2.49E-04	4.64E-04
Cr	0.00E+00	0.00E+00	0.00E+00	8.41E-05	0.00E+00	2.21E-04	1.08E-04
Mn	0.00E+00	0.00E+00	0.00E+00	1.76E-04	0.00E+00	1.17E-03	2.09E-05
Fe	1.00E-04	2.62E-04	0.00E+00	2.06E-02	2.62E-04	4.43E-02	5.56E-04
Co	0.00E+00	2.00E-06	0.00E+00	4.82E-06	2.00E-06	4.33E-05	0.00E+00
Ni	0.00E+00	2.00E-06	0.00E+00	4.26E-05	2.00E-06	6.66E-05	2.73E-04
Cu	0.00E+00	0.00E+00	0.00E+00	1.46E-04	0.00E+00	1.11E-04	8.68E-05
Zn	1.00E-04	6.82E-04	0.00E+00	5.07E-04	6.82E-04	5.15E-04	7.14E-04
Ga	0.00E+00	0.00E+00	0.00E+00	2.02E-05	0.00E+00	5.74E-07	0.00E+00
As	0.00E+00	3.00E-06	0.00E+00	1.24E-06	3.00E-06	1.77E-05	0.00E+00
Se	0.00E+00	0.00E+00	0.00E+00	2.81E-03	0.00E+00	1.10E-06	2.84E-05
Br	2.80E-04	1.50E-05	0.00E+00	2.93E-04	1.50E-05	1.23E-05	0.00E+00
Rb	1.00E-04	1.00E-06	0.00E+00	2.93E-05	1.00E-06	1.03E-04	0.00E+00
Sr	0.00E+00	1.00E-06	0.00E+00	1.34E-03	1.00E-06	3.15E-04	0.00E+00
Zr	0.00E+00	0.00E+00	0.00E+00	9.94E-05	0.00E+00	1.26E-04	0.00E+00
Mo	0.00E+00	2.00E-06	0.00E+00	9.56E-06	2.00E-06	5.23E-06	0.00E+00
Pd	5.15E-05	2.60E-05	0.00E+00	4.98E-05	2.60E-05	1.99E-05	0.00E+00
Ag	1.55E-05	6.10E-05	0.00E+00	8.57E-05	6.10E-05	4.10E-06	0.00E+00
Cd	0.00E+00	4.40E-05	0.00E+00	2.36E-05	4.40E-05	1.46E-05	0.00E+00
In	5.00E-05	1.80E-05	0.00E+00	3.64E-05	1.80E-05	7.12E-06	0.00E+00
Sn	0.00E+00	2.50E-05	0.00E+00	1.30E-04	2.50E-05	2.80E-05	0.00E+00
Sb	0.00E+00	1.90E-05	0.00E+00	8.13E-06	1.90E-05	3.22E-05	1.33E-04
Ba	2.50E-04	3.50E-04	0.00E+00	3.91E-03	3.50E-04	7.88E-04	0.00E+00
La	0.00E+00	2.72E-04	0.00E+00	1.68E-05	2.72E-04	1.14E-04	2.85E-04
Ce	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Hg	0.00E+00	0.00E+00	0.00E+00	9.20E-06	0.00E+00	2.09E-06	0.00E+00
Pb	1.00E-05	1.00E-06	0.00E+00	1.22E-04	1.00E-06	2.22E-04	0.00E+00
PMO	1.96E-02	2.42E-03	0.00E+00	4.09E-01	2.42E-03	2.57E-01	4.51E-01
Total	1.0066E+00	1.0027E+00	0.0000E+00	1.0522E+00	1.0027E+00	1.0025E+00	1.0832E+00

* "other" $PM_{2.5}$ equals to the Total with OC, EC, NH4, NO3, SO4, NCOM, H2O and S being removed.

Species	LIVESTOCK	LPGCMB	LWASTEBU	MEATALPR	MEATCOOK	MEXCMB_M	MINERALP
OC	0.00E+00	2.47E-01	3.88E-01	7.05E-02	6.54E-01	1.86E-01	7.72E-02
EC	0.00E+00	3.84E-01	1.09E-01	1.41E-02	3.41E-02	8.06E-02	1.98E-02
NH4	0.00E+00	0.00E+00	1.80E-02	4.06E-04	0.00E+00	8.39E-04	5.92E-03
NO3	0.00E+00	2.10E-02	3.50E-03	5.18E-03	4.56E-03	4.48E-04	1.32E-02
SO4	0.00E+00	8.60E-02	1.65E-02	1.84E-01	2.59E-03	1.86E-01	1.41E-01
NCOM	0.00E+00	9.88E-02	2.72E-01	2.82E-02	2.62E-01	7.45E-02	3.08E-02
MO	0.00E+00	0.00E+00	3.51E-03	1.17E-01	4.53E-03	4.15E-02	1.30E-01
H2O	0.00E+00	0.00E+00	0.00E+00	4.62E-03	9.70E-09	0.00E+00	2.07E-02
Na	0.00E+00	0.00E+00	6.55E-03	3.09E-02	3.23E-03	1.98E-04	2.94E-02
Mg	0.00E+00	0.00E+00	8.06E-04	4.60E-03	8.00E-04	0.00E+00	3.24E-03
Al	0.00E+00	0.00E+00	3.00E-04	3.48E-02	4.36E-04	1.37E-02	2.49E-02
Si	0.00E+00	0.00E+00	1.50E-04	4.01E-02	8.08E-04	2.63E-02	6.27E-02
P	0.00E+00	0.00E+00	0.00E+00	1.55E-03	4.00E-04	2.15E-03	3.90E-04
S	0.00E+00	2.87E-02	6.50E-03	6.16E-02	1.88E-03	3.45E-02	5.03E-02
Cl	0.00E+00	0.00E+00	9.05E-02	1.01E-01	8.75E-03	7.11E-04	2.28E-02
K	0.00E+00	0.00E+00	7.04E-02	3.74E-02	2.65E-03	1.36E-03	2.37E-02
Ca	0.00E+00	0.00E+00	3.30E-04	1.31E-02	5.22E-04	8.17E-03	6.63E-02
Ti	0.00E+00	0.00E+00	1.00E-05	8.86E-04	2.80E-05	1.04E-03	1.86E-03
V	0.00E+00	0.00E+00	0.00E+00	1.09E-03	1.67E-06	5.56E-05	1.23E-04
Cr	0.00E+00	0.00E+00	0.00E+00	5.04E-03	2.41E-04	2.19E-04	4.26E-04
Mn	0.00E+00	0.00E+00	0.00E+00	5.06E-03	6.68E-05	6.92E-05	6.29E-04
Fe	0.00E+00	0.00E+00	1.00E-04	7.49E-02	7.72E-04	7.35E-03	1.17E-02
Co	0.00E+00	0.00E+00	0.00E+00	2.43E-05	6.70E-06	0.00E+00	7.58E-05
Ni	0.00E+00	0.00E+00	0.00E+00	6.03E-03	1.32E-04	5.67E-05	2.16E-04
Cu	0.00E+00	0.00E+00	0.00E+00	5.48E-03	1.20E-04	1.80E-04	1.45E-04
Zn	0.00E+00	0.00E+00	1.00E-04	2.55E-03	2.58E-04	1.27E-03	1.16E-03
Ga	0.00E+00	0.00E+00	0.00E+00	1.98E-05	0.00E+00	3.05E-05	1.21E-06
As	0.00E+00	0.00E+00	0.00E+00	4.09E-04	2.93E-06	0.00E+00	5.57E-05
Se	0.00E+00	0.00E+00	0.00E+00	1.68E-04	1.01E-05	1.02E-04	7.68E-05
Br	0.00E+00	0.00E+00	2.80E-04	1.42E-03	1.67E-04	3.74E-05	3.43E-04
Rb	0.00E+00	0.00E+00	1.00E-04	5.68E-04	1.46E-04	8.17E-06	2.22E-04
Sr	0.00E+00	0.00E+00	0.00E+00	2.97E-04	2.07E-05	4.49E-04	3.27E-04
Zr	0.00E+00	0.00E+00	0.00E+00	1.56E-04	0.00E+00	5.69E-05	7.77E-05
Mo	0.00E+00	0.00E+00	0.00E+00	1.39E-03	0.00E+00	1.35E-05	6.61E-05
Pd	0.00E+00	0.00E+00	5.15E-05	1.82E-06	0.00E+00	0.00E+00	1.57E-05
Ag	0.00E+00	0.00E+00	1.55E-05	1.44E-04	3.68E-05	0.00E+00	2.26E-05
Cd	0.00E+00	0.00E+00	0.00E+00	3.01E-04	1.67E-06	0.00E+00	3.73E-05
In	0.00E+00	0.00E+00	5.00E-05	2.15E-05	1.67E-06	0.00E+00	4.50E-05
Sn	0.00E+00	0.00E+00	0.00E+00	3.78E-04	1.17E-05	0.00E+00	1.22E-04
Sb	0.00E+00	0.00E+00	0.00E+00	8.15E-04	2.26E-05	1.55E-05	1.63E-05
Ba	0.00E+00	0.00E+00	2.50E-04	2.81E-03	9.15E-04	3.05E-03	6.51E-04
La	0.00E+00	0.00E+00	0.00E+00	1.69E-06	8.37E-05	3.32E-05	1.39E-04
Ce	0.00E+00	0.00E+00	0.00E+00	4.59E-04	0.00E+00	0.00E+00	2.99E-05
Hg	0.00E+00	0.00E+00	0.00E+00	1.65E-06	8.37E-07	0.00E+00	5.55E-06
Pb	0.00E+00	0.00E+00	1.00E-05	2.10E-03	3.77E-04	1.56E-04	4.63E-04
PMO	0.00E+00	1.63E-01	1.96E-02	2.01E-01	1.69E-02	3.64E-01	3.09E-01
Total	0.0000E+00	1.0285E+00	1.0066E+00	1.0617E+00	1.0019E+00	1.0349E+00	1.0508E+00

Species	NAGASCMB	NRDIESEL	NRFUELOI	NRGASOL	NRLPG	NRNAGAS	NROTHERS
OC	4.76E-01	1.76E-01	1.00E-02	4.75E-01	4.90E-01	4.90E-01	2.53E-01
EC	8.51E-02	7.71E-01	1.00E-02	1.22E-01	6.70E-02	6.70E-02	6.03E-01
NH4	4.15E-03	0.00E+00	0.00E+00	0.00E+00	4.40E-03	4.40E-03	2.77E-05
NO3	3.34E-02	1.14E-03	0.00E+00	7.00E-04	3.41E-02	3.41E-02	1.24E-03
SO4	1.24E-01	2.95E-03	4.40E-01	5.00E-04	1.26E-01	1.26E-01	3.11E-03
NCOM	1.90E-01	4.39E-02	4.00E-03	1.19E-01	1.96E-01	1.96E-01	6.38E-02
MO	4.15E-04	1.39E-04	0.00E+00	1.95E-03	4.40E-04	4.40E-04	5.97E-04
H2O	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Na	2.01E-02	0.00E+00	0.00E+00	0.00E+00	2.13E-02	2.13E-02	1.34E-04
Mg	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Al	2.07E-03	0.00E+00	0.00E+00	0.00E+00	2.20E-03	2.20E-03	1.38E-05
Si	2.64E-03	0.00E+00	0.00E+00	1.20E-03	2.80E-03	2.80E-03	3.20E-04
P	2.83E-04	0.00E+00	0.00E+00	3.00E-04	3.00E-04	3.00E-04	7.75E-05
S	4.12E-02	2.52E-03	1.26E-01	1.67E-04	4.20E-02	4.20E-02	2.18E-03
Cl	3.66E-02	2.05E-04	3.13E-03	0.00E+00	3.88E-02	3.88E-02	3.96E-04
K	1.60E-03	3.80E-05	0.00E+00	0.00E+00	1.70E-03	1.70E-03	3.89E-05
Ca	9.81E-03	5.83E-04	3.51E-04	3.00E-04	1.04E-02	1.04E-02	5.73E-04
Ti	9.43E-04	4.00E-06	6.43E-04	0.00E+00	1.00E-03	1.00E-03	9.25E-06
V	1.70E-04	6.00E-06	8.19E-04	0.00E+00	1.80E-04	1.80E-04	5.58E-06
Cr	4.72E-04	0.00E+00	0.00E+00	0.00E+00	5.00E-04	5.00E-04	3.14E-06
Mn	1.89E-04	0.00E+00	3.70E-05	0.00E+00	2.00E-04	2.00E-04	1.26E-06
Fe	8.49E-04	2.62E-04	2.93E-04	1.00E-04	9.00E-04	9.00E-04	2.25E-04
Co	0.00E+00	2.00E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.48E-06
Ni	8.49E-04	2.00E-06	4.83E-04	0.00E+00	9.00E-04	9.00E-04	7.14E-06
Cu	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Zn	0.00E+00	6.82E-04	7.38E-05	3.00E-04	0.00E+00	0.00E+00	5.81E-04
Ga	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
As	0.00E+00	3.00E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.23E-06
Se	5.75E-04	0.00E+00	5.02E-05	0.00E+00	6.10E-04	6.10E-04	3.83E-06
Br	3.77E-04	1.50E-05	0.00E+00	0.00E+00	4.00E-04	4.00E-04	1.36E-05
Rb	1.89E-04	1.00E-06	0.00E+00	0.00E+00	2.00E-04	2.00E-04	2.00E-06
Sr	0.00E+00	1.00E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.42E-07
Zr	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Mo	0.00E+00	2.00E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.48E-06
Pd	0.00E+00	2.60E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.93E-05
Ag	0.00E+00	6.10E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.52E-05
Cd	0.00E+00	4.40E-05	0.00E+00	1.00E-04	0.00E+00	0.00E+00	5.78E-05
In	0.00E+00	1.80E-05	0.00E+00	1.00E-04	0.00E+00	0.00E+00	3.85E-05
Sn	0.00E+00	2.50E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.85E-05
Sb	0.00E+00	1.90E-05	2.35E-04	2.00E-04	0.00E+00	0.00E+00	6.45E-05
Ba	0.00E+00	3.50E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.60E-04
La	0.00E+00	2.72E-04	5.04E-04	0.00E+00	0.00E+00	0.00E+00	2.02E-04
Ce	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Hg	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Pb	0.00E+00	1.00E-06	0.00E+00	3.00E-04	0.00E+00	0.00E+00	7.63E-05
PMO	9.29E-03	2.42E-03	5.29E-01	2.78E-01	0.00E+00	0.00E+00	7.18E-02
Total	1.0415E+00	1.0027E+00	1.1256E+00	1.0002E+00	1.0423E+00	1.0423E+00	1.0023E+00

Species	OPENFIRE	ORDIESEL	ORGASOL	OTHERCMB	OTHERS	PRESCRBU	RAILROAD
OC	3.88E-01	1.84E-01	4.11E-01	2.41E-01	1.12E-01	5.03E-01	1.76E-01
EC	1.09E-01	7.47E-01	1.45E-01	1.01E-01	2.31E-02	1.05E-01	7.71E-01
NH4	1.80E-02	1.66E-04	1.00E-02	3.15E-02	7.49E-03	3.19E-03	4.85E-09
NO3	3.50E-03	1.18E-03	1.54E-03	8.81E-03	8.13E-03	1.01E-02	1.14E-03
SO4	1.65E-02	3.92E-03	1.80E-02	1.26E-01	1.39E-01	3.91E-03	2.95E-03
NCOM	2.72E-01	4.71E-02	1.15E-01	9.67E-02	4.58E-02	3.52E-01	4.39E-02
MO	3.51E-03	3.39E-03	7.49E-02	9.91E-02	5.74E-02	4.68E-03	1.39E-04
H2O	0.00E+00	2.25E-04	3.18E-03	2.93E-07	1.40E-02	0.00E+00	0.00E+00
Na	6.55E-03	2.01E-05	7.59E-04	2.19E-03	4.36E-02	2.38E-03	2.35E-08
Mg	8.06E-04	1.52E-03	3.29E-02	7.61E-04	1.00E-03	0.00E+00	0.00E+00
Al	3.00E-04	3.09E-05	1.31E-03	8.09E-03	9.45E-03	4.97E-04	2.42E-09
Si	1.50E-04	1.28E-03	2.88E-02	6.71E-02	2.95E-02	1.97E-04	6.54E-08
P	0.00E+00	2.50E-05	1.17E-03	1.12E-03	9.65E-04	6.20E-04	1.59E-08
S	6.50E-03	2.81E-03	7.61E-03	4.33E-02	4.40E-02	1.58E-03	2.52E-03
Cl	9.05E-02	3.39E-04	1.77E-03	2.77E-02	1.79E-02	2.57E-03	2.05E-04
K	7.04E-02	4.50E-05	2.06E-04	3.61E-02	1.22E-02	2.11E-03	3.80E-05
Ca	3.30E-04	7.13E-04	4.77E-03	9.98E-03	1.12E-02	7.73E-04	5.83E-04
Ti	1.00E-05	5.84E-05	1.13E-03	6.05E-04	5.53E-03	6.34E-04	4.00E-06
V	0.00E+00	1.03E-05	1.10E-04	1.08E-04	1.51E-04	1.19E-05	6.00E-06
Cr	0.00E+00	1.54E-05	3.52E-04	6.31E-05	2.61E-04	2.55E-05	5.51E-10
Mn	0.00E+00	1.62E-05	3.49E-04	1.72E-04	4.75E-04	1.14E-04	2.20E-10
Fe	1.00E-04	1.91E-03	3.66E-02	8.62E-03	6.95E-03	4.44E-04	2.62E-04
Co	0.00E+00	2.87E-06	2.50E-05	4.45E-06	1.62E-05	0.00E+00	2.00E-06
Ni	0.00E+00	1.17E-05	2.37E-04	2.05E-04	2.82E-04	2.32E-05	2.00E-06
Cu	0.00E+00	1.65E-04	3.55E-03	2.34E-03	2.74E-04	2.19E-05	0.00E+00
Zn	1.00E-04	7.64E-04	2.66E-03	7.50E-03	4.97E-04	2.16E-03	6.82E-04
Ga	0.00E+00	5.42E-08	5.20E-06	1.66E-05	4.94E-06	0.00E+00	0.00E+00
As	0.00E+00	3.10E-06	6.04E-06	2.28E-05	2.89E-05	0.00E+00	3.00E-06
Se	0.00E+00	5.81E-07	1.17E-05	1.90E-05	1.05E-05	0.00E+00	6.72E-10
Br	2.80E-04	1.52E-05	4.86E-05	5.55E-05	4.82E-04	1.03E-04	1.50E-05
Rb	1.00E-04	1.63E-06	1.57E-05	2.25E-05	5.81E-05	0.00E+00	1.00E-06
Sr	0.00E+00	9.87E-06	1.83E-04	8.56E-05	4.06E-05	0.00E+00	1.00E-06
Zr	0.00E+00	0.00E+00	5.37E-06	4.10E-05	3.78E-05	0.00E+00	0.00E+00
Mo	0.00E+00	5.24E-05	1.10E-03	3.61E-05	4.93E-05	0.00E+00	2.00E-06
Pd	5.15E-05	2.51E-05	3.52E-05	1.91E-05	1.56E-05	0.00E+00	2.60E-05
Ag	1.55E-05	5.91E-05	3.46E-05	2.76E-05	2.51E-05	1.95E-04	6.10E-05
Cd	0.00E+00	4.26E-05	4.33E-05	1.95E-04	4.02E-05	4.57E-03	4.40E-05
In	5.00E-05	1.79E-05	2.09E-05	1.07E-05	6.81E-06	0.00E+00	1.80E-05
Sn	0.00E+00	1.14E-04	2.01E-03	2.12E-05	6.86E-05	1.88E-04	2.50E-05
Sb	0.00E+00	1.96E-05	1.13E-04	4.05E-05	2.12E-04	0.00E+00	1.90E-05
Ba	2.50E-04	8.10E-04	1.05E-02	5.11E-04	5.34E-04	0.00E+00	3.50E-04
La	0.00E+00	2.59E-04	1.99E-04	1.64E-04	9.03E-04	0.00E+00	2.72E-04
Ce	0.00E+00	1.36E-07	2.93E-06	1.35E-05	7.76E-05	0.00E+00	0.00E+00
Hg	0.00E+00	0.00E+00	6.86E-06	1.40E-08	3.00E-06	0.00E+00	0.00E+00
Pb	1.00E-05	7.55E-06	3.16E-04	2.28E-05	1.36E-04	1.41E-04	1.02E-06
PMO	1.96E-02	5.57E-03	8.97E-02	1.21E-01	4.51E-01	0.00E+00	2.43E-03
Total	1.0066E+00	1.0030E+00	1.0075E+00	1.0431E+00	1.0439E+00	1.0012E+00	1.0027E+00

Species	SOLVENT	WILDFIRE	WOODFUEL	WOODSTOV	SEASALT
OC	2.74E-01	4.62E-01	3.51E-01	5.28E-01	0.00E+00
EC	4.65E-03	9.49E-02	3.71E-02	5.58E-02	0.00E+00
NH4	7.46E-04	8.79E-03	0.00E+00	1.50E-03	0.00E+00
NO3	3.55E-04	1.32E-03	0.00E+00	1.90E-03	0.00E+00
SO4	2.83E-02	1.26E-02	6.53E-02	4.10E-03	7.57E-02
NCOM	1.09E-01	3.23E-01	1.40E-01	3.70E-01	0.00E+00
MO	1.80E-01	1.28E-02	1.66E-01	3.02E-03	0.00E+00
H2O	6.98E-03	0.00E+00	0.00E+00	0.00E+00	2.40E-02
Na	2.41E-05	5.73E-03	1.51E-03	9.40E-04	3.01E-01
Mg	0.00E+00	3.14E-04	1.42E-03	1.13E-04	3.59E-02
Al	2.45E-02	6.08E-04	0.00E+00	1.10E-04	0.00E+00
Si	2.52E-02	1.82E-03	1.36E-01	3.40E-04	0.00E+00
P	0.00E+00	5.40E-05	7.53E-04	4.00E-05	0.00E+00
S	9.44E-03	4.32E-03	3.10E-02	1.82E-03	2.49E-02
Cl	3.16E-03	4.15E-02	5.35E-03	2.97E-03	5.41E-01
K	1.95E-03	2.94E-02	8.87E-02	9.67E-03	1.12E-02
Ca	2.50E-02	3.86E-03	6.36E-03	1.00E-04	1.15E-02
Ti	1.69E-01	5.15E-05	0.00E+00	0.00E+00	0.00E+00
V	0.00E+00	3.50E-06	0.00E+00	0.00E+00	0.00E+00
Cr	0.00E+00	8.00E-06	0.00E+00	0.00E+00	0.00E+00
Mn	0.00E+00	1.60E-05	0.00E+00	0.00E+00	0.00E+00
Fe	3.33E-04	4.34E-04	0.00E+00	9.00E-05	0.00E+00
Co	6.61E-05	5.00E-07	0.00E+00	0.00E+00	0.00E+00
Ni	0.00E+00	5.50E-06	0.00E+00	0.00E+00	0.00E+00
Cu	1.54E-05	1.60E-05	0.00E+00	0.00E+00	0.00E+00
Zn	1.10E-05	1.67E-04	0.00E+00	2.20E-04	0.00E+00
Ga	0.00E+00	5.00E-07	0.00E+00	0.00E+00	0.00E+00
As	0.00E+00	1.23E-04	0.00E+00	2.00E-06	0.00E+00
Se	0.00E+00	6.00E-06	0.00E+00	0.00E+00	0.00E+00
Br	9.91E-05	4.20E-04	0.00E+00	2.00E-05	0.00E+00
Rb	6.61E-05	1.90E-05	0.00E+00	6.00E-06	0.00E+00
Sr	3.30E-05	3.85E-05	0.00E+00	0.00E+00	0.00E+00
Zr	4.30E-04	6.00E-06	0.00E+00	0.00E+00	0.00E+00
Mo	3.30E-05	5.00E-07	0.00E+00	0.00E+00	0.00E+00
Pd	0.00E+00	5.00E-07	0.00E+00	1.00E-06	0.00E+00
Ag	0.00E+00	2.50E-06	0.00E+00	7.00E-06	0.00E+00
Cd	4.63E-04	5.50E-06	0.00E+00	7.00E-06	0.00E+00
In	0.00E+00	1.45E-05	0.00E+00	0.00E+00	0.00E+00
Sn	0.00E+00	1.45E-05	0.00E+00	1.00E-05	0.00E+00
Sb	4.69E-03	7.00E-06	0.00E+00	0.00E+00	0.00E+00
Ba	1.25E-02	2.25E-05	0.00E+00	0.00E+00	0.00E+00
La	1.86E-02	2.00E-06	0.00E+00	1.00E-05	0.00E+00
Ce	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Hg	0.00E+00	2.50E-06	0.00E+00	0.00E+00	0.00E+00
Pb	3.00E-04	1.60E-05	0.00E+00	6.00E-06	0.00E+00
PMO	1.10E-01	0.00E+00	0.00E+00	2.11E-02	0.00E+00
Total	1.0096E+00	1.0044E+00	1.0305E+00	1.0019E+00	1.0249E+00

Table S6. Typical normalized prediction errors (δ_i) of CTM

Species	$PM_{2.5}$	EC	OC	Sulfate	Nitrate	Ammonium	Elements
δ_i	0.4	0.6	0.8	0.3	0.8	0.4	0.35

Table S7. Brief information of the selected six CSN sites

Site ID	13089002	170310076	261630001	060658001	360050083	420030008
Metro Area	Atlanta	Chicago	Detroit	Los Angeles	New York	Pittsburgh
County	DaKalb, GA	Cook, IL	Wayne, MI	Riverside, CA	Bronx, NY	Allegheny, PA
City	Decatur	Chicago	Allen Park	Rubidoux	Bronx	Pittsburgh
Elevation	308 m	188 m	182 m	248 m	24 m	312 m
Latitude	33.688	41.751	42.229	34.000	40.865	40.466
Longitude	-84.290	-87.714	-83.208	117.417	-73.881	-79.961
Setting	suburban	suburban	suburban	suburban	urban	urban

Table S8. (a) Average daily a priori emissions estimates within a radius of approximate 54-km distance of Atlanta site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.12	0.22	0.00	0.00	1.21	0.01	0.15
AIRCRAFT	14.94	0.99	4.01	1.25	0.00	15.77	2.64	1.17
BIOGENIC	1.67	36.94	0.00	0.00	0.00	6.91	0.00	0.00
COALCMB	100.03	2.16	6.27	448.63	0.00	15.12	0.06	1.25
DIESELCMB	0.02	0.01	0.00	0.00	0.00	0.03	0.00	0.00
DUST	0.00	0.00	16.14	0.00	0.00	0.00	0.08	1.05
FUELOILCMB	4.49	0.05	0.45	20.60	0.03	0.86	0.04	0.03
LIVESTOCK	0.00	0.00	0.00	0.00	2.47	0.00	0.00	0.00
LPGCMB	1.49	0.04	0.00	0.08	0.00	0.23	0.00	0.00
LWASTEBURN	0.74	3.87	4.70	0.13	0.00	18.20	0.17	0.22
METALPRODUCT	0.03	0.00	0.00	0.00	0.00	0.02	0.00	0.00
MEATCOOKING	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	5.16	0.38	1.15	5.01	0.50	0.85	0.02	0.35
NAGASCMB	47.05	1.28	3.51	1.02	0.00	34.46	0.00	2.11
NRDIESEL	31.02	1.28	3.03	3.99	0.02	17.07	2.24	0.68
NRFUELOIL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
NRGASOLINE	3.32	24.48	0.85	0.11	0.01	441.94	0.07	0.67
NRLPG	7.64	1.89	0.03	0.01	0.01	29.73	0.00	0.03
NRNAGAS	0.95	0.01	0.00	0.00	0.00	3.76	0.00	0.00
NROTHERS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OPENFIRE	0.72	1.75	2.43	0.00	0.00	24.19	0.10	1.63
ORDIESEL	120.82	1.94	3.32	4.15	0.25	28.78	1.94	0.98
ORGASOLINE	172.37	171.61	1.76	3.77	11.36	2790.59	0.20	0.50
OTHERCMB	0.19	0.03	0.00	0.00	0.00	0.11	0.00	0.00
OTHERS	1.00	9.11	0.89	0.09	0.14	3.42	0.02	0.04
PRESCRBURN	0.67	0.97	2.60	0.18	0.14	31.14	0.42	2.00
RAILROAD	15.80	0.23	0.37	0.93	0.00	1.88	0.27	0.08
SEASALT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SOLVENT	0.11	299.31	0.00	0.00	0.00	0.08	0.00	0.00
WILDFIRE	0.03	0.04	0.11	0.01	0.01	1.29	0.02	0.08
WOODFUEL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
WOODSTOVE	1.00	52.42	25.09	0.35	0.00	93.21	2.70	14.19

Table S8. (b) Average daily a priori emissions estimates within a radius of approximate 54-km distance of Chicago site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
AIRCRAFT	12.65	2.36	0.12	1.18	0.00	22.14	0.08	0.03
BIOGENIC	0.77	0.73	0.00	0.00	0.00	0.30	0.00	0.00
COALCMB	117.21	2.00	5.99	205.23	0.06	23.99	0.06	1.20
DIESELCMB	0.66	0.14	0.05	0.04	0.00	0.26	0.04	0.01
DUST	0.00	0.00	16.31	0.00	0.00	0.00	0.08	1.05
FUELOILCMB	26.69	0.95	3.04	92.33	0.46	3.39	0.39	0.21
LIVESTOCK	0.00	0.00	0.00	0.00	0.99	0.00	0.00	0.00
LPGCMB	1.28	0.06	0.29	0.06	0.00	0.27	0.00	0.18
LWASTEBURN	1.85	4.66	6.41	0.01	0.00	61.56	0.25	4.11
METALPRODUCT	25.88	18.73	47.50	42.90	1.54	962.36	0.32	7.27
MEATCOOKING	0.02	1.14	5.55	0.02	0.00	2.33	0.07	5.39
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	11.05	1.00	3.85	15.85	0.00	6.32	0.04	0.26
NAGASCMB	271.19	6.85	21.78	137.60	6.35	71.87	0.00	12.93
NRDIESEL	77.13	1.95	5.00	7.01	0.05	25.02	3.70	1.12
NRFUELOIL	10.66	0.29	0.43	5.73	0.00	1.40	0.04	0.05
NRGASOLINE	12.06	77.05	2.77	0.18	0.04	681.29	0.22	2.18
NRLPG	19.05	4.76	0.09	0.02	0.00	74.61	0.00	0.08
NRNAGAS	2.07	0.03	0.01	0.00	0.00	8.23	0.00	0.01
NROTHERS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OPENFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ORDIESEL	89.53	2.19	2.63	2.45	0.18	34.06	1.58	0.79
ORGASOLINE	276.49	211.64	2.35	5.11	16.44	4991.52	0.24	0.59
OTHERCMB	80.83	4.04	7.42	88.79	4.46	152.98	0.01	3.87
OTHERS	17.09	36.46	16.82	133.36	2.43	63.98	0.17	2.56
PRESCRBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
RAILROAD	37.00	0.52	0.86	2.05	0.00	4.27	0.64	0.19
SEASALT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SOLVENT	3.32	438.88	3.38	30.87	0.11	0.68	0.08	2.50
WILDFIRE	0.00	0.00	0.00	0.00	0.00	0.03	0.00	0.00
WOODFUEL	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
WOODSTOVE	2.50	55.82	25.30	0.38	0.00	185.42	2.73	14.31

Table S8 (c) Average daily a priori emissions estimates within a radius of approximate 54-km distance of Detroit site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
AIRCRAFT	4.65	0.98	0.09	0.44	0.00	11.69	0.06	0.03
BIOGENIC	0.74	1.21	0.00	0.00	0.00	0.55	0.00	0.00
COALCMB	156.40	1.48	13.28	443.41	0.01	17.82	0.13	2.66
DIESELCMB	2.01	0.11	0.07	0.21	0.00	0.38	0.05	0.02
DUST	0.00	0.00	11.96	0.00	0.40	0.00	0.06	0.81
FUELOILCMB	129.22	0.12	0.97	369.65	0.13	1.39	0.13	0.07
LIVESTOCK	0.00	0.00	0.00	0.00	8.44	0.00	0.00	0.00
LPGCMB	1.05	0.04	0.06	0.03	0.00	0.24	0.00	0.03
LWASTEBURN	2.18	5.53	7.58	0.01	0.00	72.18	0.30	4.80
METALPRODUCT	3.53	1.37	2.61	3.21	0.00	53.41	0.02	0.43
MEATCOOKING	0.00	0.53	2.85	0.00	0.00	1.17	0.03	2.74
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	12.14	8.84	3.10	18.56	0.17	5.05	0.05	0.14
NAGASCMB	134.96	2.80	9.47	54.77	0.56	36.39	0.00	5.68
NRDIESEL	48.80	1.49	3.21	4.34	0.03	17.65	2.38	0.72
NRFUELOIL	4.99	0.14	0.24	3.45	0.00	0.65	0.02	0.03
NRGASOLINE	8.89	56.77	1.73	0.55	0.02	588.42	0.14	1.36
NRLPG	12.94	3.24	0.06	0.01	0.00	50.91	0.00	0.05
NRNAGAS	1.30	0.02	0.01	0.00	0.00	5.19	0.00	0.01
NROTHERS	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00
OPENFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ORDIESEL	141.29	3.45	4.31	3.75	0.29	50.76	2.65	1.27
ORGASOLINE	209.87	190.27	1.88	4.05	10.94	3875.01	0.23	0.58
OTHERCMB	13.33	0.57	0.57	12.92	0.01	8.38	0.00	0.34
OTHERS	9.12	11.23	1.70	3.87	3.32	10.09	0.03	0.17
PRESCRBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
RAILROAD	6.29	0.10	0.16	0.35	0.00	1.04	0.12	0.04
SEASALT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SOLVENT	0.50	285.68	0.54	0.06	0.00	0.34	0.03	0.28
WILDFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
WOODFUEL	0.01	0.00	0.08	0.00	0.00	0.10	0.01	0.03
WOODSTOVE	2.74	53.15	28.16	0.41	0.05	202.49	3.03	15.93

Table S8 (d) Average daily a priori emissions estimates within a radius of approximate 54-km distance of Los Angeles site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
AIRCRAFT	1.33	0.28	0.03	0.13	0.00	3.73	0.02	0.01
BIOGENIC	2.76	63.72	0.00	0.00	0.00	14.62	0.00	0.00
COALCMB	0.04	0.05	0.01	0.06	0.00	1.05	0.00	0.00
DIESELCMB	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
DUST	0.00	0.00	12.87	0.00	0.00	0.00	0.05	0.76
FUELOILCMB	17.45	0.95	4.43	2.96	0.00	1.44	0.37	0.18
LIVESTOCK	0.00	0.00	0.00	0.00	9.26	0.00	0.00	0.00
LPGCMB	1.76	0.11	0.97	0.12	0.00	0.39	0.00	0.58
LWASTEBURN	1.76	4.46	6.87	0.00	0.00	63.61	0.27	4.60
METALPRODUCT	0.08	0.06	0.16	0.00	0.00	0.11	0.00	0.03
MEATCOOKING	0.00	1.06	9.79	0.00	0.00	0.00	0.12	9.53
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	12.50	0.91	1.61	0.99	0.00	6.16	0.03	0.12
NAGASCMB	55.29	6.76	6.91	0.60	0.04	16.99	0.00	4.14
NRDIESEL	42.09	1.69	3.40	0.26	0.06	22.71	2.52	0.76
NRFUELOIL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
NRGASOLINE	6.30	44.86	1.38	0.03	0.03	726.21	0.11	1.08
NRLPG	8.05	2.01	0.04	0.01	0.00	31.57	0.00	0.03
NRNAGAS	0.95	0.01	0.00	0.00	0.00	3.77	0.00	0.00
NROTHERS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OPENFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
ORDIESEL	42.74	1.15	1.35	1.59	0.09	15.28	0.78	0.41
ORGASOLINE	140.01	146.88	1.46	3.19	10.01	2119.82	0.15	0.38
OTHERCMB	0.99	0.07	0.12	0.10	0.00	0.32	0.03	0.06
OTHERS	0.26	8.70	4.70	0.05	2.34	1.14	0.17	2.93
PRESCRBURN	0.00	0.03	0.26	0.00	0.00	1.33	0.04	0.20
RAILROAD	24.46	0.33	0.56	0.07	0.00	2.71	0.41	0.13
SEASALT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SOLVENT	0.50	156.19	0.04	0.01	0.00	0.08	0.00	0.03
WILDFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
WOODFUEL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
WOODSTOVE	2.26	7.26	23.33	0.30	0.00	175.67	2.51	13.19

Table S8 (e) Average daily a priori emissions estimates within a radius of approximate 54-km distance of New York site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
AIRCRAFT	16.07	3.06	0.17	1.45	0.00	29.27	0.11	0.05
BIOGENIC	0.36	1.47	0.00	0.00	0.00	0.57	0.00	0.00
COALCMB	32.62	1.95	11.71	58.40	0.57	3.11	0.12	2.34
DIESELCMB	2.21	0.04	0.05	0.15	0.00	0.32	0.04	0.01
DUST	0.00	0.00	10.67	0.00	0.00	0.00	0.07	0.86
FUELOILCMB	250.74	4.65	15.54	405.03	4.37	34.29	1.78	1.17
LIVESTOCK	0.00	0.00	0.00	0.00	0.93	0.00	0.00	0.00
LPGCMB	5.52	0.11	0.18	0.80	0.00	0.86	0.00	0.11
LWASTEBURN	0.08	0.18	0.24	0.01	0.01	1.41	0.01	0.07
METALPRODUCT	0.00	0.04	0.00	0.00	0.00	0.00	0.00	0.00
MEATCOOKING	0.00	1.95	11.31	0.00	0.00	3.59	0.14	11.00
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	0.07	0.04	0.12	0.02	0.00	0.20	0.00	0.01
NAGASCMB	237.89	19.58	13.02	171.52	1.20	68.06	0.00	7.81
NRDIESEL	88.45	3.56	8.08	11.73	0.11	46.11	5.99	1.81
NRFUELOIL	1.63	0.04	0.07	0.95	0.00	0.21	0.01	0.01
NRGASOLINE	17.74	109.05	3.22	0.25	0.06	1528.22	0.26	2.53
NRLPG	28.21	6.97	0.13	0.03	0.00	109.07	0.00	0.11
NRNAGAS	3.87	0.06	0.02	0.00	0.00	15.29	0.00	0.02
NROTHERS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OPENFIRE	3.28	8.01	11.14	0.00	0.00	110.71	0.45	7.46
ORDIESEL	196.52	4.79	5.72	6.95	0.42	72.70	3.36	1.69
ORGASOLINE	442.50	369.50	3.87	8.15	25.09	7863.88	0.43	1.06
OTHERCMB	7.83	1.86	0.51	1.72	0.02	3.72	0.16	0.19
OTHERS	22.52	167.42	4.88	11.19	19.20	12.81	0.10	0.27
PRESCRBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
RAILROAD	3.91	0.09	0.15	0.25	0.00	1.73	0.11	0.03
SEASALT	0.00	0.00	0.57	0.00	0.00	0.00	0.00	0.00
SOLVENT	1.86	498.77	0.67	0.42	0.01	0.45	0.01	0.61
WILDFIRE	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
WOODFUEL	0.37	0.01	0.24	0.02	0.00	10.21	0.03	0.09
WOODSTOVE	1.10	58.12	12.12	0.17	0.12	101.17	1.31	6.86

Table S8 (f) Average daily a priori emissions estimates within a radius of approximate 54-km distance of Pittsburgh site (tons/day)

Category	NO_x	VOC	$PM_{2.5}$	SO_2	NH_3	CO	PEC	POA
AGRIBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
AIRCRAFT	3.21	0.73	0.08	0.31	0.00	9.26	0.05	0.02
BIOGENIC	0.55	2.54	0.00	0.00	0.00	0.92	0.00	0.00
COALCMB	437.51	2.69	59.71	1338.95	0.02	34.61	0.60	11.94
DIESELCMB	0.37	0.02	0.01	0.03	0.00	0.11	0.01	0.00
DUST	0.00	0.00	9.08	0.00	0.00	0.00	0.06	0.74
FUELOILCMB	153.42	0.43	0.73	558.02	0.02	1.71	0.12	0.05
LIVESTOCK	0.00	0.00	0.00	0.00	2.51	0.00	0.00	0.00
LPGCMB	0.38	0.02	0.02	0.00	0.00	0.07	0.00	0.01
LWASTEBURN	2.53	6.44	8.22	0.07	0.02	75.51	0.32	4.70
METALPRODUCT	31.76	3.95	25.38	25.82	0.40	227.46	0.04	4.88
MEATCOOKING	0.00	0.29	1.67	0.00	0.00	0.69	0.02	1.62
MEXCMB_M	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
MINERALPRODUCT	16.24	0.53	3.09	11.83	0.00	1.99	0.06	0.18
NAGASCMB	93.51	1.44	3.49	145.92	0.21	24.51	0.00	2.09
NRDIESEL	59.35	1.13	3.19	4.20	0.02	14.58	2.36	0.71
NRFUELOIL	10.87	0.30	0.45	6.00	0.00	1.43	0.04	0.05
NRGASOLINE	2.37	22.44	0.61	0.07	0.01	266.45	0.05	0.48
NRLPG	5.72	1.43	0.03	0.01	0.00	22.43	0.00	0.02
NRNAGAS	0.66	0.01	0.00	0.00	0.00	2.64	0.00	0.00
NROTHERS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OPENFIRE	0.07	0.18	0.25	0.00	0.00	2.48	0.01	0.17
ORDIESEL	51.34	1.23	1.55	1.89	0.11	18.48	0.91	0.46
ORGASOLINE	100.94	85.78	0.81	1.72	5.25	1765.17	0.09	0.23
OTHERCMB	26.57	0.40	4.74	25.81	0.08	11.20	0.03	2.34
OTHERS	2.88	17.80	1.85	1.33	3.75	12.72	0.02	0.24
PRESCRBURN	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
RAILROAD	14.32	0.21	0.33	0.78	0.00	1.67	0.25	0.07
SEASALT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
SOLVENT	0.19	100.54	0.19	0.07	0.00	0.12	0.02	0.09
WILDFIRE	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
WOODFUEL	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
WOODSTOVE	1.30	24.48	13.76	0.20	0.00	101.76	1.48	7.78

Table S9 (a) Calculated R_j for each measurement day at Atlanta site

Category	1/4/04	1/7/04	1/10/04	1/13/04	1/16/04	1/19/04	1/22/04
AGRIBURN	0.100	0.733	0.307	0.171	0.404	0.762	0.313
AIRCRAFT	0.914	0.894	0.974	0.922	0.812	1.002	0.915
BIOGENIC	1.001	0.996	1.002	0.980	0.985	1.002	0.999
COALCMB	0.921	0.936	0.947	0.903	0.896	0.856	0.945
DIESELCMB	1.000	1.000	1.000	1.000	1.000	1.000	1.000
DUST	0.100	0.100	0.100	0.100	0.100	0.100	0.100
FUELOILCMB	0.786	0.875	0.967	0.769	0.756	0.888	0.887
LIVESTOCK	0.998	0.992	1.004	0.957	0.944	1.009	0.990
LPGCMB	1.000	1.000	1.000	1.000	0.999	1.000	1.000
LWASTEBURN	0.100	0.100	0.100	0.100	0.100	0.100	0.100
METALPRODUCT	0.945	0.960	0.752	0.504	0.886	0.684	0.765
MEATCOOKING	1.000	0.984	0.923	0.921	0.940	0.884	0.940
MEXCMB_M	1.000	1.000	1.000	0.998	0.999	1.000	1.000
MINERALPRODUCT	0.817	0.936	0.862	0.753	0.877	0.834	0.818
NAGASCMB	0.454	0.447	0.259	0.264	0.336	0.403	0.259
NRDIESEL	0.984	0.973	0.996	0.987	0.956	1.002	0.976
NRFUELOIL	0.998	1.000	1.000	0.998	0.998	0.999	0.999
NRGASOLINE	0.994	0.997	1.001	0.994	0.977	0.998	0.996
NRLPG	1.000	1.001	1.000	1.000	1.001	1.000	1.000
NRNAGAS	1.000	1.000	1.000	1.000	1.000	1.000	1.000
NROTHERS	1.000	1.000	1.000	1.000	0.999	1.000	1.000
OPENFIRE	0.100	0.257	0.100	0.143	0.100	0.141	0.102
ORDIESEL	0.906	0.957	0.957	0.863	0.840	0.962	0.921
ORGASOLINE	0.641	0.734	0.901	0.482	0.444	0.753	0.753
OTHERCMB	0.468	0.992	0.976	0.950	0.974	0.917	0.983
OTHERS	0.386	0.671	0.453	0.352	0.404	0.384	0.394
PRESCRBURN	0.100	1.080	0.432	0.305	0.347	0.763	0.434
RAILROAD	0.997	0.999	0.999	1.000	1.001	1.001	0.996
SEASALT	0.982	0.994	0.981	0.976	0.993	0.997	0.986
SOLVENT	0.941	0.995	0.987	0.901	0.867	0.844	0.984
WILDFIRE	0.439	0.949	0.587	0.703	0.647	0.924	0.636
WOODFUEL	0.756	0.997	0.955	0.974	0.935	0.746	0.972
WOODSTOVE	0.100	0.100	0.100	0.100	0.100	0.100	0.100

Table S9. (b) Calculated R_j for each measurement day at Chicago site

Category	1/4/04	1/19/04	1/25/04	1/28/04
AGRIBURN	0.994	0.972	1.000	0.991
AIRCRAFT	1.000	0.998	0.999	0.999
BIOGENIC	1.000	0.999	1.000	1.000
COALCMB	0.954	0.927	0.960	0.921
DIESELCMB	1.000	1.000	1.000	1.000
DUST	0.100	0.100	0.100	0.100
FUELOILCMB	0.831	0.606	0.719	0.685
LIVESTOCK	0.998	0.982	0.958	0.964
LPGCMB	1.000	0.999	0.999	0.999
LWASTEBURN	0.100	0.100	0.229	0.154
METALPRODUCT	0.112	0.227	0.111	0.247
MEATCOOKING	0.439	0.536	0.537	0.549
MEXCMB_M	1.000	0.999	1.000	1.000
MINERALPRODUCT	0.681	0.758	0.750	0.751
NAGASCMB	0.182	0.330	0.244	0.267
NRDIESEL	0.978	0.966	0.970	0.951
NRFUELOIL	0.989	0.972	0.971	0.980
NRGASOLINE	0.996	0.986	0.989	0.991
NRLPG	1.000	0.999	1.000	0.999
NRNAGAS	1.000	0.999	1.000	1.000
NROTHERS	1.000	0.999	1.000	1.000
OPENFIRE	1.000	0.982	1.000	0.992
ORDIESEL	0.970	0.959	0.968	0.939
ORGASOLINE	0.763	0.611	0.731	0.607
OTHERCMB	0.476	0.644	0.533	0.708
OTHERS	0.149	0.168	0.169	0.121
PRESCRBURN	1.000	0.965	1.000	0.978
RAILROAD	0.996	0.995	0.993	0.987
SEASALT	0.997	0.999	1.000	1.000
SOLVENT	0.601	0.637	0.320	0.384
WILDFIRE	0.996	0.962	0.997	0.925
WOODFUEL	0.945	0.873	0.983	0.962
WOODSTOVE	0.100	0.100	0.129	0.100

Table S9. (c) Calculated R_j for each measurement day at Detroit site

Category	1/4/04	1/7/04	1/10/04	1/13/04	1/16/04	1/19/04	1/22/04	1/28/04
AGRIBURN	0.972	0.975	1.000	0.986	1.000	0.951	0.522	0.949
AIRCRAFT	0.988	0.999	1.000	0.997	0.992	0.988	0.995	1.000
BIOGENIC	0.917	1.000	1.000	0.995	1.000	0.988	0.996	1.002
COALCMB	0.979	0.967	0.978	0.948	0.985	0.928	0.963	0.974
DIESELCMB	0.998	1.000	1.000	0.999	0.999	0.998	0.998	1.000
DUST	0.100	0.100	0.100	0.100	0.100	0.100	0.100	0.100
FUELOILCMB	0.916	0.709	0.842	0.842	0.744	0.382	0.744	0.908
LIVESTOCK	0.993	0.983	0.993	1.011	1.000	0.938	0.966	1.009
LPGCMB	0.996	0.995	1.000	1.000	0.999	0.991	0.997	1.000
LWASTEBURN	0.100	0.100	0.264	0.100	0.103	0.100	0.100	0.100
METALPRODUCT	0.772	0.633	0.786	0.525	0.719	0.733	0.548	0.452
MEATCOOKING	0.128	0.916	1.014	0.429	0.487	0.286	0.280	0.325
MEXCMB_M	1.000	1.000	1.000	1.000	1.000	0.997	0.997	1.000
MINERALPRODUCT	0.839	0.883	0.920	0.803	0.943	0.868	0.865	0.854
NAGASCMB	0.302	0.582	0.603	0.321	0.345	0.346	0.170	0.202
NRDIESEL	0.826	0.992	1.006	0.900	0.803	0.819	0.938	1.004
NRFUELOIL	0.989	0.990	0.991	0.995	0.972	0.908	0.987	0.996
NRGASOLINE	0.733	1.000	1.003	0.978	0.980	0.880	0.987	1.006
NRLPG	0.993	0.999	1.000	0.999	0.999	0.990	0.997	1.000
NRNAGAS	0.999	0.999	1.000	1.000	1.000	0.997	0.997	1.000
NROTHERS	0.999	1.000	1.000	0.999	1.000	0.997	0.999	0.999
OPENFIRE	0.999	0.999	1.000	0.994	1.000	0.952	0.950	0.668
ORDIESEL	0.796	0.999	1.003	0.789	0.787	0.723	0.902	0.990
ORGASOLINE	0.817	0.867	0.876	0.496	0.638	0.541	0.782	0.936
OTHERCMB	0.882	0.944	0.975	0.872	0.959	0.808	0.864	0.845
OTHERS	0.458	0.519	0.770	0.343	0.490	0.378	0.346	0.374
PRESCRBURN	0.993	0.980	1.000	0.989	1.000	0.922	0.891	0.993
RAILROAD	0.984	0.997	1.001	0.995	0.989	0.971	0.995	1.000
SEASALT	1.000	1.000	1.000	0.999	1.000	0.999	0.998	0.992
SOLVENT	0.733	0.737	0.944	0.661	0.590	0.724	0.687	0.921
WILDFIRE	0.972	0.935	1.000	0.942	1.000	0.915	0.728	0.846
WOODFUEL	0.913	0.963	0.993	0.898	0.979	0.836	0.939	0.931
WOODSTOVE	0.100	0.100	0.100	0.100	0.100	0.100	0.100	0.100

Table S9. (d) Calculated R_j for each measurement day at Los Angeles site

Category	1/7/04	1/10/04	1/13/04	1/19/04	1/22/04	1/25/04	1/28/04
AGRIBURN	0.834	0.993	0.614	0.652	0.764	0.515	0.381
AIRCRAFT	0.997	0.998	1.000	1.001	0.997	1.013	1.000
BIOGENIC	0.981	1.013	0.975	1.032	0.808	0.999	1.006
COALCMB	0.998	0.976	0.998	0.998	0.986	0.999	0.999
DIESELCMB	1.000	1.000	1.000	1.000	0.999	1.004	1.000
DUST	0.100	0.174	0.116	0.100	0.606	0.100	0.100
FUELOILCMB	0.189	0.284	0.648	0.622	0.398	0.644	0.675
LIVESTOCK	0.997	0.881	1.004	1.005	0.801	1.011	1.003
LPGCMB	0.875	0.929	0.989	1.008	0.954	1.009	1.001
LWASTEBURN	0.100	0.100	0.100	0.100	0.100	0.100	0.100
METALPRODUCT	0.955	0.990	0.985	0.931	0.983	0.970	0.961
MEATCOOKING	0.190	0.259	0.316	0.312	0.164	0.415	0.255
MEXCMB_M	0.956	0.998	0.993	1.001	0.998	1.010	0.999
MINERALPRODUCT	0.715	0.853	0.929	0.611	0.888	0.514	0.709
NAGASCMB	0.206	0.244	0.362	0.289	0.347	0.256	0.209
NRDIESEL	0.882	0.965	1.007	0.986	0.944	1.035	1.015
NRFUELOIL	0.990	1.000	1.000	1.002	0.998	1.019	0.996
NRGASOLINE	0.981	0.986	0.997	1.024	0.943	1.012	1.005
NRLPG	0.997	1.000	1.000	1.000	1.006	1.011	1.000
NRNAGAS	0.999	1.000	1.000	1.000	0.999	1.010	0.999
NROTHERS	1.000	1.000	1.000	1.000	1.001	0.991	1.001
OPENFIRE	0.992	1.000	1.000	1.001	0.972	1.262	0.988
ORDIESEL	0.867	0.936	0.991	0.960	0.965	0.976	0.996
ORGASOLINE	0.554	0.756	0.880	0.617	0.674	0.670	0.808
OTHERCMB	0.972	0.998	0.999	0.915	0.989	0.834	0.825
OTHERS	0.408	0.522	0.724	0.325	0.375	0.342	0.385
PRESCRBURN	0.862	0.730	4.070	0.825	0.542	1.100	0.789
RAILROAD	0.974	0.977	1.003	0.998	0.981	1.016	1.003
SEASALT	0.985	0.994	0.993	1.000	0.994	0.978	0.943
SOLVENT	0.997	1.011	1.004	0.879	0.993	0.897	0.940
WILDFIRE	0.985	1.000	0.999	1.002	0.952	4.653	0.979
WOODFUEL	0.999	1.000	1.000	0.962	0.986	0.980	0.990
WOODSTOVE	0.100	0.100	0.100	0.100	0.100	0.100	0.100

Table S9. (e) Calculated R_j for each measurement day at New York site

Category	1/4/04	1/7/04	1/10/04	1/13/04	1/16/04	1/19/04	1/22/04	1/25/04	1/28/04
AGRIBURN	0.557	0.995	1.000	0.637	1.000	0.954	0.964	1.000	0.947
AIRCRAFT	1.000	1.000	1.000	0.999	0.998	0.993	1.000	1.000	1.000
BIOGENIC	1.004	0.999	1.000	0.961	0.995	0.884	0.997	0.997	0.999
COALCMB	0.752	0.982	0.998	0.779	0.895	0.899	0.801	0.879	0.959
DIESELCMB	1.000	1.000	1.000	1.000	1.000	0.999	1.000	1.000	1.000
DUST	0.100	0.108	0.424	0.100	0.100	0.100	0.100	0.100	0.100
FUELOILCMB	0.831	0.900	1.413	0.646	0.502	0.503	0.646	0.506	0.803
LIVESTOCK	0.996	0.996	1.001	0.966	0.988	0.978	0.993	0.982	0.995
LPGCMB	1.000	1.000	1.000	0.999	0.998	0.990	1.000	0.999	1.000
LWASTEBURN	0.112	0.100	0.579	0.100	0.345	0.114	0.218	0.197	0.100
METALPRODUCT	0.817	0.916	1.000	0.728	0.946	0.935	0.865	0.978	0.850
MEATCOOKING	0.744	0.549	0.983	0.299	0.174	0.100	0.679	0.186	0.310
MEXCMB_M	1.000	1.000	1.000	0.999	1.000	0.999	1.000	1.000	1.000
MINERALPRODUCT	0.789	0.971	0.999	0.751	0.976	0.938	0.885	0.979	0.949
NAGASCMB	0.471	0.492	0.832	0.339	0.691	0.319	0.488	0.300	0.258
NRDIESEL	1.016	1.022	1.015	0.981	0.901	0.708	1.018	0.975	1.019
NRFUELOIL	0.999	1.000	1.001	0.999	0.999	0.996	0.998	0.999	1.000
NRGASOLINE	1.003	0.998	1.003	0.960	0.908	0.673	0.995	0.951	1.001
NRLPG	1.000	1.000	1.000	1.001	0.995	0.978	1.001	1.000	1.000
NRNAGAS	1.000	1.000	1.000	1.000	0.999	0.996	1.000	1.000	1.000
NROTHERS	1.000	1.000	1.000	1.000	1.000	0.999	1.000	1.000	1.000
OPENFIRE	0.100	0.100	0.172	0.100	0.100	0.100	0.143	0.100	0.100
ORDIESEL	0.949	1.016	1.025	0.919	0.909	0.760	0.959	0.940	0.990
ORGASOLINE	0.694	1.061	1.068	0.566	0.722	0.672	0.630	0.702	0.890
OTHERCMB	0.896	0.979	1.001	0.919	0.950	0.898	0.907	0.950	0.947
OTHERS	0.326	0.647	0.959	0.285	0.442	0.375	0.246	0.317	0.390
PRESCRBURN	0.350	0.998	1.000	0.572	1.000	0.954	0.310	1.000	0.983
RAILROAD	1.001	1.000	1.001	1.012	0.999	0.998	1.001	0.997	1.000
SEASALT	0.997	0.975	1.003	0.973	0.999	0.995	0.996	1.000	0.991
SOLVENT	0.824	0.986	0.998	0.819	0.962	0.771	0.667	0.960	0.984
WILDFIRE	0.858	0.943	1.000	0.863	1.000	0.969	0.969	1.000	0.987
WOODFUEL	0.847	0.989	1.004	0.903	0.838	0.868	0.949	0.854	0.966
WOODSTOVE	0.100	0.153	0.282	0.100	0.100	0.100	0.110	0.100	0.100

Table S9. (f) Calculated R_j for each measurement day at Pittsburgh site

Category	1/4/04	1/7/04	1/10/04	1/13/04	1/19/04	1/22/04	1/25/04	1/28/04
AGRIBURN	0.645	0.988	1.000	0.765	0.981	0.724	1.000	0.991
AIRCRAFT	0.891	1.001	0.998	0.999	0.998	0.998	0.998	1.000
BIOGENIC	0.384	1.001	0.999	0.978	0.990	1.000	1.000	1.001
COALCMB	0.924	0.968	0.902	0.811	0.920	0.878	0.787	0.939
DIESELCMB	0.991	1.000	1.000	1.000	1.000	1.000	1.000	1.000
DUST	0.100	0.100	0.100	0.100	0.100	0.100	0.100	0.100
FUELOILCMB	0.872	1.056	0.514	0.923	0.857	0.885	0.822	0.988
LIVESTOCK	1.000	1.031	0.995	0.983	0.992	1.015	0.970	1.027
LPGCMB	0.978	1.000	1.000	1.000	1.000	1.000	1.000	1.000
LWASTEBURN	0.100	0.116	0.100	0.100	0.100	0.100	0.100	0.100
METALPRODUCT	0.622	0.787	0.788	0.472	0.334	0.435	0.494	0.458
MEATCOOKING	0.100	0.976	0.928	0.649	0.353	0.610	0.671	0.673
MEXCMB_M	1.000	1.000	1.000	0.999	1.000	1.000	1.000	1.000
MINERALPRODUCT	0.816	0.940	0.893	0.603	0.784	0.797	0.681	0.867
NAGASCMB	0.230	0.825	0.678	0.441	0.265	0.394	0.491	0.398
NRDIESEL	0.352	1.016	0.967	0.974	0.955	0.960	0.949	1.011
NRFUELOIL	0.792	1.002	0.947	0.995	0.976	0.985	0.967	0.996
NRGASOLINE	0.373	1.002	0.989	0.987	0.979	0.997	0.990	1.004
NRLPG	0.962	1.000	1.000	1.000	0.999	1.000	1.000	1.000
NRNAGAS	0.995	1.000	1.000	1.000	1.000	1.000	1.000	1.000
NROTHERS	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
OPENFIRE	0.739	0.816	0.771	0.596	0.269	0.140	0.514	0.367
ORDIESEL	0.629	1.011	0.983	0.945	0.963	0.978	0.946	1.002
ORGASOLINE	0.731	1.053	0.904	0.763	0.871	0.925	0.758	1.027
OTHERCMB	0.801	0.985	0.970	0.870	0.786	0.876	0.954	0.860
OTHERS	0.313	1.045	0.754	0.417	0.464	0.467	0.437	0.546
PRESCRBURN	0.654	1.022	1.000	0.908	0.988	0.896	0.953	0.999
RAILROAD	0.885	1.002	0.999	0.997	0.996	0.999	0.996	1.001
SEASALT	1.000	1.000	1.000	0.996	0.995	0.998	1.000	0.997
SOLVENT	0.768	1.029	0.978	0.833	0.971	0.844	0.937	0.966
WILDFIRE	0.542	0.975	1.000	0.848	0.949	0.856	0.998	0.855
WOODFUEL	0.937	0.998	0.999	0.962	0.986	0.974	0.980	0.979
WOODSTOVE	0.100	0.296	0.287	0.124	0.100	0.100	0.100	0.100

Table S10. (a) Initial and refined source impacts in percentage contributions at Atlanta site

Category	1/4/04		1/7/04		1/10/04		1/13/04		1/16/04		1/19/04		1/22/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	2.5	0.5	0.0	0.0	0.1	0.0	0.3	0.1	0.1	0.1	0.1	0.1	0.1	0.0
AIRCRAFT	11.7	22.0	14.0	21.6	7.3	10.0	11.1	19.9	16.0	27.4	3.7	5.8	12.2	20.8
BIOGENIC	8.9	18.4	1.5	2.6	2.0	2.9	5.9	11.3	3.0	6.2	1.4	2.3	3.7	6.9
COALCMB	6.6	12.5	11.3	18.3	30.6	40.8	8.4	14.7	6.9	13.0	23.7	32.0	6.2	11.0
DIESELCMB	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
DUST	15.4	3.2	11.8	2.0	7.9	1.1	13.9	2.7	14.8	3.1	8.9	1.4	15.0	2.8
FUELOILCMB	6.1	9.9	1.4	2.2	7.7	10.5	3.2	4.8	2.1	3.4	7.9	11.1	1.9	3.2
LIVESTOCK	0.6	1.2	4.5	7.8	2.9	4.1	4.0	7.5	2.6	5.1	4.1	6.5	6.0	11.1
LPGCMB	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.1	0.2	0.3	0.0	0.1
LWASTEBURN	4.2	0.9	3.9	0.7	2.6	0.4	3.9	0.8	5.1	1.1	2.4	0.4	4.4	0.8
METALPRODUCT	0.2	0.4	0.2	0.3	0.8	0.9	1.3	1.3	0.3	0.5	1.5	1.6	0.4	0.6
MEATCOOKING	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.2	0.1	0.2	0.3	0.4	0.1	0.1
MEXCMB_M	0.0	0.0	0.0	0.0	0.1	0.2	0.1	0.1	0.0	0.0	0.1	0.1	0.0	0.0
MINERALPRODUCT	0.5	0.9	1.0	1.6	1.0	1.2	1.8	2.6	0.8	1.4	1.1	1.5	1.4	2.1
NAGASCMB	2.4	2.3	3.8	3.0	2.7	1.0	3.8	1.9	3.4	2.4	7.2	4.6	4.0	1.9
NRDIESEL	1.6	3.3	3.3	5.6	0.9	1.3	2.4	4.6	3.1	6.2	2.0	3.2	3.0	5.5
NRFUELOIL	0.2	0.3	0.0	0.0	0.1	0.1	0.0	0.1	0.0	0.1	0.3	0.4	0.0	0.1
NRGASOLINE	1.1	2.2	1.2	2.0	0.9	1.2	0.9	1.8	1.3	2.7	1.5	2.4	1.0	1.9
NRLPG	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.2	0.1	0.1
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
NROTHERS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OPENFIRE	1.8	0.4	0.4	0.2	0.8	0.1	0.4	0.1	1.4	0.3	0.7	0.2	0.5	0.1
ORDIESEL	2.3	4.4	2.9	4.8	2.2	3.0	2.8	4.7	3.5	6.2	2.6	3.9	3.7	6.3
ORGASOLINE	4.5	5.9	12.6	16.0	11.6	14.7	12.4	11.6	10.2	9.5	9.0	10.7	11.1	15.5
OTHERCMB	2.1	2.0	0.1	0.1	0.2	0.3	0.2	0.4	0.2	0.4	1.3	1.9	0.2	0.3
OTHERS	3.1	2.4	2.5	2.9	2.3	1.5	2.8	1.9	1.8	1.6	3.4	2.1	2.3	1.7
PRESCRBURN	12.2	2.5	1.1	2.1	0.5	0.3	1.1	0.7	1.0	0.7	0.3	0.3	0.6	0.4
RAILROAD	0.3	0.7	0.3	0.6	0.3	0.4	0.3	0.5	0.3	0.5	0.5	0.8	0.5	1.0
SEASALT	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SOLVENT	0.2	0.5	0.7	1.2	1.1	1.5	1.1	2.0	1.4	2.5	2.2	3.0	0.9	1.6
WILDFIRE	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
WOODFUEL	0.4	0.6	0.3	0.6	0.3	0.5	0.2	0.3	0.3	0.6	0.5	0.6	0.1	0.2
WOODSTOVE	10.8	2.2	20.9	3.6	13.1	1.9	17.7	3.4	20.1	4.2	12.6	2.0	20.5	3.8

Table S10. (b) Initial and refined source impacts in percentage contributions at Chicago site

Category	1/4/04		1/19/04		1/22/04		1/25/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
AIRCRAFT	0.0	0.1	0.1	0.2	0.0	0.1	0.1	0.1
BIOGENIC	0.4	1.1	0.7	1.7	0.0	0.1	0.4	0.8
COALCMB	3.9	10.7	4.0	9.1	1.6	4.5	5.3	10.5
DIESELCMB	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.1
DUST	4.5	1.3	8.4	2.0	7.3	2.1	14.6	3.2
FUELOILCMB	3.3	7.9	3.4	4.9	1.4	2.9	3.7	5.4
LIVESTOCK	2.5	7.0	3.5	8.4	5.0	13.6	11.9	24.9
LPGCMB	0.1	0.3	0.1	0.2	0.2	0.5	0.1	0.2
LWASTEBURN	2.0	0.6	3.9	0.9	2.2	1.4	2.9	1.0
METALPRODUCT	23.0	7.4	11.5	6.3	22.6	7.2	6.6	3.5
MEATCOOKING	3.4	4.3	4.4	5.8	2.6	4.0	2.9	3.4
MEXCMB_M	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MINERALPRODUCT	1.8	3.5	1.4	2.6	1.4	2.9	1.2	1.9
NAGASCMB	16.6	8.7	13.7	11.0	15.4	10.7	10.4	6.0
NRDIESEL	2.2	6.2	3.6	8.5	1.8	4.9	3.0	6.1
NRFUELOIL	0.4	1.0	0.4	0.9	0.2	0.6	0.3	0.6
NRGASOLINE	1.0	2.8	2.6	6.1	0.9	2.5	2.0	4.2
NRLPG	0.0	0.1	0.1	0.1	0.0	0.1	0.1	0.3
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
NROTHERS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OPENFIRE	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
ORDIESEL	1.0	2.8	1.6	3.7	0.8	2.2	2.1	4.2
ORGASOLINE	5.2	11.5	7.1	10.6	7.5	15.7	8.3	10.9
OTHERCMB	8.1	11.0	2.7	4.2	8.6	13.1	2.4	3.7
OTHERS	9.3	4.0	10.1	4.1	7.8	3.8	10.1	2.6
PRESCRBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
RAILROAD	0.5	1.4	0.5	1.3	0.4	1.2	1.0	2.2
SEASALT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SOLVENT	2.2	3.7	2.1	3.2	2.1	2.0	1.7	1.4
WILDFIRE	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.2
WOODFUEL	0.1	0.3	0.2	0.5	0.0	0.1	0.1	0.2
WOODSTOVE	8.5	2.4	13.7	3.3	10.0	3.7	8.9	1.9

Table S10. (c) Initial and refined source impacts in percentage contributions at Detroit site

Category	1/4/04		1/7/04		1/10/04		1/13/04		1/16/04		1/19/04		1/22/04		1/25/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
AIRCRAFT	0.1	0.3	0.1	0.2	0.0	0.1	0.1	0.3	0.2	0.5	0.2	0.5	0.2	0.4	0.1	0.2
BIOGENIC	0.6	1.6	0.9	1.7	0.1	0.1	1.0	2.2	0.0	0.1	0.4	1.3	1.3	2.9	0.6	1.0
COALCMB	3.8	10.7	5.2	9.1	3.2	5.5	6.3	13.4	2.6	6.9	4.2	11.5	5.5	11.8	4.6	8.1
DIESELCMB	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.2	0.1	0.2	0.0	0.1
DUST	11.4	3.3	14.9	2.7	7.7	1.4	10.5	2.4	5.7	1.5	9.3	2.8	13.5	3.0	10.3	1.8
FUELOILCMB	2.6	6.8	2.4	3.1	1.7	2.5	4.0	7.6	2.3	4.6	3.2	3.6	2.7	4.5	2.3	3.7
LIVESTOCK	2.0	5.8	11.4	20.3	6.8	12.0	5.7	13.1	0.2	0.4	0.7	2.0	9.4	20.4	13.1	23.7
LPGCMB	0.1	0.3	0.2	0.4	0.1	0.1	0.1	0.3	0.1	0.2	0.1	0.3	0.2	0.5	0.1	0.2
LWASTEBURN	7.0	2.0	3.5	0.6	5.3	2.5	4.9	1.1	6.3	1.8	6.8	2.0	4.6	1.0	3.3	0.6
METALPRODUCT	2.3	5.0	3.6	4.1	2.5	3.5	2.4	2.8	2.0	4.0	1.9	4.1	1.7	2.1	4.6	3.7
MEATCOOKING	4.1	1.5	1.8	3.0	3.2	5.8	3.1	3.0	4.7	6.3	3.8	3.2	2.5	1.6	1.8	1.0
MEXCMB_M	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
MINERALPRODUCT	0.9	2.1	1.3	2.1	1.3	2.1	0.7	1.2	0.6	1.5	0.6	1.7	0.7	1.4	0.9	1.4
NAGASCMB	12.3	10.7	9.0	9.4	9.0	9.6	11.1	8.0	15.3	14.3	13.6	13.9	10.1	3.9	7.2	2.6
NRDIESEL	3.1	7.3	2.6	4.7	2.1	3.7	3.5	7.2	5.3	11.6	4.4	10.7	3.3	6.9	2.3	4.1
NRFUELOIL	0.4	1.3	0.2	0.3	0.2	0.3	0.3	0.7	0.4	1.2	0.4	1.2	0.3	0.6	0.2	0.3
NRGASOLINE	3.0	6.3	1.6	2.9	1.9	3.4	2.6	5.8	3.2	8.4	2.6	6.7	1.9	4.2	2.0	3.7
NRLPG	0.0	0.1	0.1	0.2	-0.1	-0.2	0.1	0.2	0.1	0.2	0.1	0.3	0.1	0.3	0.1	0.1
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
NROTHERS	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OPENFIRE	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
ORDIESEL	3.3	7.5	2.9	5.3	2.1	3.8	3.9	6.9	5.1	11.0	4.8	10.2	3.9	7.9	2.7	4.8
ORGASOLINE	3.4	8.0	8.8	13.7	17.6	27.4	7.2	8.0	4.4	7.7	3.7	6.0	6.3	11.2	16.0	27.0
OTHERCMB	0.6	1.5	1.3	2.2	0.7	1.2	1.4	2.7	0.5	1.4	0.7	1.7	0.8	1.6	1.1	1.7
OTHERS	3.1	4.1	6.5	6.1	6.1	8.3	4.4	3.4	2.5	3.3	2.4	2.7	4.7	3.6	6.1	4.1
PRESCRBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0
RAILROAD	0.3	0.9	0.8	1.5	0.2	0.3	0.4	0.9	0.2	0.6	0.3	0.8	0.5	1.1	0.3	0.5
SEASALT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SOLVENT	1.0	2.0	1.5	2.0	0.7	1.3	1.2	1.8	1.1	1.7	0.9	1.9	1.1	1.7	0.9	1.5
WILDFIRE	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.2	0.0	0.1
WOODFUEL	0.2	0.6	0.2	0.4	0.1	0.3	0.3	0.5	0.2	0.5	0.2	0.6	0.3	0.7	0.3	0.4
WOODSTOVE	34.2	9.8	18.8	3.4	27.5	4.9	24.4	5.5	36.9	10.0	34.5	10.2	23.7	5.3	19.2	3.5

Table S10. (d) Initial and refined source impacts in percentage contributions at Los Angeles site

Category	1/7/04		1/10/04		1/13/04		1/19/04		1/22/04		1/25/04		1/28/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	0.0	0.0	0.0	0.0	0.1	0.1	0.1	0.1	0.0	0.1	0.3	0.4	0.1	0.1
AIRCRAFT	0.0	0.1	0.0	0.1	0.0	0.1	0.1	0.2	0.0	0.1	0.4	0.9	0.2	0.5
BIOGENIC	1.4	4.9	6.7	19.3	7.0	15.9	5.0	12.1	4.3	8.6	1.2	2.7	1.8	4.6
COALCMB	0.1	0.4	1.2	3.3	0.6	1.4	0.5	1.1	2.2	5.4	0.3	0.6	0.2	0.5
DIESELCMB	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.4	0.1	0.1
DUST	10.4	3.7	12.4	6.2	13.0	3.5	7.8	1.8	11.6	17.4	8.7	2.1	9.8	2.5
FUELOILCMB	8.5	5.7	7.5	6.1	8.7	13.0	5.8	8.4	8.8	8.7	5.3	8.1	6.6	11.4
LIVESTOCK	0.4	1.3	2.0	5.1	0.8	1.9	1.3	3.0	1.9	3.9	2.9	6.8	1.2	3.0
LPGCMB	1.9	5.9	1.8	4.7	1.9	4.4	1.3	3.0	1.8	4.2	1.3	3.2	1.5	3.9
LWASTEBURN	8.2	2.9	7.5	2.1	7.4	1.7	6.1	1.4	6.8	1.7	6.3	1.5	6.7	1.7
METALPRODUCT	0.1	0.3	0.0	0.1	0.1	0.3	0.1	0.2	0.1	0.3	0.2	0.5	0.1	0.3
MEATCOOKING	17.6	11.8	11.9	8.8	11.6	8.6	13.2	9.5	10.8	4.4	13.0	12.7	12.4	8.0
MEXCMB_M	0.1	0.3	0.0	0.0	0.2	0.4	0.0	0.1	0.0	0.0	0.2	0.4	0.0	0.1
MINERALPRODUCT	2.1	5.2	1.6	3.9	1.5	3.2	1.8	2.5	2.0	4.3	1.9	2.3	2.1	3.8
NAGASCMB	9.8	7.1	8.1	5.6	7.7	6.5	8.5	5.7	7.6	6.5	8.5	5.1	9.2	4.9
NRDIESEL	3.9	12.2	1.4	3.9	3.2	7.5	2.7	6.2	3.0	7.0	2.8	6.8	4.5	11.5
NRFUELOIL	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.5	1.2	0.3	0.9
NRGASOLINE	2.0	6.9	1.3	3.6	1.1	2.5	3.5	8.4	1.0	2.4	2.0	4.7	1.6	4.2
NRLPG	0.0	-0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.3	0.7	0.2	0.4
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.4	0.1	0.2
NROTHERS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	-0.2	-0.4	0.0	-0.1
OPENFIRE	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.5	0.0	0.1
ORDIESEL	1.3	3.9	1.0	2.7	1.3	2.9	1.3	2.9	1.2	2.8	2.0	4.6	2.1	5.2
ORGASOLINE	2.3	4.6	2.4	5.2	2.2	4.4	9.2	13.2	2.8	4.7	6.9	10.9	5.9	12.1
OTHERCMB	0.1	0.4	0.0	0.1	0.0	0.1	0.2	0.5	0.1	0.2	0.7	1.3	0.5	1.1
OTHERS	6.9	9.9	5.3	8.0	5.1	8.5	8.2	6.2	8.4	7.8	12.2	9.9	9.7	9.5
PRESCRBURN	0.1	0.4	0.6	1.2	0.5	5.2	0.2	0.3	0.4	0.6	0.3	0.9	0.2	0.5
RAILROAD	0.7	2.3	0.7	2.0	0.7	1.6	0.7	1.5	0.9	2.3	0.9	2.1	1.0	2.6
SEASALT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1
SOLVENT	0.6	2.2	0.2	0.4	0.2	0.4	3.4	7.0	0.2	0.5	0.7	1.4	0.4	0.9
WILDFIRE	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	1.9	0.0	0.1
WOODFUEL	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.3	0.2	0.5	0.1	0.2
WOODSTOVE	21.4	7.5	26.3	7.5	25.1	5.8	18.9	4.4	23.7	5.8	19.6	4.6	21.4	5.4

Table S10. (e) Initial and refined source impacts in percentage contributions at New York site

Category	1/4/04		1/7/04		1/10/04		1/13/04		1/16/04		1/19/04		1/22/04		1/25/04		1/28/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	0.2	0.2	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
AIRCRAFT	0.1	0.3	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.1	0.2	0.1	0.2	0.0	0.1
BIOGENIC	2.6	5.0	0.4	0.5	0.3	0.4	2.5	5.1	0.1	0.2	0.5	1.0	1.1	2.0	0.1	0.3	0.7	1.2
COALCMB	16.2	23.3	13.3	18.7	6.6	8.9	11.5	19.2	10.3	18.9	11.9	24.3	13.6	20.4	9.0	19.9	15.6	26.1
DIESELCMB	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
DUST	7.6	1.4	9.2	1.4	6.4	3.7	12.9	2.8	5.6	1.1	5.3	1.2	8.8	1.7	7.4	1.9	5.5	1.0
FUELOILCMB	10.5	16.7	10.1	13.0	12.7	24.2	8.7	12.1	13.2	13.6	12.2	13.9	9.5	11.5	14.5	18.5	12.7	17.7
LIVESTOCK	2.6	5.0	6.4	9.1	3.4	4.6	5.5	11.4	1.2	2.4	1.8	3.9	3.8	7.0	1.2	2.9	2.2	3.8
LPGCMB	0.1	0.2	0.2	0.2	0.1	0.1	0.1	0.2	0.1	0.2	0.2	0.3	0.2	0.3	0.1	0.3	0.1	0.1
LWASTEBURN	1.9	0.4	1.5	0.2	0.3	0.2	2.6	0.6	0.3	0.2	0.8	0.2	2.0	0.8	0.2	0.1	1.1	0.2
METALPRODUCT	0.9	1.4	0.9	1.2	0.1	0.1	1.5	2.3	3.2	6.2	2.5	5.3	1.1	1.8	1.1	2.6	2.9	4.3
MEATCOOKING	6.0	8.6	5.8	4.6	8.5	11.3	6.3	4.0	9.6	3.4	8.7	2.0	6.6	8.4	13.3	6.2	7.1	3.8
MEXCMB_M	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MINERALPRODUCT	0.8	1.2	1.0	1.4	0.2	0.3	0.8	1.2	0.4	0.8	0.8	1.8	0.7	1.2	0.2	0.4	0.6	1.0
NAGASCMB	5.5	4.9	7.2	5.1	6.2	6.9	4.5	3.3	8.0	11.3	7.9	5.7	5.7	5.2	7.8	5.9	6.1	2.7
NRDIESEL	2.2	4.2	4.5	6.6	3.2	4.4	3.9	8.3	6.5	12.0	5.9	9.4	4.6	8.7	3.5	8.5	4.3	7.6
NRFUELOIL	0.1	0.2	0.1	0.1	0.2	0.3	0.0	0.1	0.0	0.1	0.1	0.3	0.1	0.1	0.0	0.1	0.1	0.1
NRGASOLINE	1.4	2.7	1.6	2.3	1.7	2.2	2.6	5.4	2.5	4.6	2.5	3.8	2.2	4.1	2.4	5.9	2.4	4.1
NRLPG	0.1	0.1	0.2	0.3	0.1	0.1	-0.1	-0.1	0.1	0.2	0.1	0.1	0.0	0.0	0.0	0.0	0.0	-0.1
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
NROTHERS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OPENFIRE	7.0	1.3	4.1	0.6	7.1	1.6	5.1	1.1	7.8	1.6	6.9	1.6	5.7	1.5	9.7	2.4	5.5	1.0
ORDIESEL	2.2	4.0	3.6	5.3	3.7	5.2	1.0	1.9	3.9	7.2	3.4	5.9	2.4	4.3	3.2	7.7	2.2	3.8
ORGASOLINE	4.7	6.3	9.7	14.8	5.3	7.6	5.6	6.8	3.9	5.7	4.4	6.6	8.6	10.1	2.9	5.2	6.9	10.7
OTHERCMB	0.6	1.0	0.6	0.9	0.3	0.4	0.5	1.0	0.4	0.7	0.6	1.2	0.6	1.1	0.3	0.7	0.5	0.8
OTHERS	5.1	3.2	9.6	9.0	3.8	5.0	5.3	3.2	3.6	3.2	4.9	4.1	5.9	2.7	4.1	3.3	5.2	3.5
PRESCRBURN	2.4	1.6	0.0	0.0	0.0	0.0	0.5	0.7	0.0	0.0	0.0	0.0	1.5	0.9	0.0	0.0	0.0	0.0
RAILROAD	0.3	0.7	0.5	0.7	0.2	0.3	-0.5	-1.1	0.1	0.2	0.1	0.3	0.3	0.6	0.2	0.4	0.0	0.0
SEASALT	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SOLVENT	1.0	1.5	1.0	1.4	0.4	0.5	3.5	6.2	0.7	1.3	1.2	2.1	1.6	2.0	0.7	1.6	1.8	3.0
WILDFIRE	0.1	0.2	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
WOODFUEL	0.5	0.9	0.2	0.4	0.2	0.3	0.3	0.6	0.5	0.9	0.3	0.7	0.3	0.5	0.3	0.7	0.3	0.4
WOODSTOVE	17.0	3.3	8.1	1.8	28.9	11.0	15.0	3.2	18.0	3.7	16.8	3.8	12.7	2.6	17.5	4.4	16.3	2.8

Table S10. (f) Initial and refined source impacts in percentage contributions at Pittsburgh site

Category	1/4/04		1/7/04		1/10/04		1/13/04		1/19/04		1/22/04		1/25/04		1/28/04	
	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.	Init.	Refnd.
AGRIBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
AIRCRAFT	0.1	0.3	0.1	0.1	0.2	0.3	0.1	0.2	0.2	0.3	0.2	0.3	0.2	0.4	0.1	0.1
BIOGENIC	1.5	1.5	1.2	1.5	0.1	0.2	2.3	4.1	0.7	1.4	1.3	2.3	0.1	0.3	0.9	1.4
COALCMB	5.5	13.2	10.0	12.8	21.3	29.3	9.5	14.2	12.6	22.9	12.5	19.8	13.3	20.3	16.8	24.1
DIESELCMB	0.1	0.2	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.1	0.1	0.1	0.0	0.0
DUST	10.2	2.7	14.7	1.9	4.2	0.6	18.4	3.4	10.2	2.0	12.3	2.2	8.0	1.5	13.2	2.0
FUELOILCMB	1.7	3.8	0.9	1.3	5.6	4.4	3.0	5.1	2.5	4.3	2.4	3.9	2.3	3.7	4.3	6.4
LIVESTOCK	6.0	15.6	18.4	25.1	1.9	2.8	12.2	22.2	4.4	8.7	8.7	15.9	5.2	9.8	14.2	22.3
LPGCMB	0.1	0.4	0.1	0.1	0.0	0.1	0.1	0.2	0.1	0.1	0.2	0.3	0.1	0.1	0.1	0.1
LWASTEBURN	11.9	3.1	5.4	0.8	10.7	1.6	5.3	1.0	10.0	2.0	8.4	1.5	12.3	2.4	4.4	0.7
METALPRODUCT	5.0	8.0	4.8	5.0	5.0	6.1	4.0	3.5	5.5	3.7	5.3	4.1	5.8	5.6	5.9	4.1
MEATCOOKING	3.9	1.0	1.6	2.0	3.8	5.4	1.5	1.8	3.2	2.2	2.5	2.7	4.0	5.2	1.2	1.2
MEXCMB_M	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MINERALPRODUCT	2.7	5.7	1.0	1.3	1.7	2.3	1.4	1.6	2.0	3.0	1.5	2.2	2.9	3.8	1.2	1.6
NAGASCMB	5.1	3.1	3.2	3.5	4.5	4.7	4.6	3.8	5.4	2.9	4.8	3.4	5.6	5.4	4.0	2.5
NRDIESEL	5.1	4.7	2.7	3.6	5.2	7.6	2.4	4.3	4.7	8.9	4.4	7.5	5.6	10.3	1.9	2.9
NRFUELOIL	0.9	1.8	0.2	0.3	1.1	1.5	0.2	0.4	0.8	1.5	0.7	1.3	1.3	2.5	0.3	0.4
NRGASOLINE	1.7	1.7	0.7	1.0	1.8	2.7	1.2	2.1	1.7	3.3	1.4	2.6	1.6	3.0	2.0	3.0
NRLPG	0.1	0.1	0.0	0.1	0.0	0.1	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1
NRNAGAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
NROTHERS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OPENFIRE	0.0	0.0	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.1	0.9	0.2	0.2	0.2	0.1	0.1
ORDIESEL	1.6	2.7	1.5	2.0	2.3	3.4	1.6	2.7	2.0	3.8	2.1	3.7	2.8	5.2	1.4	2.1
ORGASOLINE	6.6	12.5	12.1	16.8	9.0	12.4	11.1	15.6	8.7	15.1	8.3	13.8	7.1	10.4	8.8	13.8
OTHERCMB	0.5	1.1	0.8	1.1	0.6	0.8	0.8	1.3	0.7	1.1	0.8	1.2	0.2	0.4	1.0	1.3
OTHERS	13.5	11.0	9.9	13.6	4.5	5.2	7.8	6.0	7.8	7.2	5.9	4.9	5.8	4.9	6.4	5.4
PRESCRBURN	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.2	0.1	0.2	0.0	0.0
RAILROAD	0.4	1.0	0.4	0.6	0.3	0.5	0.7	1.2	0.4	0.7	0.5	0.9	0.4	0.8	0.2	0.4
SEASALT	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SOLVENT	0.3	0.7	1.1	1.5	0.5	0.7	1.3	2.0	0.7	1.3	1.3	1.9	0.3	0.6	1.2	1.8
WILDFIRE	0.0	0.0	0.1	0.1	0.0	0.0	0.1	0.2	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.1
WOODFUEL	0.1	0.2	0.2	0.2	0.1	0.1	0.1	0.2	0.1	0.2	0.2	0.3	0.0	0.1	0.3	0.5
WOODSTOVE	15.0	3.9	8.9	3.5	15.4	6.8	9.7	2.2	15.5	3.1	13.0	2.3	14.5	2.8	9.8	1.5

Table S11. Initial and refined source impacts (%) regrouped to 9 sources and compared with CAMx/PSAT results (Burr and Zhang 2011b)

Category*	Atlanta			Chicago			New York		
	Init.	Refnd.	CAMx/PSAT**	Init.	Refnd.	CAMx/PSAT	Init.	Refnd.	CAMx/PSAT
Biogenic	4.3	7.8	4.2	0.3	0.9	1.3	1.4	2.7	0.9
Biomass	24.9	5.5	20.8	13.0	4.1	8.0	25.4	6.8	15.4
Coal	11.8	19.8	18.7	3.7	8.6	9.1	12.9	20.8	13.0
Diesel	5.3	9.0	10.4	4.0	9.6	10.9	6.3	11.7	10.3
Gas	11.3	13.9	13.6	8.9	16.1	13.6	7.9	11.9	12.9
Industrial	2.9	4.3	7.1	18.6	10.5	19.8	3.8	6.2	10.9
Misc. & Waste	6.1	8.3	4.3	19.0	23.6	9.1	16.0	16.2	8.4
Other Comb.	8.4	9.4	16.6	22.0	21.5	14.6	17.6	21.3	23.2
Other Mob.	25.0	22.0	4.2	10.5	5.0	13.6	8.7	2.4	5.0

* Category definitions can be found in Table 2 of Burr and Zhang (2011a). **At the JST SEARCH site. Note that the CAMx/PSAT results are averages for January 2002.

Table S12. January 2004 average initial and refined source impacts on $PM_{2.5}$ at the six sites: regrouped to 13 sources

Category (each includes both primary and secondary impacts)	Atlanta				Chicago				Detroit				Los Angeles				New York				Pittsburgh			
	Init.		Refnd.		Init.		Refnd.		Init.		Refnd.		Init.		Refnd.		Init.		Refnd.		Init.		Refnd.	
	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank	%	Rank
LDGV*	11.3	6	13.9	4	8.9	6	16.2	2	10.2	4	19.0	2	7.8	6	15.5	2	7.9	6	11.9	5	10.6	5	16.6	3
HDDV*	5.6	7	9.7	5	4.7	7	11.4	3	7.6	6	15.1	3	5.4	8	13.5	3	6.5	8	12.0	4	6.3	6	10.4	4
DUST	13.2	2	2.4	8	9.5	5	2.4	11	10.2	5	2.3	10	9.8	4	3.3	9	8.4	5	1.8	10	12.3	4	2.2	11
BURN*	24.9	1	5.5	7	13.0	4	4.2	9	33.8	1	8.6	6	28.7	1	9.0	5	25.4	1	6.8	6	21.1	1	5.1	5
COALCMB	11.8	4	19.8	1	3.7	8	8.7	4	4.4	7	9.7	4	0.5	10	1.2	10	12.9	2	20.9	1	12.7	3	19.8	2
MEATCOOKING	0.1	12	0.1	12	3.2	9	4.2	8	3.3	8	3.3	9	13.6	3	9.8	4	7.4	7	6.3	7	2.6	10	2.8	9
SEASALT	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13	0.0	13
METALPRODUCT	0.7	11	0.8	11	15.2	2	5.7	6	2.5	10	3.7	8	0.1	11	0.3	12	1.5	10	2.5	9	5.1	7	4.6	6
MINERALPRODUCT	1.1	10	1.7	10	1.4	11	2.6	10	0.8	11	1.7	11	1.9	9	3.3	8	0.7	11	1.1	11	1.8	11	2.4	10
NATURALGAS*	3.7	9	2.2	9	13.6	3	8.7	5	11.3	3	9.1	5	8.7	5	5.9	7	6.1	9	5.1	8	4.8	8	3.8	8
FUELOIL*	4.2	8	6.4	6	3.1	10	5.1	7	3.0	9	4.7	7	7.1	7	8.7	6	11.0	4	15.0	3	3.6	9	4.4	7
AIRCRAFT	11.4	5	18.7	2	0.0	12	0.1	12	0.1	12	0.3	12	0.1	12	0.3	11	0.1	12	0.2	12	0.1	12	0.3	13
AllOthers*	12.0	3	18.6	3	23.7	1	30.7	1	12.6	2	22.6	1	16.4	2	29.2	1	12.2	3	16.4	2	19.2	2	27.6	1

* Regrouped sources: LDGV – light-duty gasoline vehicles, merged from NRGASOLINE and ORGASOLINE; HDDV – heavy-duty diesel vehicles, merged from NRDIESEL, ORDIESEL, RAILROAD and DIESELCMB; BURN – vegetative burning, merged from AGRIBURN, LWASTEBURN, OPENFIRE, PRESCRBU, WILDFIRE, WOODFUEL and WOODSTOVE; NATURALGAS – merged from NAGASCMB and NRNAGAS; FUELOIL - merged from FUELOILC and NRFUELOI; AllOthers – merged from the leftover hybrid sources: BIOGENIC, LIVESTOCK, LPGCMB, MEXCMB_M, NRLPG, NROTHERS, OTHERCMB, OTHERS and SOLVENT.

Table S13. Diesel versus gasoline vehicle impacts at the six sites

Diesel/gasoline ratios	Atlanta	Chicago	Detroit	Los Angeles	New York	Pittsburgh
Refined source impacts (primary only)	2.61	2.19	1.97	2.0	2.17	2.2
$PM_{2.5}$ Emissions	2.57	1.67	2.14	1.87	1.97	3.58
Source impacts from the chosen RM study	0.43	-	0.28	3.0	-	0.49
Refined source impacts (primary and secondary combined)	0.69	0.70	0.79	0.87	1.01	0.63
Total vehicle impacts (diesel and gasoline combined, in percentage)						
Refined source impacts (primary only)	14.1	13.4	21.1	18.0	19.0	14.4
Source impacts from the chosen RM study	19.6	31	19.2	14.8	15.5	14.2
Refined source impacts (primary and secondary combined)	23.6	27.6	34.1	29.0	23.9	27.0

Table S14. Primary versus secondary impacts at the six sites

Refined source impacts (in percentage)	Atlanta	Chicago	Detroit	Los Angeles	New York	Pittsburgh
Primary	53.5	49.1	54.9	64.1	60.9	47.9
Secondary	46.5	50.9	45.1	35.9	39.1	52.1
Source impacts from the chosen RM study (in percentage)						
Primary	31.9	49	40.9	36.3	41.7	42.3
Secondary	68.1	51	59.1	63.7	58.3	57.7

Table S15. Contribution to secondary species with refined source impacts results regrouped to 13 sources

Metro area (site ID)	Atlanta (130890002)		Chicago (170310076)		Detroit (261630001)		Los Angeles (060658001)		New York (360050083)		Pittsburgh (420030008)	
Contribution (%)*	SOC	SO4	SOC	SO4	SOC	SO4	SOC	SO4	SOC	SO4	SOC	SO4
LDGV	1	1	28	2	20	2	3	7	11	1	19	1
HDDV				1		3		12				
DUST											4	
BURN	1	1	3	1	3	2		10	7	1		1
COALCMB		61		39		43		7		43		56
MEATCOOKING								1				
SEASALT												7
METALPRODUCT		1		10		8		2		6		3
MINERALPRODUCT		1		5		3		6		1		4
NATURALGAS		3		10		10		11		5		4
FUELOIL		23		20		20		26		36		21
AIRCRAFT		3						2				
AllOthers	98	6	69	11	77	9	96	17	82	5	77	6

*Note that less then 0.5% contributions are not shown in the table.

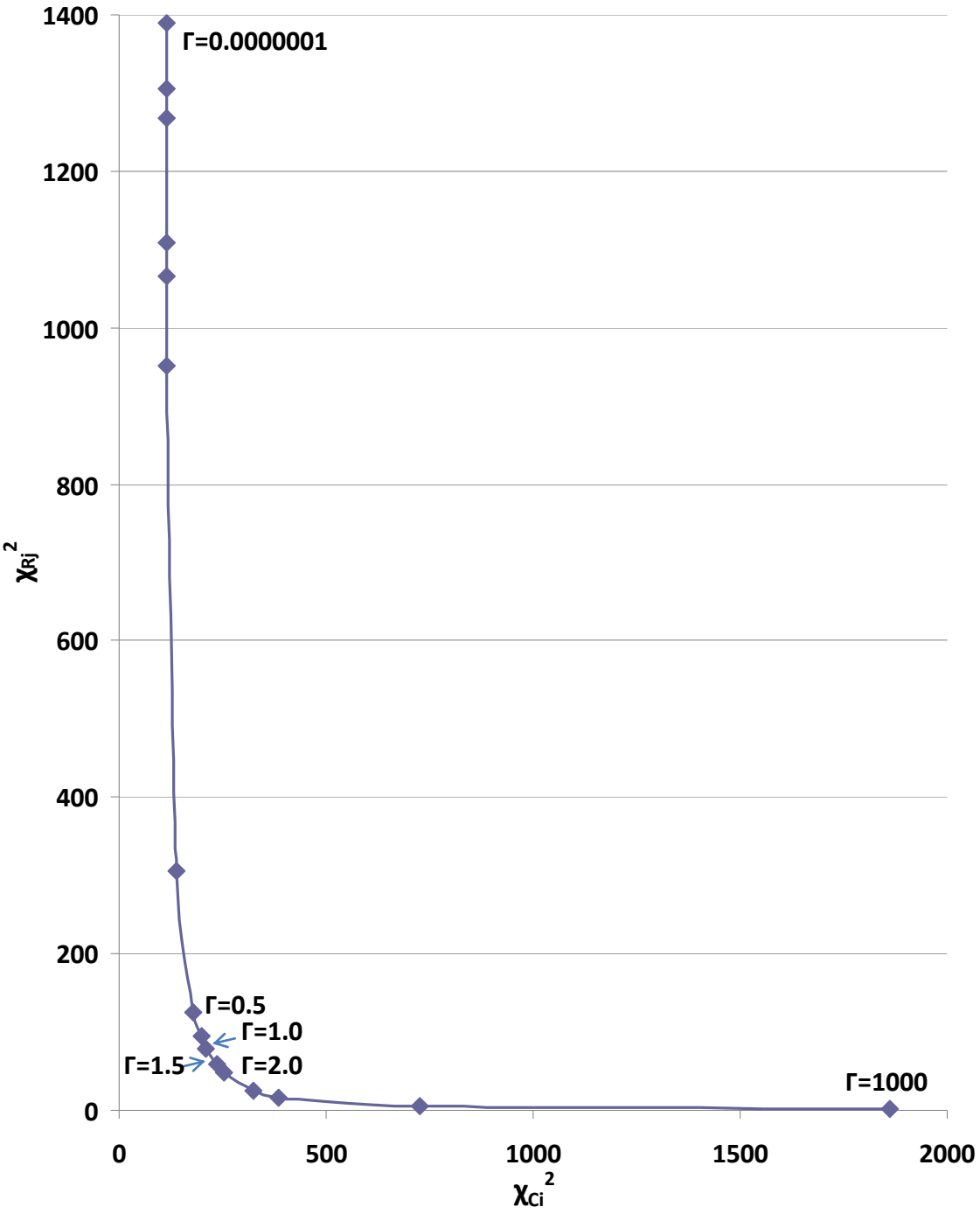


Fig. S1. L-curve plot: objective function terms $\chi_{Rj}^{\ 2}$ versus $\chi_{Ci}^{\ 2}$ at different Γ values, calculated using the case of January 4, 2004 at the Atlanta CSN site.

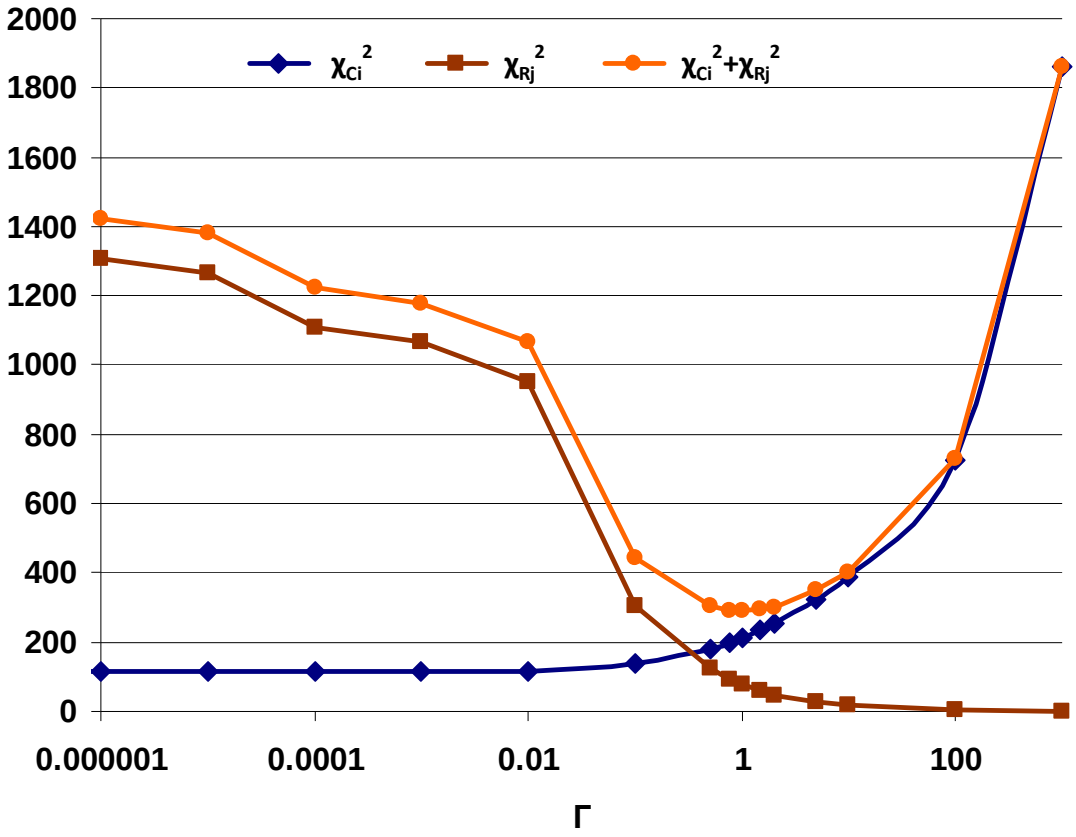
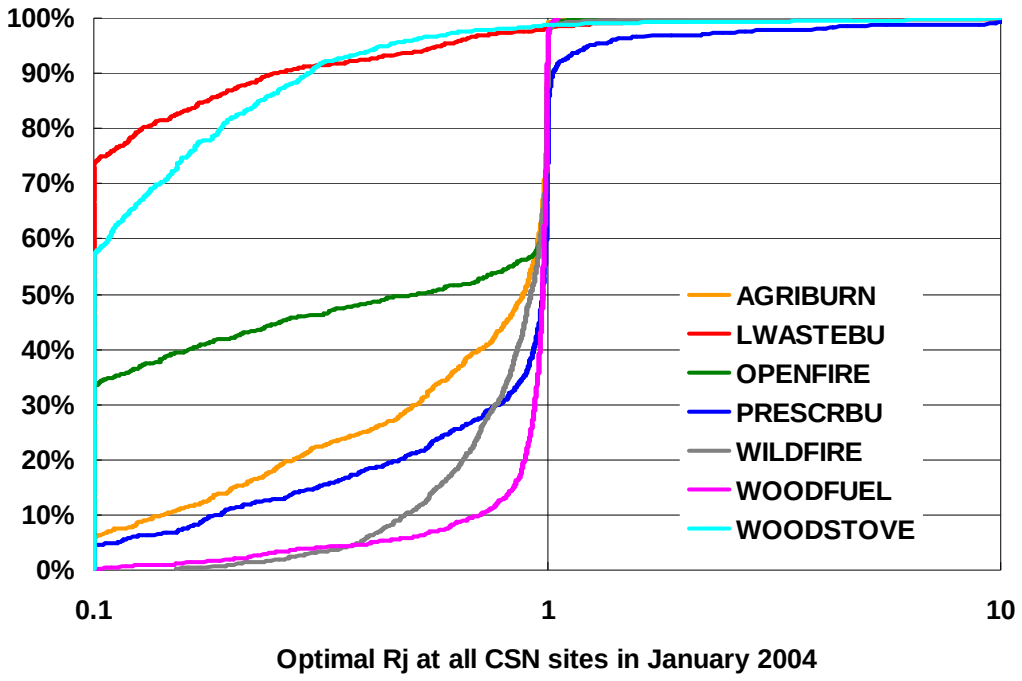


Fig. S2. Objective function terms $\chi_{Ci}^{\ 2}$, $\chi_{Rj}^{\ 2}$ and $\chi_{Ci}^{\ 2} + \chi_{Rj}^{\ 2}$ against different Γ values calculated using the case of January 4, 2004 at the Atlanta CSN site.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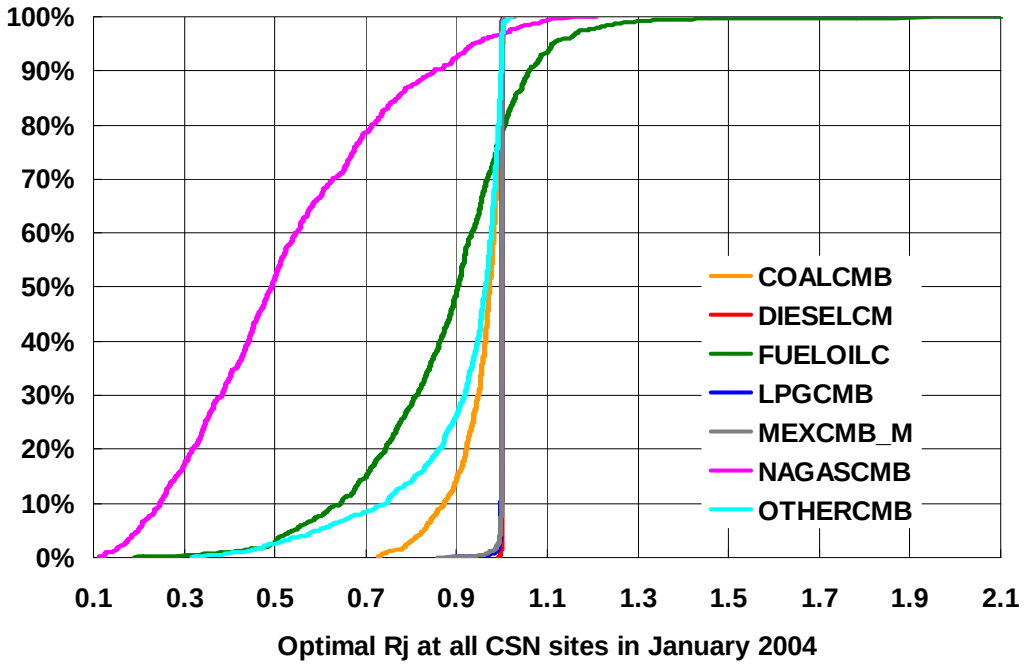
20%
10%
0%
0.1
1
10
AGRIBURN
LWASTEBU
OPENFIRE
PRESCRBU
WILDFIRE
WOODFUEL
WOODSTOVE
Optimal Rj at all CSN sites in January 2004

(a)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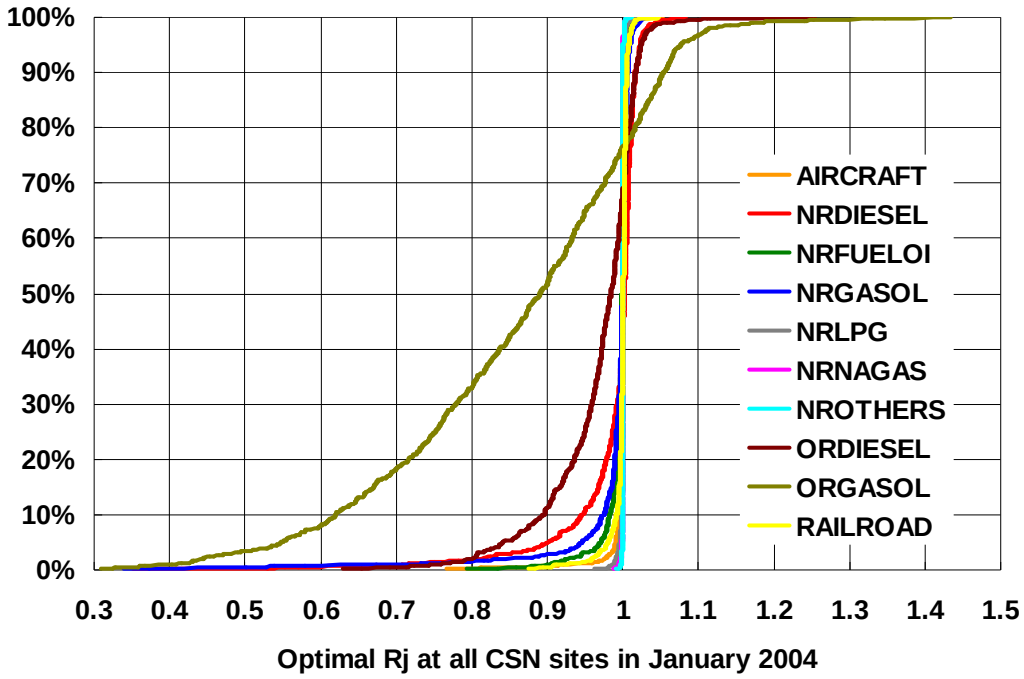
2.1
COALCMB
DIESELCM
FUELOILC
LPGCMB
MEXCMB_M
NAGASCMB
OTHERCMB
Optimal Rj at all CSN sites in January 2004

(b)



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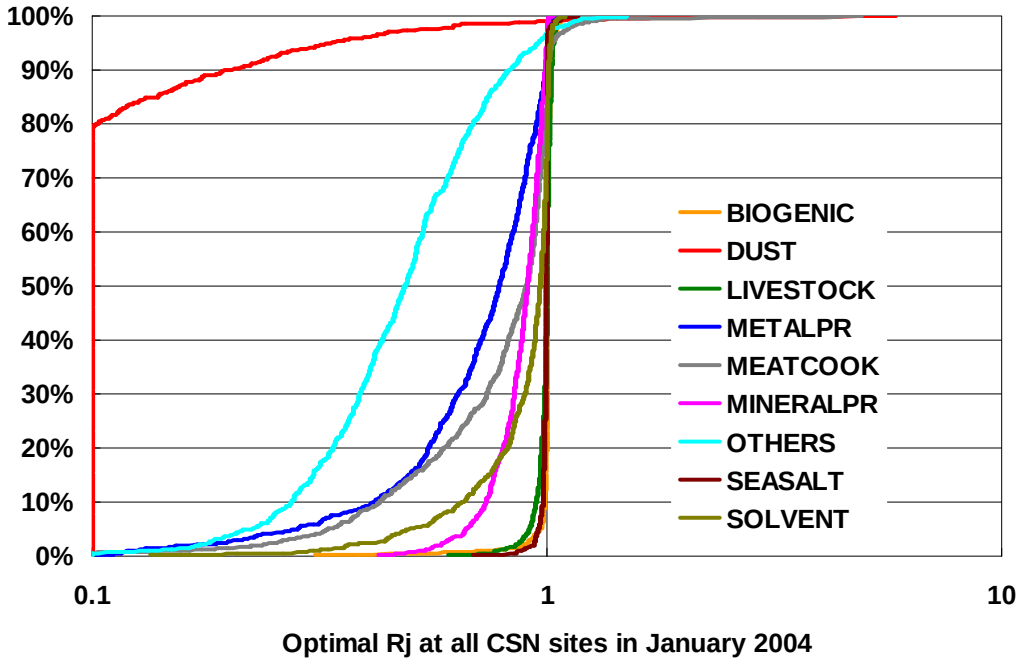

(d)

Fig. S3. Cumulative distribution functions of calculated R_j across 164 CSN sites with valid measurements in January 2004, with source categories grouped by top tier sectors: (a) Biomass-burning, (b) Combustion, (c) Non-road and On-road, and (d) Industrial and Others.

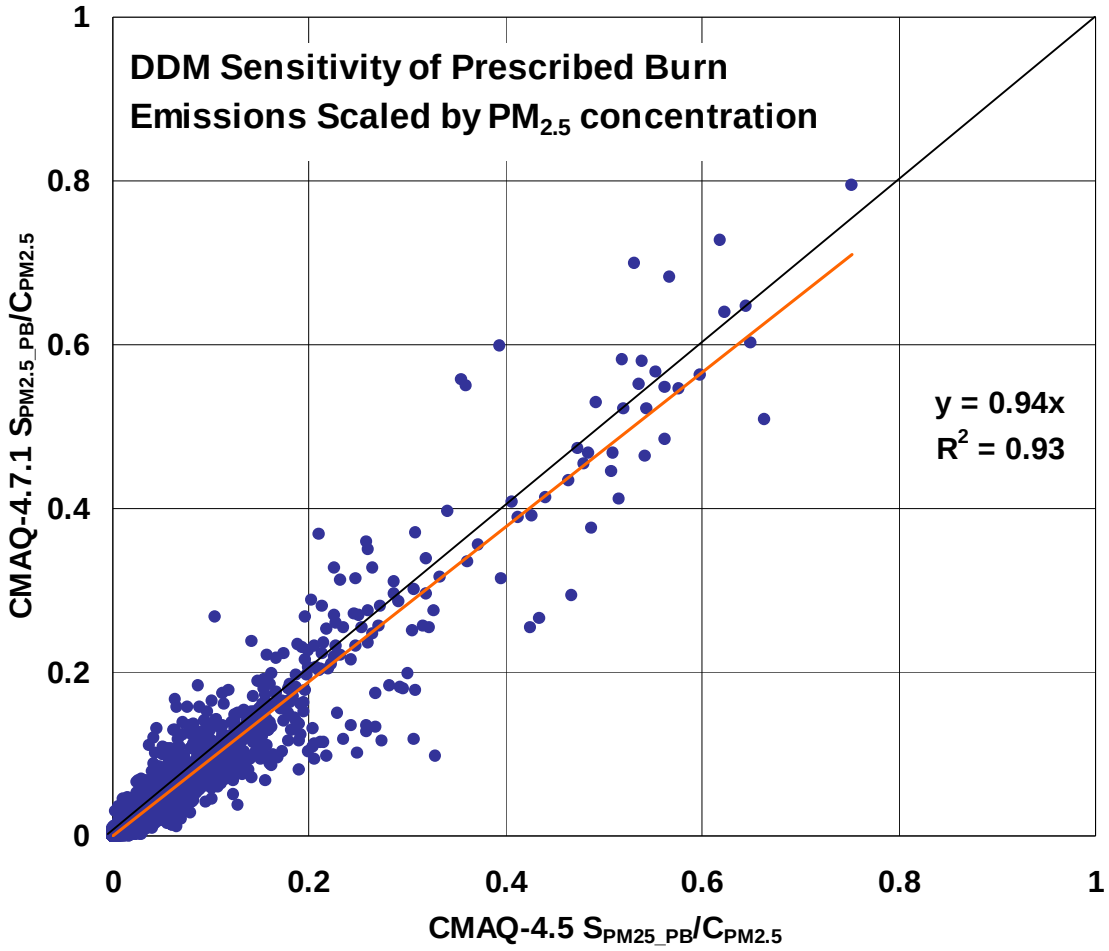

Fig. S4. CMAQ modeling results using different versions: CMAQv4.5 versus CMAQv4.7.1 for DDM sensitivity of prescribed burn emissions to $PM_{2.5}$ concentration (S_{PM25_PB}) scaled by simulated $PM_{2.5}$ concentration ($C_{PM2.5}$) at CSN sites.