Supplement of

Source Apportionment of Organic Carbon in Centreville, AL using Organosulfates in Organic Tracer-based Positive Matrix Factorization

Hettiyadura et al.,

S1. Stability of the 8 factor solution

Three error-estimation methods were used to analyze the stability of the PMF solution: displacement error (DISP), bootstrapping (BS) and BS-DISP error. The results obtained for error estimation for the 8 factor solution are summarized in Table S5 and Figure S1. For both DISP and BS-DISP dQ is <1%. No factor swaps were observed in DISP at dQ_{max} 4, 8 or 16. However, 3 factor swaps were observed at dQ_{max} 32 between SOC by photochemical reactions and monoterpene SOC formed under high-NO_x conditions, suggesting that there is some mixing in these sources, which may be due to the role of ozone in both of the above SOC formation pathways as an oxidant and a reactant. Seven of the 8 factors mapped with ≥80% of the BS runs. Only 55% of the BS runs were mapped with the base run for the monoterpene SOC formed under high-NO_x conditions. Of these 14% mapped with SOC by photochemical reactions, 10% mapped with sulfuric acid-influenced SOC, 8% mapped with isoprene SOC formed under high-NO_x condition, and <5% with remaining sources. The BS-DISP results show that there is some factor interdependence and rotational ambiguity which may be due to the use of a small data set (n=49) (Zhang et al., 2009). Overall the error-estimation results suggest that the 8 factor solution is relatively stable.

Table S1: Summary of PMF settings

Parameter	Description
Data type; sample collection/averaging time frame	$PM_{2.5}$[a]; 13 June – 13 July 2013 in Centreville, AL based on daytime (08:00 – 19:00 LT) and nighttime (20:00 – 07:00) schedule
Excluded data	03 July nighttime – 08 July nighttime (composited samples)
# of species	54
Total variable	Organic carbon
# of samples	49
# of factors	2 to 9
Treatment of missing data	No missing data
Treatment of data below detection limit (BDL)	No modifications or censoring of BDL data
Treatment of concentrations equal to or less than zero	No modifications or censoring of data ≤ 0
Lower limit for normalized factor contributions g_{jk}	-0.2
Robust mode	Yes
Constraints	None
Seed value	A constant seed value (32) when developing a solution (2-9 factors) and a random seed value (5-9 factors) when determining a final solution
# of base runs	20 when developing a solution (2-9 factors) and 100 (5-9 factors) when determining a final solution
# of bootstraps in BS	100
R^2 for BS	0.6
BS block size	1
DISP dQmax	4, 8, 16, 32
# of DISP active species	43 (only the species categorized as strong)
# of bootstraps and r^2 for BS in BS-DISP	100 and 0.6
BS-DISP active species	Elemental carbon, glycolic acid sulfate, hydroxyacetone sulfate, aromatic di and poly carboxylic acids, aliphatic dicarboxylic acids, isoprene SOA tracers, pinonic acid, pinic acid, 3-hydroxyglutaric acid, 2-hydroxy-4,4-dimethylglutaric acid, 3-methyl-1,2,3-butanetricarboxylic acid, levoglucosan, norhopane and hopane
BS-DISP dQmax	0.5. 1, 2, 4
Computer run times for BS-DISP	3 – 9 hours

[a]Water soluble organic carbon (WSOC) in $PM_{2.5}$ was measured until 22 June 2013, after WSOC in PM_1 was measured

Table S2: Summary of PMF input data and statistics

Variable	Concentration (μg m^{-3})			S/N	Category
	Min	Max	Average		
Organic carbon (OC) - *total variable*	9.40E-01	6.01E+00	3.48E+00	9.9	Weak
Elemental carbon (EC)	4.67E-05	3.99E-01	1.77E-01	2.4	Strong
Water soluble organic carbon (WSOC)	5.09E-01	4.80E+00	2.13E+00	7.6	Strong
Hydroxyacetone sulfate	1.67E-03	1.43E-02	6.69E-03	10.0	Strong
Glycolic acid sulfate	4.35E-03	5.25E-02	2.46E-02	2.5	Strong
Lactic acid sulfate	3.72E-03	3.67E-02	1.93E-02	8.1	Weak
2-Methyltetrol sulfate	1.18E-01	2.33E+00	8.01E-01	2.3	Strong
$C_5H_7SO_7^-$ (*m/z* 211)	3.43E-03	9.97E-02	4.24E-02	2.3	Strong
$C_5H_9SO_7^-$ (*m/z* 213)	5.15E-03	9.14E-02	3.79E-02	2.3	Strong
$C_{10}H_{16}NSO_{10}^-$ (*m/z* 342)	6.91E-04	3.17E-02	6.51E-03	2.3	Strong
$C_7H_{11}SO_7^-$ (*m/z* 239)	2.35E-03	1.91E-02	8.09E-03	2.3	Strong
$C_4H_7SO_6^-$ (*m/z* 183)	1.90E-03	2.31E-02	1.10E-02	2.3	Strong
$C_5H_{11}SO_6^-$ (*m/z* 199)	2.88E-04	7.89E-03	3.03E-03	2.3	Strong
$C_3H_7SO_5^-$ (*m/z* 155)	9.87E-05	4.53E-03	1.26E-03	2.3	Strong
2-Methylglyceric acid sulfate	1.15E-03	4.87E-02	1.02E-02	2.3	Strong
Octacosane	5.96E-05	1.13E-03	3.77E-04	1.1	Strong
Nonacosane	2.93E-04	4.22E-03	1.31E-03	2.3	Strong
Triacontane	-8.48E-05	9.72E-04	2.68E-04	0.2	Weak
Hentriacontane	3.64E-05	2.81E-03	7.72E-04	1.1	Strong
Dotriacontane	-1.66E-04	6.37E-04	1.79E-04	0.0	Weak
Tritriacontane	1.11E-04	9.54E-04	3.89E-04	0.3	Weak
Tetratriacontane	-4.79E-04	8.45E-04	2.09E-04	0.0	Weak
Docosanoic acid	2.72E-04	4.84E-03	1.07E-03	2.7	Strong
Tricosanoic acid	0.00E+00	1.53E-03	6.59E-04	0.9	Weak
Tetracosanoic acid	2.86E-04	4.19E-03	1.59E-03	2.6	Strong
Pentacosanoic acid	0.00E+00	9.00E-04	2.57E-04	0.2	Weak

Table S2 (continued)

Variable	Concentration (μg m^{-3})			S/N	Category
	Min	Max	Average		
Hexacosanoic acid	2.43E-04	2.10E-03	1.01E-03	1.5	Strong
Heptacosanoic acid	0.00E+00	7.63E-04	1.64E-04	0.1	Weak
Octacosanoic acid	4.38E-04	3.82E-03	1.38E-03	3.1	Strong
Phthalic acid	8.39E-04	2.08E-02	5.43E-03	3.8	Strong
Isophthalic acid	0.00E+00	3.05E-03	1.04E-03	2.9	Strong
Terephthalic acid	1.27E-03	1.63E-02	5.05E-03	3.9	Strong
1,2,4-Benzenetricarboxylic acid	2.62E-04	3.51E-03	1.55E-03	3.4	Strong
1,2,4,5-Benzenetetracarboxylic acid	0.00E+00	9.55E-04	4.70E-04	2.0	Strong
Methylphthalic acid	-5.06E-05	2.87E-03	8.38E-04	2.6	Strong
Succinic acid	0.00E+00	1.35E-02	5.97E-03	3.9	Strong
Glutaric acid	0.00E+00	4.04E-03	1.91E-03	3.6	Strong
Adipic acid	0.00E+00	6.90E-03	2.76E-03	3.7	Strong
Suberic acid	0.00E+00	5.86E-03	1.60E-03	3.2	Strong
Azelaic acid	7.39E-04	4.88E-03	2.19E-03	3.7	Strong
2-Methylglyceric acid	1.64E-03	3.86E-02	1.41E-02	4.0	Strong
2-Methylthreitol	1.22E-03	1.13E-01	3.83E-02	4.0	Strong
2-Methylerythritol	1.55E-02	2.30E-01	6.59E-02	4.0	Strong
Pinonic acid	5.70E-03	2.03E-01	6.22E-02	4.0	Strong
3-Hydroxyglutaric acid	9.67E-03	2.99E-01	8.70E-02	4.0	Strong
Pinic acid	7.48E-03	1.02E-01	3.30E-02	4.0	Strong
2-Hydroxy-4,4-dimethylglutaric acid	4.31E-03	9.76E-02	4.03E-02	4.0	Strong
3-Acetyl hexanedioic acid	1.21E-02	1.35E-01	5.37E-02	4.0	Strong
3-Methyl-1,2,3-butanetricarboxylic acid	0.00E+00	1.67E-02	1.52E-03	1.7	Strong
Levoglucosan	2.81E-03	1.06E-01	2.87E-02	3.6	Strong
Dehydroabietic acid	5.75E-05	1.08E-02	1.48E-03	2.1	Weak
7-Oxodehydroabietic acid	0.00E+00	2.01E-03	4.21E-04	0.8	Weak

Table S2 (continued)

Variable	Concentration (μg m^{-3})			S/N	Category
	Min	Max	Average		
17α(H)-21β(H)-30-Norhopane	0.00E+00	9.57E-05	1.88E-05	1.4	Strong
17α(H)-21β(H)-Hopane	0.00E+00	1.26E-04	2.91E-05	1.9	Strong

Table S3: Summary of PMF and error estimation diagnostics obtained for 5 to 9 factor solutions.

Diagnostic	5 Factors	6 Factors	7 Factors	8 Factors	9 Factors
$Q_{expected}$	1647	1555	1463	1371	1279
Q_{robust} (minimum)	4150	3542	3060	2657	2286
Q_{true} (corresponding to min Qrobust)	4196	3561	3072	2664	2294
$Q_{robust}/Q_{expected}$	2.52	2.28	2.09	1.94	1.79
Species with $Q_{true}/Q_{expected} > 3$	$C_{10}H_{16}NSO_{10}^-$(*m/z* 342), 2-methylglyceric acid sulfate, terephthalic acid, succinic acid, 2-methylglyceric acid, 2-methylerythritol, 2-hydroxy-4,4-dimethylglutaric acid, 3-methyl-1,2,3-butanetricarboxylic acid, levoglucosan, 17α(H)-21β(H)-hopane	$C_{10}H_{16}NSO_{10}^-$(*m/z* 342), 2-methylglyceric acid sulfate, suberic acid, 2-methylglyceric acid, 3-methyl-1,2,3-butanetricarboxylic acid, levoglucosan	$C_{10}H_{16}NSO_{10}^-$ (*m/z* 342), suberic acid, 2-methylglyceric acid, levoglucosan, 17α(H)-21β(H)-hopane	$C_{10}H_{16}NSO_{10}^-$(*m/z* 342), succinic acid, 2-methylglyceric acid, 17α(H)-21β(H)-hopane	$C_{10}H_{16}NSO_{10}^-$(*m/z* 342), 2-methylglyceric acid, 17α(H)-21β(H)-hopane
DISP %dQ	0	0	0	0	0
DISP swaps	0	Only at dQmax = 32	At dQmax = 16 and 32	Only at dQmax = 32	At dQmax = 16 and 32
Factors with BS mapping < 80 %	Oxidatively aged biogenic SOC (66 %)	Sulfuric acid-influenced SOC (77 %)	Monoterpene SOC (high-NO_x) (67%)	Monoterpene SOC (high-NO_x) (55 %)	Monoterpene SOC (high-NO_x) (75 %)
BS-DISP % cases with swaps	16	14	30	48	56

Table S4: PMF diagnostics for 8 factor solution at Fpeak = 0

Variable	Observed/predicted graph		Q_{true}/Q_{exp}
	Slope	R^2	
Organic carbon (OC)	1.078	0.931	0.359
Elemental carbon (EC)	0.681	0.740	1.997
Water soluble organic carbon (WSOC)	0.942	0.936	1.700
Hydroxyacetone sulfate	0.974	0.995	0.651
Glycolic acid sulfate	0.625	0.827	1.253
Lactic acid sulfate	0.681	0.811	0.909
2-Methyltetrol sulfate	0.434	0.519	1.630
$C_5H_7SO_7^-$ (*m/z* 211)	0.797	0.783	0.807
$C_5H_9SO_7^-$ (*m/z* 213)	0.549	0.547	1.458
$C_{10}H_{16}NSO_{10}^-$ (*m/z* 342)	0.032	0.023	4.365
$C_7H_{11}SO_7^-$ (*m/z* 239)	0.768	0.829	0.356
$C_4H_7SO_6^-$ (*m/z* 183)	0.760	0.762	0.901
$C_5H_{11}SO_6^-$ (*m/z* 199)	0.376	0.573	2.728
$C_3H_7SO_5^-$ (*m/z* 155)	0.407	0.407	1.443
2-Methylglyceric acid sulfate	0.325	0.540	2.561
Octacosane	0.789	0.876	0.350
Nonacosane	0.486	0.537	1.700
Triacontane	0.614	0.681	0.030
Hentriacontane	0.437	0.522	1.286
Dotriacontane	0.402	0.420	0.030
Tritriacontane	0.621	0.529	0.026
Tetratriacontane	0.105	0.096	0.114
Docosanoic acid	0.516	0.598	1.384
Tricosanoic acid	0.883	0.838	0.036
Tetracosanoic acid	0.869	0.907	0.974
Pentacosanoic acid	0.422	0.542	0.050

Table S4 (continued)

Variable	Observed/predicted graph		Qt_{rue}/Q_{exp}
	Slope	R^2	
Hexacosanoic acid	0.884	0.840	0.508
Heptacosanoic acid	0.319	0.398	0.050
Octacosanoic acid	0.672	0.686	1.965
Phthalic acid	0.548	0.709	2.419
Isophthalic acid	0.605	0.751	2.309
Terephthalic acid	0.490	0.732	2.734
1,2,4-Benzenetricarboxylic acid	0.770	0.835	1.550
1,2,4,5-Benzenetetracarboxylic acid	0.584	0.506	2.730
Methylphthalic acid	0.526	0.637	1.561
Succinic acid	0.623	0.476	3.772
Glutaric acid	0.868	0.780	1.842
Adipic acid	0.657	0.759	2.525
Suberic acid	0.690	0.771	2.991
Azelaic acid	0.986	0.855	1.139
2-Methylglyceric acid	0.485	0.534	3.979
2-Methylthreitol	0.606	0.672	3.288
2-Methylerythritol	0.588	0.657	3.309
Pinonic acid	0.781	0.776	2.221
3-Hydroxyglutaric acid	0.584	0.706	2.421
Pinic acid	0.792	0.793	2.045
2-Hydroxy-4,4-dimethylglutaric acid	0.746	0.744	2.227
3-Acetyl hexanedioic acid	0.702	0.794	2.565
3-Methyl-1,2,3-butanetricarboxylic acid	0.576	0.716	1.933
Levoglucosan	0.774	0.918	1.240
Dehydroabietic acid	0.267	0.553	0.529
7-Oxodehydroabietic acid	0.661	0.800	0.133

Table S4 (continued)

Variable	Observed/predicted graph		Q_{true}/Q_{exp}
	Slope	R^2	
17α(H)-21β(H)-30-Norhopane	0.753	0.827	2.120
17α(H)-21β(H)-Hopane	0.785	0.807	4.069

Table S5: Error estimation summary for 8 factor solution at Fpeak = 0.

BS-DISP diagnostics									
# of Cases Accepted:	52								
% of Cases Accepted:	52%								
Largest Decrease in Q:	-13.47500038								
%dQ:	-0.507137576								
# of Decreases in Q:	0								
# of Swaps in Best Fit:	9								
# of Swaps in DISP:	39								
Swaps by Factor:		Isoprene SOC (high-NO_x)	Sulfuric acid-influenced SOC	Oxidatively aged biogenic SOC	SOC by photochemical reactions	Biomass burning	Isoprene SOC (low-NO_x)	Monoterpene SOC (high-NO_x)	Vehicle emissions
	# of swaps at dQmax = 0.5	2	5	12	7	8	2	1	8

Table S5 (continued)

DISP diagnostics

Error Code:	0
Largest Decrease in Q:	0
%dQ:	0

Swaps by Factor:	Isoprene SOC (high-NO_x)	Sulfuric acid-influenced SOC	Oxidatively aged biogenic SOC	SOC by photochemical reactions	Biomass burning	Isoprene SOC (low-NO_x)	Monoterpene SOC (high-NO_x)	Vehicle emissions
dQmax = 4, 8 and 16	0	0	0	0	0	0	0	0
dQmax = 32	0	0	0	3	0	0	3	0

BS mapping

	Isoprene SOC (high-NO_x)	Sulfuric acid-influenced SOC	Oxidatively aged biogenic SOC	SOC by photochemical reactions	Biomass burning	Isoprene SOC (low-NO_x)	Monoterpene SOC (high-NO_x)	Vehicle emissions	Unmapped
Boot isoprene SOC formed under high-NO_x conditions	80	4	0	5	1	2	0	4	4
Boot sulfuric acid-influenced SOC	1	94	0	2	1	0	1	1	0
Boot oxidatively aged biogenic SOC	0	0	80	3	2	2	2	3	8
Boot SOC by photochemical reactions	1	2	0	92	0	2	0	1	2
Boot biomass burning	0	4	0	0	88	2	2	2	2
Boot isoprene SOC formed under low-NO_x conditions	1	1	0	0	0	97	0	0	1
Boot monoterpene SOC formed under high-NO_x conditions	8	10	1	14	3	4	55	5	0
Boot vehicle emissions	0	1	0	1	0	0	0	98	0

Table S6: Pearson's correlations (r) for organosulfates with isoprene, its gas phase oxidation products, isoprene secondary organic aerosol (SOA) tracers, sulfate, aerosol acidity, and aerosol water in Centreville, AL during SOAS.

		N	[a]Hydroxyacetone sulfate	[a]Glycolic acid sulfate	[a]Lactic acid sulfate	2-Methyltetrol sulfate	$C_5H_7SO_7^-$ (*m/z* 211)	$C_5H_9SO_7^-$ (*m/z* 213)	$C_{10}H_{16}NSO_{10}^-$ (*m/z* 342)	$C_7H_{11}SO_7^-$ (*m/z* 239)	$C_4H_7SO_6^-$ (*m/z* 183)	$C_5H_{11}SO_6^-$ (*m/z* 199)	$C_3H_7SO_5^-$ (*m/z* 155)	2-Methylglyceric acid sulfate
	Isoprene	59	.447**	.443**	.397**	.432**	.358**	.445**	-0.079	.359**	.312*	.289*	.260*	0.240
Isoprene oxidation products	Isoprene hydroxynitrates (ISOPN)	42	0.295	.316*	.398**	0.270	0.260	0.202	-0.058	0.057	0.274	0.232	0.262	0.084
	Isoprene epoxydiols (IEPOX)	38	0.141	.404*	.410*	.540**	.587**	.368*	-0.107	-0.001	0.157	.529**	0.178	0.018
	Isoprene hydroxyl hydroperoxides (ISOPOOH)	38	0.315	.520**	.475**	.674**	.541**	.496**	-.323*	0.100	0.281	.473**	0.314	0.081
	Methacrolein (MACR)	59	.592**	.673**	.670**	.633**	.633**	.570**	0.239	.514**	.520**	.571**	.411**	.327*
	Methylvinyl ketone (MVK)	59	.351**	.303*	.433**	.447**	.436**	.294*	0.146	0.158	.380**	.395**	.289*	0.050
	Glyoxal	60	.564**	.599**	.649**	.562**	.495**	.554**	0.062	.438**	.533**	.493**	.490**	.365**
	Formaldehyde	60	.689**	.729**	.760**	.619**	.615**	.650**	0.210	.600**	.598**	.532**	.463**	.489**
	Glycolaldehyde	38	.388*	.428**	.501**	.521**	.405*	.419**	-0.132	0.154	.392*	.447**	.325*	0.157
	Hydroxyacetone	42	.629**	.682**	.703**	.627**	.597**	.620**	-0.124	.454**	.473**	.569**	.436**	.402**
Isoprene SOA tracers	2-Methylglyceric acid	49	.461**	.703**	.603**	.536**	.678**	.565**	0.053	.463**	.395**	.562**	.422**	.386**
	2-Methylthreitol	49	.512**	.502**	.474**	.383**	.310*	.527**	-0.189	.427**	.309*	.324*	.315*	.341*
	2-Methylerythritol	49	.428**	.324*	0.269	0.237	0.193	.483**	-0.179	.483**	0.212	0.189	0.072	.459**
	Sulfate	60	.629**	.691**	.741**	.474**	.440**	.466**	.289*	.524**	.575**	.493**	.767**	.272*
	Aerosol acidity ($[H^+]$)	49	0.196	-0.140	0.126	-0.027	-0.151	-0.035	-0.056	0.002	.343*	-0.127	0.162	0.006
	Aerosol water	56	.334*	.316*	0.259	0.083	0.155	0.233	.428**	.485**	0.217	0.100	0.217	.265*

**Correlation is significant at the 0.01 level (2-tailed). *Correlation is significant at the 0.05 level (2-tailed). [a]Correlations of hydroxyacetone sulfate, lactic acid sulfate, and glycolic acid sulfate were obtained from Hettiyadura et al. (2017).

Table S7: Pearson's correlations (r) of organic tracer-based PMF factors resolved for $PM_{2.5}$ OC in this study with AMS-PMF factors resolved for PM_1 in Centreville during SOAS by Xu et al. (2015a) and Xu et al. (2015b). Also, given are the 'r' for sum of the several organic tracer-based PMF factors with AMS-PMF factors. The AMS-PMF factors include biomass burning organic aerosols (BBOA), isoprene-derived organic aerosols (Isoprene-OA), less-oxidized oxygenated organic aerosols (LO-OOA), and more-oxidized oxygenated organic aerosols (MO-OOA).

	AMS-PMF factors			
Organic tracer-based PMF factors	**BBOA**	**Isoprene-OA**	**LO-OOA**	**MO-OOA**
Biomass burning	0.602**	0.108	0.472**	0.076
Vehicle emissions	0.596**	0.427**	0.527**	0.493**
Isoprene SOC formed under low-NO_x conditions	-0.130	0.494**	-0.173	-0.021
Isoprene SOC formed under high-NO_x conditions	0.093	0.194	0.005	0.334*
SOC by photochemical reactions	-0.016	0.051	-0.194	0.332*
Oxidatively aged biogenic SOC	0.307*	0.249	0.092	0.382**
Sulfuric acid-influenced SOC	0.176	0.173	0.051	0.467**
Monoterpene SOC formed under high-NO_x conditions	0.385**	-0.051	0.621**	0.049
Sum of isoprene SOC formed under low-NO_x conditions and isoprene SOC formed under high-NO_x conditions	-0.043	0.473**	-0.125	0.174
Sum of sulfuric acid-influenced SOC, SOC by photochemical reactions and oxidatively aged biogenic SOC	0.272	0.267	0.012	0.661**

**Correlation is significant at the 0.01 level (2-tailed). *Correlation is significant at the 0.05 level (2-tailed).

Figure S1: Change of the ratio of Q_{robust} (goodness-of-fit parameter calculated excluding points not fit by the model by PMF) to $Q_{expected}$ (the difference between the number of non-weak species, m*, by number of samples, n, and the m* by number of factors, p, and n by p, taken together) when increasing the number of factors from 5 to 9 for source apportionment of $PM_{2.5}$ organic carbon in Centreville, AL during the SOAS using organic tracer-based positive matrix factorization model.

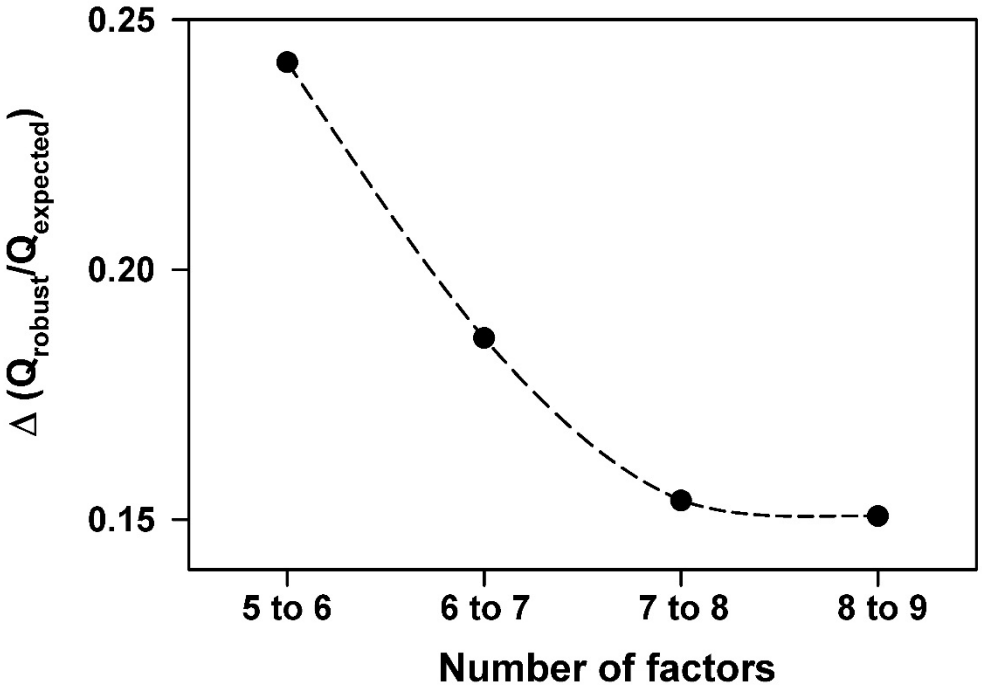


Figure S2: Average source contributions to $PM_{2.5}$ organic carbon (OC; μg m^{-3}, squares) in Centreville, AL during SOAS using organic tracer-based PMF. The circles represent the median OC from bootstrapping (BS) and the average OC from displacement (DISP) and BS-DISP error estimation methods. The error bars represent the concentration range of OC from DISP, BS and BS-DISP. The lower and the upper whiskers represent the 5th and the 95th percentiles of the OC concentrations from BS and BS-DISP and the minimum and maximum OC concentrations from DISP.

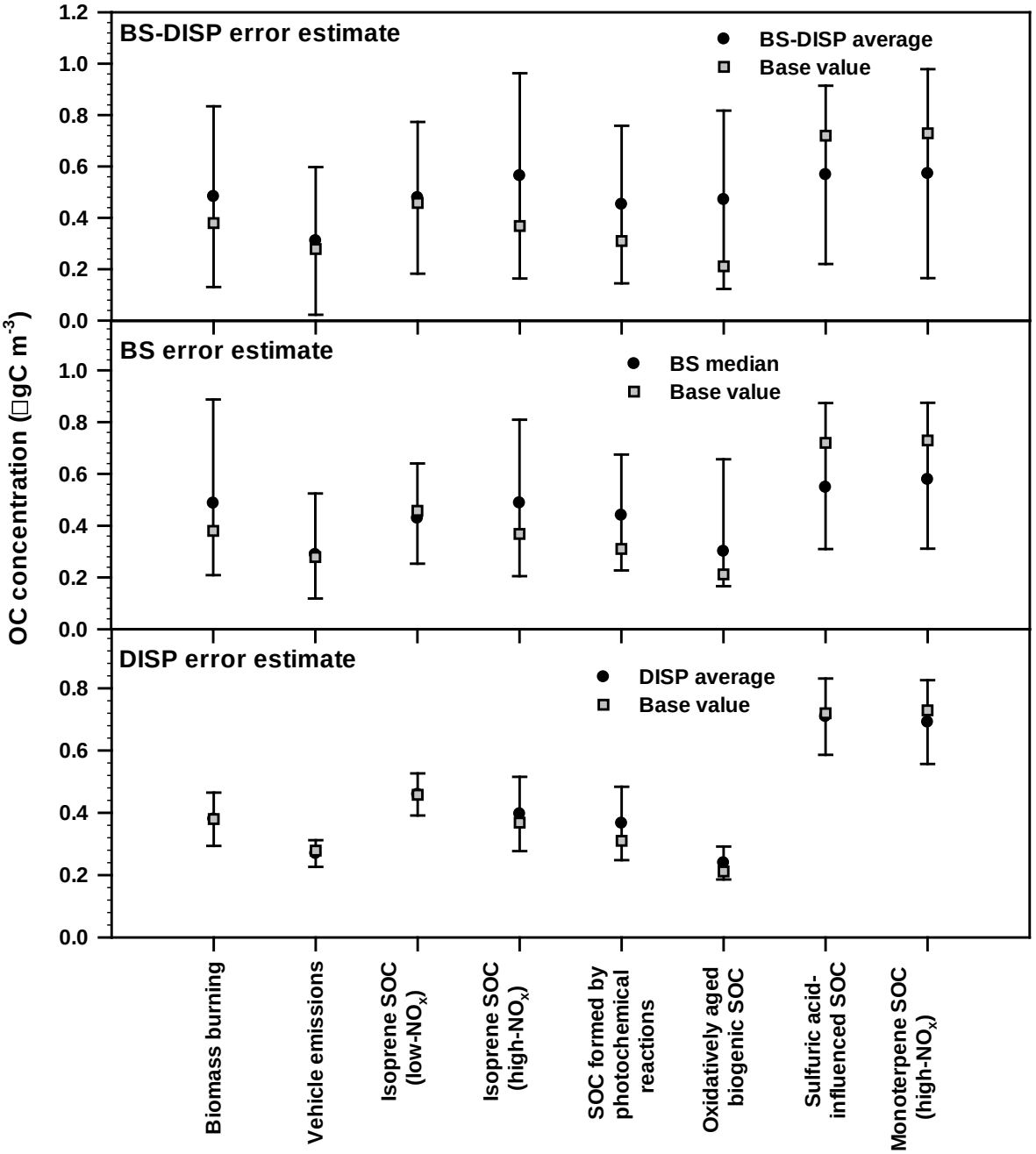


Figure S3: The average source contributions to $PM_{2.5}$ organic carbon in daytime, nighttime, and for both day and nighttime samples in Centreville, AL during the SOAS using organic tracer-based PMF model.

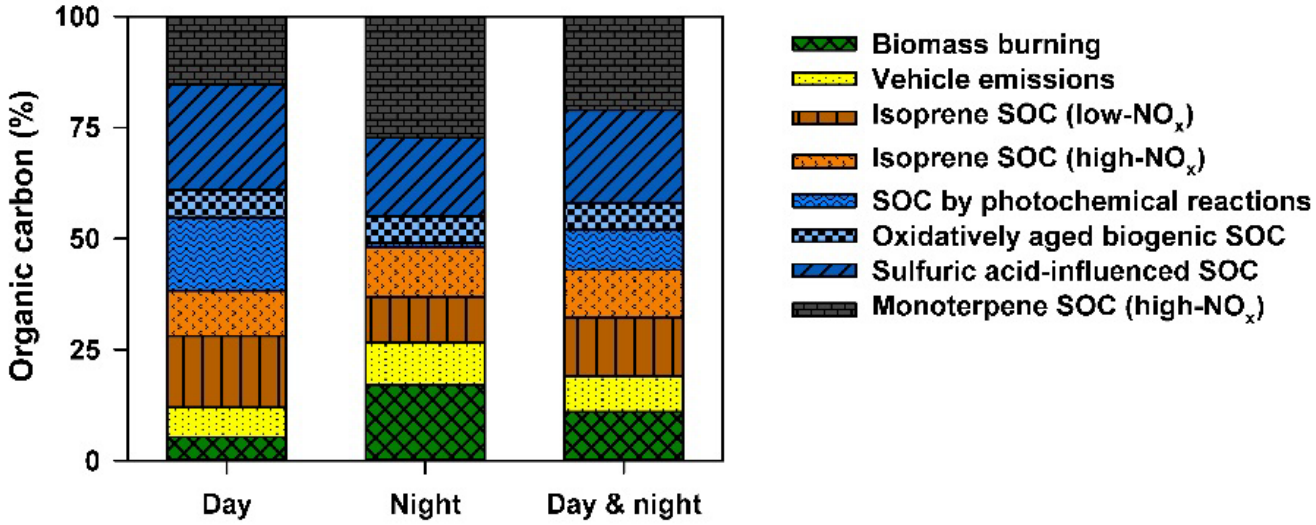
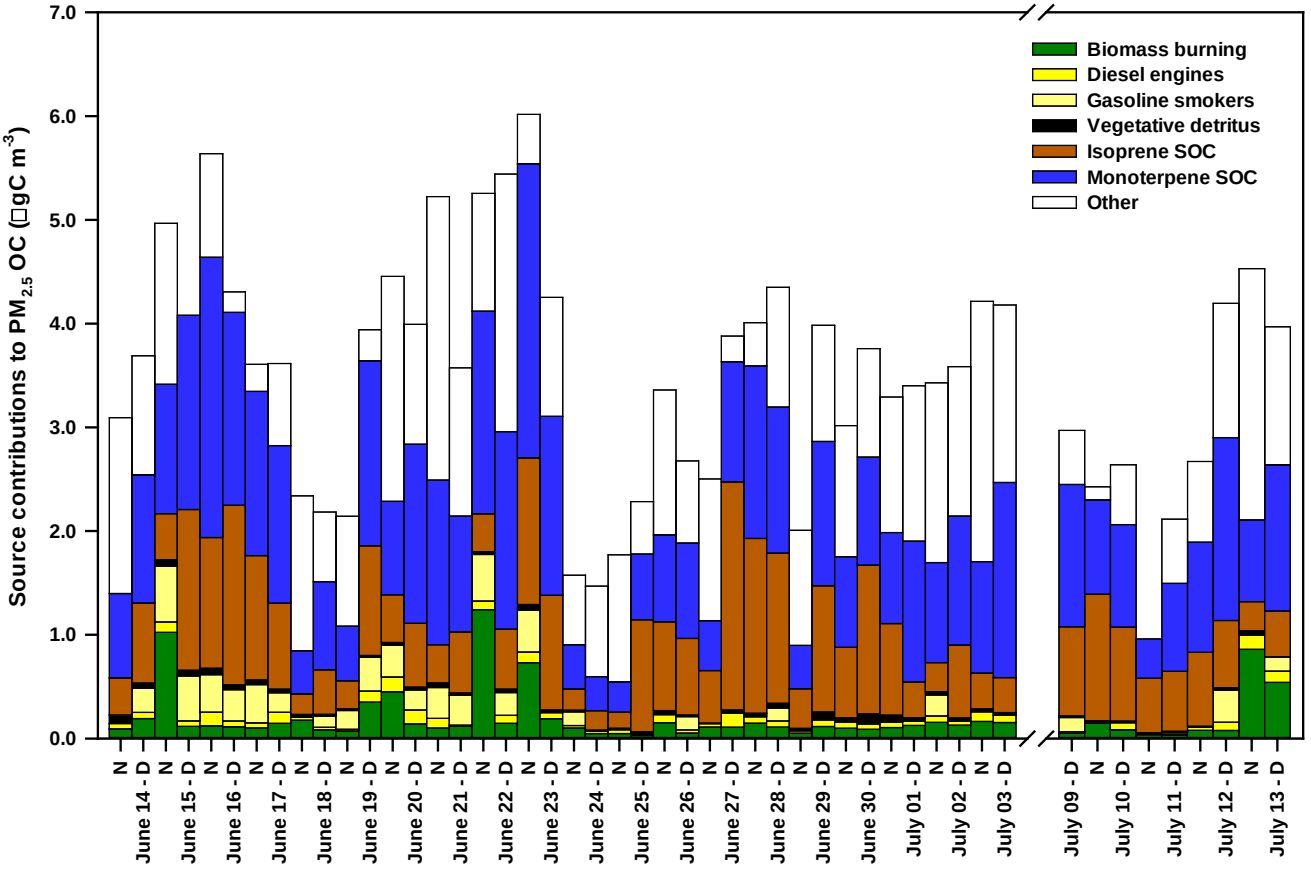


Figure S4: Source contributions to $PM_{2.5}$ organic carbon (OC) in Centreville, AL during the SOAS using chemical mass balance model; D: samples collected during daytime (8:00 AM to 7:00 PM); N: Samples collected during nighttime (8:00 PM to 7:00 AM).



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