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Supplement of

Simulating secondary organic aerosol in a regional air quality model using the statistical oxidation model – Part 2: Assessing the influence of vapor wall losses

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The Supplemental Material consists of six figures and two tables.

Influence of NO_x Parameterization: In the manuscript the influence of vapor wall losses was calculated for the low vapor wall loss (VWL) and high VWL cases as:

$$R_{wall}(lowVWL) = \frac{[SOA]_{lowVWL,lowNO_x}+[SOA]_{lowVWL,highNO_x}}{[SOA]_{noVWL,lowNO_x}+[SOA]_{noVWL,highNO_x}} \quad \text{(S1)}$$

or

$$R_{wall}(highVWL) = \frac{[SOA]_{highVWL,lowNO_x}+[SOA]_{highVWL,highNO_x}}{[SOA]_{noVWL,lowNO_x}+[SOA]_{noVWL,highNO_x}} \quad \text{(S2)}$$

and where "low NO_x" and "high NO_x" refer to the experimental conditions under which the experiments were performed to which the SOM was fit (see Table S1). The influence of the NO_x parameterization for a given VWL case can also be assessed by comparing R_{wall} values calculated individually for each NO_x condition:

$$R_{wall}(low\ NO_x) = \frac{[SOA]_{lowVWL,lowNO_x}}{[SOA]_{noVWL,lowNO_x}}\ or = \frac{[SOA]_{highVWL,lowNO_x}}{[SOA]_{noVWL,lowNO_x}} \quad \text{(S3)}$$

and

$$R_{wall}(high\ NO_x) = \frac{[SOA]_{lowVWL,highNO_x}}{[SOA]_{noVWL,highNO_x}}\ or = \frac{[SOA]_{highVWL,highNO_x}}{[SOA]_{noVWL,highNO_x}} \quad \text{(S4)}$$

The R_{wall} values from Eqns. S3 and S4 are compared in Figure S3. It is clear that there is some difference between the simulated R_{wall} values between the low-NO_x and high-NO_x parameterizations, although most points fall close to the one-to-one line. At very low R_{wall} values, the high-NO_x parameterization gives slightly lower R_{wall} than does the low-NO_x parameterization for both the SOM-lowVWL and SOM-highWVL cases. But as the absolute R_{wall} values increase the opposite is true. Regardless, the differences between the NO_x-parameterizations are much smaller than the absolute values of the simulated R_{wall} values.

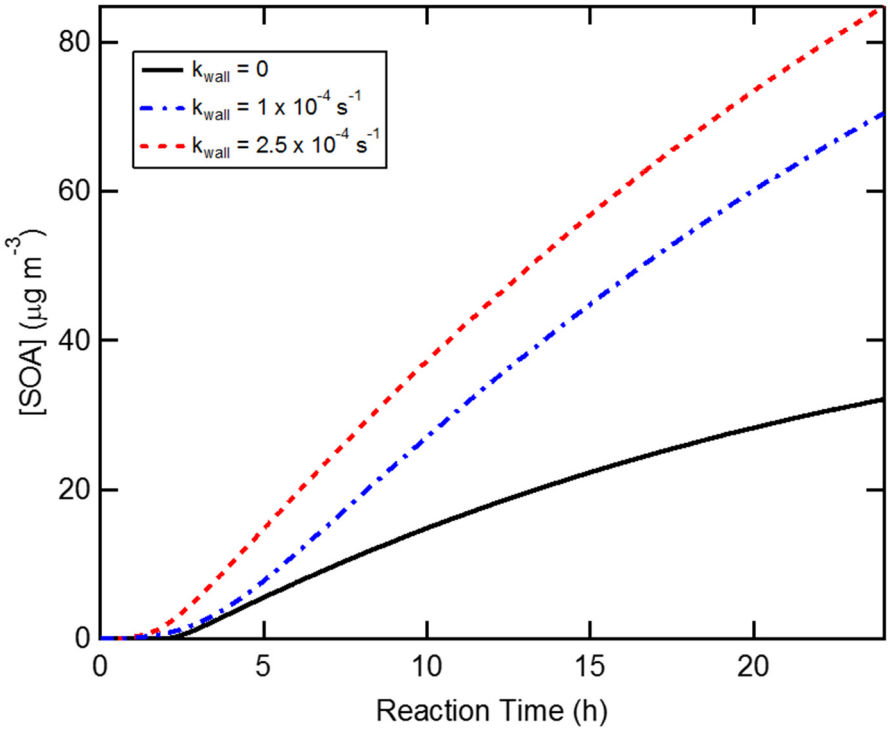

Figure S1. Box model simulations of SOA formation using SOM parameters determined from fitting low-NO_x toluene + OH SOA data assuming k_{wall} = 0, 1 x 10^{-4} and 2.5 x 10^{-4} s^{-1}, but where the simulations are run with k_{wall} = 0 s^{-1}. Reaction conditions here are $[toluene]_{t=0}$ = 100 μg m^{-3} and [OH] = 2 x 10^6 molecules cm^{-3}.

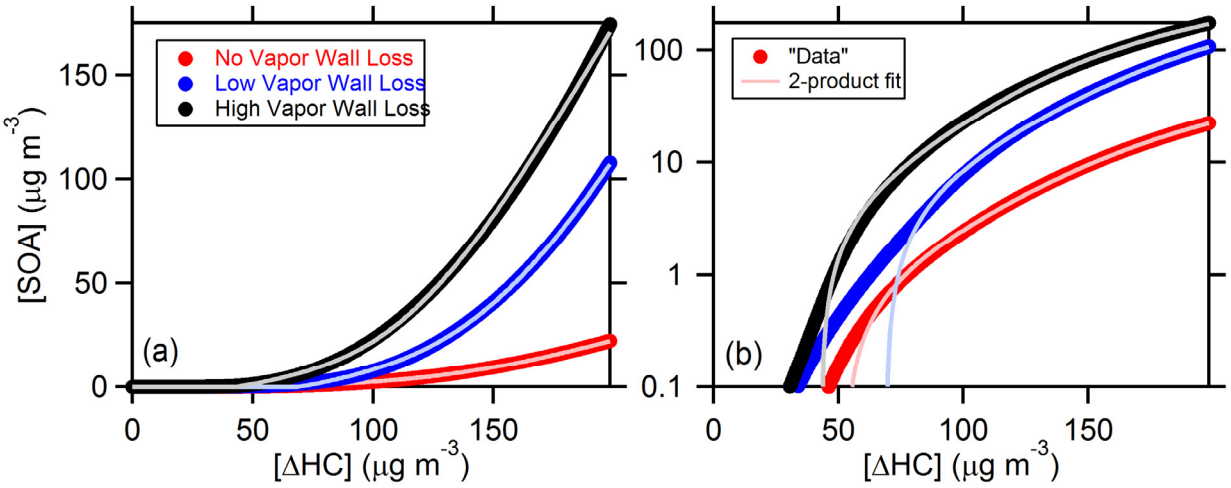

Figure S2. Example of 2-product fitting to SOA yield curves for dodecane + OH SOA formed under low-NO_x conditions. The 2-product model was fit to simulated vapor wall-loss-corrected yield curves (circles) that were generated using the SOM model. The original SOM fits were performed using variable k_{wall} values to account for vapor wall losses, but the subsequent simulated yield curves were generated with k_{wall} = 0. The lines are colored according to the wall-loss condition used when SOM was fit to the chamber observations, no wall loss (red), low wall loss (blue) and high wall loss (black).The best 2-product fits are shown as solid lines. Panel (a) shows the curves and fits on a linear scale and panel (b) shows the same on a log scale. Note that on a linear scale the deviations between the fit curves and the "data" at low [SOA] is not visibly evident.

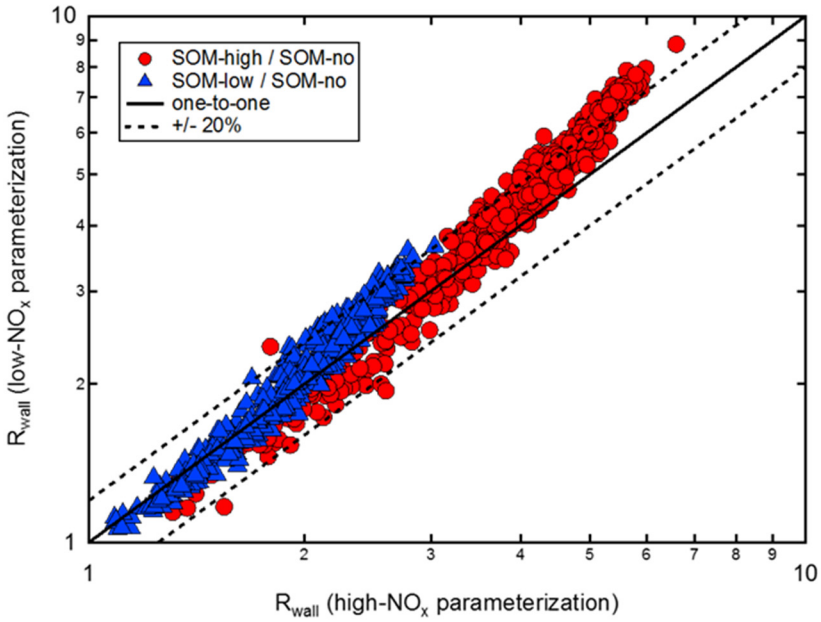

Figure S3. Comparison of R_{wall} values calculated for the low-NO_x parameterization (y-axis) or high-NO_x parameterization (x-axis) for the low vapor wall loss case (blue triangles) and high vapor wall loss case (red circles). The solid black line shows the 1-to-1 relationship and the dashed black lines the +/- 20% deviation from the 1-to-1 line.

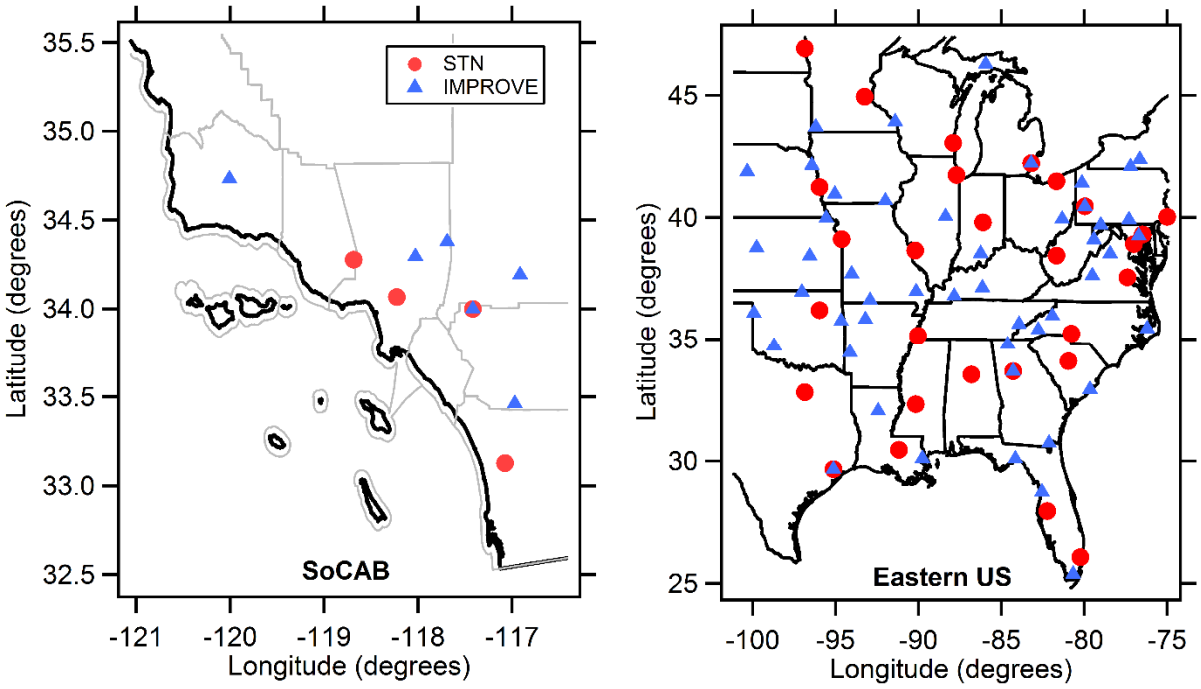

Figure S4. Map of STN and IMPROVE sites in the (left) SoCAB and (right) eastern US. STN sites are shown as red circles and IMPROVE sites as blue triangles.

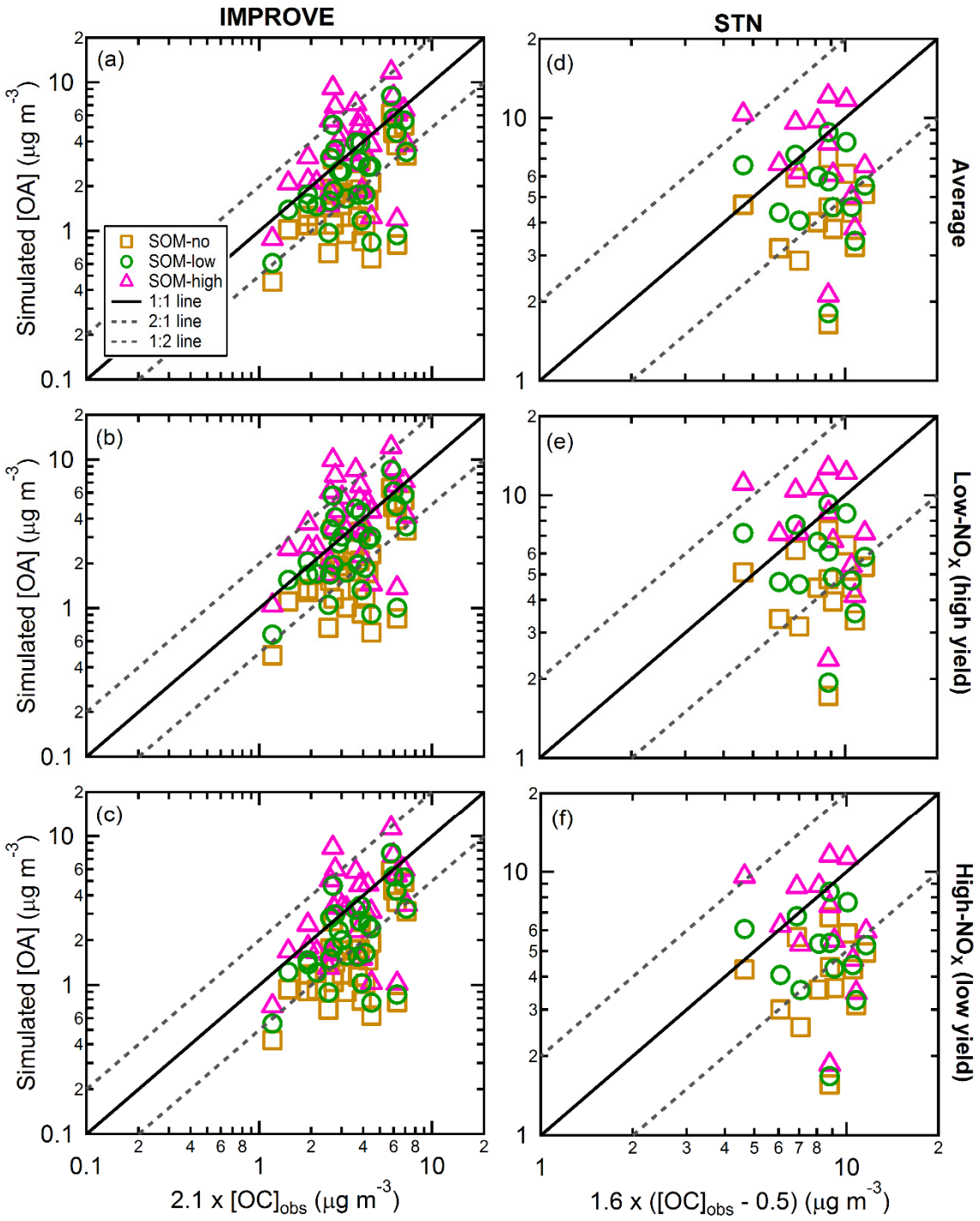

Figure S5. Scatter plots of simulated versus observed total OA (SOA + POA) concentrations for SoCAB for (left panels) IMPROVE and (right panels) STN sites. Simulation results are shown for SOM-no (orange), SOM-low (green) and SOM-high (pink). Results are reported from simulations run using the (top) average, (middle) low-NO_x / high-yield, and (bottom) high-NO_x / low-yield parameterizations.

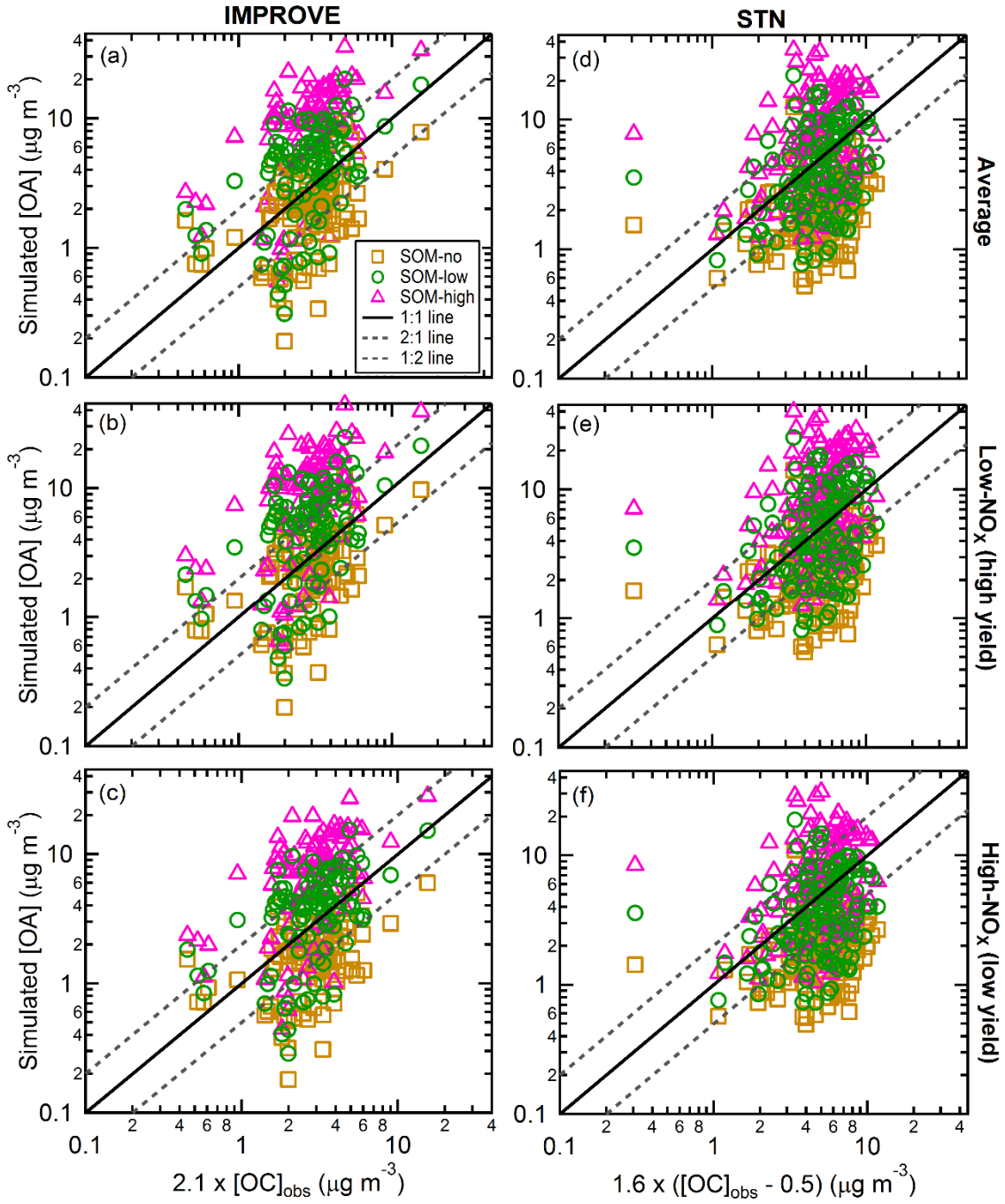

Figure S6. Scatter plots of simulated versus observed total OA (SOA + POA) concentrations for SoCAB for (left panels) IMPROVE and (right panels) STN sites. Simulation results are shown for SOM-no (orange), SOM-low (green) and SOM-high (pink). Results are reported from simulations run using the (top) average, (middle) low-NO_x / high-yield, and (bottom) high-NO_x / low-yield parameterizations. Only every other data point (one-in-two) is shown for visual clarity.

Table S1. List of best-fit SOM parameters determined by fitting SOM to experimental observations of SOA formation in the Caltech environmental chamber assuming that k_{wall} = 1 x 10^{-4} s^{-1} or 2.5 x 10^{-4} s^{-1}.

VOC Precursor Class	SAPRC-11 Species Name	VOC Surrogate	NO_x	m_{frag}	ΔLVP	p_{1O}	p_{2O}	p_{3O}	p_{4O}	Ref.^
				k_{wall} = 1 x 10^{-4} s^{-1}						
Long Alkanes	ALK5*	dodecane	low	0.677	1.57	0.97	0.023	0.003	0.004	(Cappa et al., 2013; Loza et al., 2014)
			high	0.186	1.45	0.961	0.001	0.002	0.036	
Benzene	Benzene	benzene	low	0.01	2.31	0.324	0.001	0.607	0.068	(Ng et al., 2007)
			high	0.73	1.47	0.018	0.001	0.981	0.001	
Toluene	ARO1	toluene	low	0.843	1.70	0.066	0.001	0.106	0.827	(Zhang et al., 2014)
			high	5	1.37	0.865	0.001	0.065	0.069	
m-xylene	ARO2	m-xylene	low	0.236	1.97	0.001	0.123	0.8	0.075	(Ng et al., 2007)
			high	0.0389	1.46	0.001	0.001	0.905	0.093	
Isoprene	Isoprene	isoprene	low	0.01	2.20	0.097	0.13	0.748	0.025	(Chhabra et al., 2011)
			high	0.745	2.15	0.808	0.189	0.002	0.001	
Terpenes	TRP1/ SESQ+	α-pinene	low	0.156	1.89	0.316	0.554	0.087	0.043	(Chhabra et al., 2011)
			high	0.0588	1.92	0.064	0.865	0.063	0.008	
				k_{wall} = 2.5 x 10^{-4} s^{-1}						
Long Alkanes	ALK5*	dodecane	low	2	1.83	0.999	0.001	0.001	0.001	(Cappa et al., 2013; Loza et al., 2014)
			high	0.266	1.47	0.965	0.001	0.002	0.032	
Benzene	Benzene	benzene	low	0.0807	1.97	0.637	0.001	0.002	0.360	(Ng et al., 2007)
			high	0.824	1.53	0.008	0.001	0.991	0.001	
Toluene	ARO1	toluene	low	1.31	1.77	0.185	0.001	0.002	0.812	(Zhang et al., 2014)
			high	4.61	1.42	0.856	0.001	0.002	0.141	
m-xylene	ARO2	m-xylene	low	1.08	2.05	0.102	0.001	0.878	0.019	(Ng et al., 2007)
			high	0.0671	1.46	0.001	0.001	0.942	0.056	
Isoprene	Isoprene	isoprene	low	0.0839	2.44	0.096	0.379	0.518	0.007	(Chhabra et al., 2011)
			high	5	1.78	0.874	0.039	0.085	0.001	
Terpenes	TRP1/ SESQ+	α-pinene	low	0.305	1.97	0.419	0.426	0.140	0.014	(Chhabra et al., 2011)
			high	0.16	1.91	0.500	0.422	0.070	0.008	

^These are the primary references for the experimental data. The data for the specific experiments used are presented in the supplemental material of (Zhang et al., 2014)

*For SOM, the ALK5 class is separated into long alkane species grouped according to carbon number. See (Jathar et al., 2015) for details.

+Although the same set of parameters are used to describe the formation of oxidation products and SOA from monoterpenes and sesquiterpenes, the SOA yield from sesquiterpenes is larger than for monoterpenes due to the larger number of carbon atoms comprising sesquiterpenes.

Table S2. Comparison between calculated non-fossil fractions of secondary organic aerosol (SOA) and secondary organic carbon (SOC).

Vapor Wall Loss Case	NO_x condition				
		Central LA		Riverside	
		SOA	SOC	SOA	SOC
SOM-no	high-NO_x	0.27	0.24	0.28	0.25
	low-NO_x	0.44	0.41	0.40	0.37
SOM-low	high-NO_x	0.22	0.23	0.27	0.28
	low-NO_x	0.32	0.30	0.35	0.33
SOM-high	high-NO_x	0.22	0.25	0.28	0.31
	low-NO_x	0.33	0.32	0.37	0.36
		Atlanta		Smokey Mountains	
		SOA	SOC	SOA	SOC
SOM-no	high-NO_x	0.10	0.08	0.14	0.12
	low-NO_x	0.17	0.15	0.15	0.13
SOM-low	high-NO_x	0.19	0.18	0.27	0.27
	low-NO_x	0.18	0.17	0.22	0.20
SOM-high	high-NO_x	0.25	0.27	0.32	0.35
	low-NO_x	0.20	0.19	0.24	0.23

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