

Supporting information for ‘Unspeciated organic emissions from combustion sources and their influence on the secondary organic aerosol budget in the United States’

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Experiments

Details of the experiments and experimental procedures are described in companion publications (1-5). Briefly, combustion emissions were handled into two stages. The primary emissions were characterized after the first stage of dilution; the smog chamber experiments of secondary organic aerosol (SOA) formation were performed after the second stage of dilution.

In the first stage, emissions were lightly (<20:1) diluted in a dilution tunnel (6, 7) or injected into a larger burn chamber (1). At these low levels of dilution the organic aerosol concentrations were typically greater than 100 $\mu\text{g}/\text{m}^3$ for the vehicle tests and greater than 1 mg/m^3 for the biomass burning experiments. Here, the mass of total organic gas was measured using a flame ionization detector calibrated with propane. Canister or bag samples were analyzed offline using conventional gas-chromatography techniques (1, 5). This analysis identified (speciated) emissions of single/double-ring aromatics and smaller (< C_{12}) alkanes (straight, branched and cyclic), alkenes and carbonyls (formaldehyde, acetaldehyde, etc). Organic carbon (or primary organic aerosol; POA) measurements were made using an OC/EC analyzer.

The second stage of dilution involved transferring emissions from the dilution tunnel or burn chamber into a smog chamber (1, 2, 4). This involved substantial additional dilution (~30:1) with clean air. Since the smog-chamber experiment (second stage) was run at a much higher dilution, some POA evaporated between the first and second stage, increasing gas-phase SOA precursor concentrations. The mass of evaporated POA was calculated as the difference between the dilution corrected POA between the two stages (6-8) and considered part of the unspeciated organic emissions. Inside the smog chamber the organic aerosol concentrations were representative of those typically found in urban areas or emission plumes (1 to 100 $\mu\text{g m}^{-3}$).

To initiate photo-oxidation, the smog chamber was exposed to natural or artificial sunlight. Inside the smog chamber, four to six different gas-phase organics were measured online with a proton transfer reaction mass spectrometer (PTR-MS); the decay of these organics and their reaction rates with the hydroxyl radical (OH) were used to calculate OH concentrations. Particle-phase measurements were made online with a scanning mobility particle sizer (SMPS) and aerosol mass spectrometer (AMS). Both SMPS and AMS measurements were used to estimate wall-loss corrected concentrations of POA and SOA. The measured SOA was defined as the increase in wall-loss corrected mass during the photo-oxidation phase of the experiment.

Sources

Gasoline vehicles: Experiments were performed with 15 different light-duty gasoline vehicles recruited from the California in-use fleet (model years 1987 to 2011). The fleet was not designed to represent the distribution of vehicles in the current, in-use California fleet; instead, vehicles (from private owners located within 50 miles of the laboratory) were selected to span a wide range of model years, vehicle types, engine technologies and emission control technologies. All vehicles were port fuel injected except for one LEV-II, which was an early generation gasoline direct injection vehicle. For discussion, the vehicles are grouped based on model year: “pre-LEV” were vehicles manufactured prior to 1995; “LEV-I” vehicles were manufactured between 1995 and 2003; and “LEV-II” vehicles were manufactured 2004 or later. In this work, the LEV designation simply refers to a range of model years; it does not refer to the emissions certification standard. The certification standard and other details for each vehicle are listed in the table below. All of the vehicles were operated on the same California commercial summertime gasoline. Every vehicle was tested using the cold-start Unified Cycle (UC) driving schedule, which was designed to simulate driving patterns in Southern California. It has a similar three-bag structure as the Federal Test Procedure (FTP), but is a more aggressive cycle with higher speeds, higher acceleration, fewer stops per mile and less idle time. Additional details on the gasoline vehicle testing are in Gordon et al. (2).

Designation	Model Year	Vehicle Class	Engine Size (L)	Emission Standard	Mileage
Pre-LEV	1987	Passenger Car	4.1	Tier I	197,631
Pre-LEV	1988	Passenger Car	1.6	Tier I	224,758
Pre-LEV	1990	SUV	5.0	Tier I	58,617
LEV-I	1996	Passenger Car	2.7	Tier I	51,826
LEV-I	1997	Passenger Car	3.0	LEV	130,485
LEV-I	1998	Passenger Car	3.0	LEV	90,638
LEV-I	1999	Passenger Car	2.0	TLEV	118,294
LEV-I	2000	Passenger Car	2.2	LEV I, ULEV	104,446
LEV-I	2003	Passenger Car	3.5	LEV I, NLEV	110,445
LEV-II	2007	SUV	3.9	LEV II	29,433
LEV-II	2008	Light duty truck	4.2	LEV II	43,378
LEV-II	2008	Passenger Car	3.5	LEV II	35,786
LEV-II	2010	Light duty truck	3.6	ULEV; Tier II	18,236
LEV-II	2011	Passenger Car	2.0	ULEV	10,911

LEV-II	2011	Passenger Car	3.6	LEV II, ULEV	29,249
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Diesel Vehicles: Experiments were performed with three diesel vehicles without diesel particulate filters. One was a 2006 heavy-duty (class 8) diesel tractor equipped with a 10.8L, 6 cylinder, in-line, direct injection, turbocharged diesel engine. This tractor had been driven 94,000 miles at the time of testing. The heavy duty diesel vehicle experiments were conducted using the Urban Dynamometer Driving Schedule (UDDS), which represents urban operation of heavy duty diesel vehicles. There were two medium duty diesel vehicles (MDDV) that were full-sized pickup trucks with gross vehicle weight ratings between 8,500 and 9,500 lbs. One was a 2005 MDDV equipped with a 6.6L, V8 turbocharged engine and a diesel oxidation catalyst (DOC). This vehicle had been driven 65,934 miles at the time of testing. The other was a 2001 MDDV equipped with a 5.9L, 6 cylinder, in-line, turbocharged diesel engine with no after-treatment. This vehicle had been driven 158,850 miles at the time of testing. The MDDVs were tested using the cold-start Unified Cycle (UC). The vehicles were tested with a commercial ultra-low-sulfur diesel that met California specifications. Additional details on the diesel vehicle testing are in Gordon et al. (4).

Biomass Fuels: Experiments were performed as part of the FLAME-III study at the U.S. Forest Service Fire Science Laboratory (FSL) in Missoula, MT. The fuels are listed in the table below. The fuels represent different regions in North America commonly impacted by prescribed burning and wildfires, including the Southeast (e.g., gallberry and pocosin), southern California (e.g., sagebrush and chamise) and forest regions of the western United States and Canada (e.g., ponderosa pine, lodgepole pine, and black spruce). Additional details on the biomass burning experiments are in Hennigan et al. (1)

Fuel	Name	Material
Alaskan Duff	Hylocomium splendens, Sphagnum spp.	decayed vegetation matter
Black Spruce	Picea mariana	branches, leaves
Chamise	Adenostoma fasciculatum	branches, leaves
Gallberry	Ilex glabra	branches, leaves
Lodgepole Pine	Pinus contorta	branches, leaves
Pocosin	Lyonia lucida, Pinus serotina	shrub branches, leaves; dried pine litter
Ponderosa Pine	Pinus ponderosa	branches, leaves
Sage	Artemisia tridentata	branches, leaves
Saw Grass	Cladium mariscus	leaves (grass-like)
Turkey Oak	Quercus laevis	leaves
White Spruce	Picea glauca	branches, leaves
Wire Grass	Aristida beyrichiana	grass

Speciated-SOA Model

SOA precursors (benzene, cyclodecane, m-xylene, etc) are lumped into SAPRC07 model species (9) and then allowed to react with the OH radical to form SOA as per Carlton et al. (10). The reaction is semi-empirically represented using a set of semi-volatile surrogate products in

the volatility basis set (VBS) (11). The loss of the precursor and production of semi-volatile species are described using the following equations:

$$\frac{d[X_j]}{dt} = -k_{OH,X_j}[OH][X_j] \quad (1)$$

$$\frac{d[M_i|_{g+p}]}{dt} = \sum_j \alpha_{i,j} k_{OH,X_j}[OH][X_j] \quad (2)$$

Equation (1) represents the first-generation oxidation of an SOA precursor where $k_{OH,X}$ is the reaction rate constant between the oxidant $[OH]$ and SOA precursor $[X]$. The index j indicates different precursors. Equation (2) tracks the total (gas+particle) concentration of the semi-volatile products, $M_i|_{g+p}$. The index i indicates different C^* bins in the VBS. The mass yields, $\alpha_{i,j}$ are those used by Carlton et al. (10) and listed in Table S4 below. The OH concentration used to evaluate equations (1) and (2) were derived from the measured decay of individual organics. The VOC/ NO_x influence on the SOA yields for speciated organics is parameterized using Presto et al. (12):

$$\alpha = \beta \alpha_{low NO_x} + (1 - \beta) \alpha_{high NO_x}$$

$$\beta = \frac{VOC}{NO_x} \Big|_{t=0} \quad \text{if} \quad \frac{VOC}{NO_x} \Big|_{t=0} < 10 \quad (3)$$

$$\beta = 1 \quad \text{if} \quad \frac{VOC}{NO_x} \Big|_{t=0} \geq 10$$

The amount of SOA at equilibrium is defined by the gas-particle partitioning of the surrogate products.

$$\xi_i = \left(1 + \frac{C_i^*}{C_{OA}} \right)^{-1} \quad (4)$$

$$C_{OA} = \sum_{i=1}^N \xi_i \times M_i|_{g+p}$$

where, ξ_i is the fraction of mass of $M_i|_{g+p}$ in the particulate phase, C_i^* is the effective saturation concentration of M_i and C_{OA} is the total particulate OA concentration.

Although important in the atmosphere, the model does not explicitly account for multigenerational aging because the SOA mass yields used in Carlton et al. (10) were derived from smog chamber experiments that span similar lengths in reaction times and OH exposure. Therefore, the yields in Carlton et al. (10) should account for some multi-generational aging, comparable to what occurred in our experiments.

Measurement Uncertainty/Variability

Figure 1 indicates that there is significant variability in POA and NMOG emissions and SOA formation within each source category. This variability is mainly driven by source-to-source differences in emissions, not experimental uncertainty. The level of variability is not surprising given the diversity of sources (e.g. a fleet of 15 gasoline vehicles manufactured over a 20+ year period with different engine and emissions control technology to meet very different emissions standards). This sort of variability is apparent in any large compilation of emissions data.

Repeat experiments were performed with the same source to empirically characterize the experimental uncertainty. These results are described in detail in the companion publications. For example, May et al. (6, 7) describe the repeatability of the primary emissions for the gasoline and diesel fleet. Emissions of gas-phase pollutants typically agreed to within ~10% (relative percent difference between repeated tests for the same vehicle), while repeated measurements of particle-phase pollutants were somewhat more variable (~25%). The largest variability was observed with low emitting vehicles. Gordon et al. (2, 4) presented results from ten sets of duplicate chamber experiments. There is good agreement ($\pm 25\%$ of the average of each duplicate pair) in SOA production for 7 of the 10 pairs of duplicate experiments. In two of the paired sets of experiments with higher experiment-to-experiment variability, the variability was due to actual differences in emissions. That vehicle had a malfunctioning oxygen sensor, which resulted in highly variable and high emissions. In other words the experiments were not true repeats. The final paired experiment with higher experiment-to-experiment variability was on a very low emitting vehicle. The bottom line is that the variability in repeat experiments with the same source is much smaller than the order of magnitude variability shown in Figure 1. Therefore, the scatter shown in Figure 1 is due to source-to-source differences, not experimental uncertainty.

For the on-road gasoline source, pre-LEV vehicles have higher POA, SOA and NMOG emissions/production than LEV-I and LEV-II vehicles and LEV-I vehicles have higher NMOG emissions than LEV-II vehicles. However, even within a given class of vehicles (e.g. LEV-I) there is a wide variation in emission rates depending on the engine/emissions control system. Except for those two, there are no trends in POA, SOA and NMOG emissions/production with engine certification (LEV-type), with vehicle size (medium-duty versus heavy-duty) or biomass fuel type (trees/pines, grasses, shrubs/duff). The biomass burning data are more variable than the on-road gasoline and on-road diesel data because of the poorly controlled nature of biomass burning and the fact that the fuel types for biomass burning were very different.

Model Uncertainty

In our model, the following variables are resolved for each experiment based on measurements: composition of speciated organics, mass of unspeciated organics, OH exposure and dependency of speciated organic's SOA yields on the VOC-to-NO_x ratio. For the model that simulates SOA formation from both speciated and unspeciated organics, there are two known sources of uncertainty. The first source of uncertainty comes from the assumption that the composition of the unspeciated emissions and therefore its propensity to form SOA remains the same for all sources in a given source category. This is likely a poor assumption because the composition of the speciated SOA precursors (lumped SAPRC model species) varied significantly from source to source – for example the speciated SOA precursors varied by $\pm 57\%$ ($\pm 1\sigma$) for the on-road diesel sources. It is likely that the composition of the unspeciated emissions varied by a similar amount and resulted in proportionally different amounts of SOA formation. Since our parameterization is intended to represent the average source-class behavior of the unspeciated organics, we expect it to remove the bias in the data. This is critical for chemical transport modeling and emissions inventories, which represent the behavior of emissions from a large set of sources, not an individual source. The parameterization will only eliminate the scatter if the unspeciated organics from all sources within a category are the same. We attribute much of the scatter in Figure S2 to source-to-source variability in the composition of the unspeciated organic emissions.

The second source of uncertainty comes from the SOA yields used to model SOA formation from speciated organics. A comparison of SOA yield data from published single precursor smog chamber experiments suggests that these yields are uncertain within a factor of two. Note that this factor of two accounts only for the variability in SOA yields between studies, not potential systematic biases such as loss of condensable vapors to the chamber walls (13) which have not been accounted for by smog chamber studies. To cite a few examples that illustrate the variability in single precursor data, Hildebrandt et al. (14) found that the estimated SOA mass yields for toluene from half a dozen experiments varied within a factor of two. When compared against Ng et al. (15), the average SOA mass yields were a factor of two higher. Similarly, SOA mass yields for similar experiments on naphthalene from Chan et al. (16) and Shakya and Griffin (17) vary by a factor of two. If we assume that the composition of emissions varied by $\pm 50\%$ and the SOA yields are uncertain within a factor of two ($\pm\sqrt{2}$), model predictions would have an uncertainty slightly larger than a factor of two (illustrated as a band in Figure S2).

There are several other sources of uncertainty: dependency of unspciated organic's SOA yields on the VOC-to-NO_x ratio, absolute concentrations of VOCs and NO_x, relative proportions of NO, NO₂, RO₂ and HO₂ radicals. These are probably important in modeling SOA formation and would need to be included in future modeling efforts to explain the measured variability in SOA formation better.

Although the model predictions for an individual source are uncertain by more than a factor of two, the model predictions for the average source are less uncertain. To derive an uncertainty estimate for the average source, we fit the ratio of predicted to measured SOA for all the experiments to a lognormal distribution. For the updated model, the median ratio was 1 and the geometric standard deviation was 1.6, which corresponds to a 5 to 95% confidence interval of -40% to +60% for the average estimate. This uncertainty estimate for SOA is applied to the model prediction in Figure 4.

Primary PM_{2.5} emissions

Primary PM_{2.5} emissions are from the National Emissions Inventory (NEI) for 2008. These emissions were speciated into primary organic carbon (POC) using profiles from EPA's SPECIATE database (18); POC was converted to POA using an organic-matter-to-organic-carbon ratio of 1.4. For off-road gasoline, residential wood combustion, and off-road diesel a single profile was used for each lumped source category. For both on-road categories, tailpipe was the largest source of PM_{2.5} (76-96%) followed by brake wear (3-19%) and then tire wear (1-5%). On-road diesel tailpipe emissions included profiles for both heavy duty (92035) and light duty (92042) vehicle exhaust. The 2008 NEI estimated that 97% of on-road diesel PM_{2.5} comes from heavy duty vehicles (the remainder being light duty).

The speciation profiles used in this work are listed in Table S7 and the actual speciation is in Table S8. Speciation is based on the CMAQ v5.0 AERO6 aerosol module (19).

NMOG emissions

On-road gasoline: On-road gasoline emissions were calculated using the MOVES model. The emissions included contributions from tailpipe (65%), evaporation (24%), refueling (6%) and evaporative permeation (5%). The NMOG emissions were speciated using profiles 8750/8751 for tailpipe, 8753/8754 for evaporation, 8869/8870 for refueling and 8766/69 for evaporative permeation.

Off-road gasoline: Off-road gasoline vehicle emissions included contributions from tailpipe (76%), evaporative (17%) and refueling (7%). The NMOG emissions were speciated using profiles 8750/8751 for tailpipe, 8753/8754 for evaporation and 8869/8870 for refueling.

On-road diesel: On-road diesel emissions, also calculated with MOVES, included contributions from tailpipe (95%) and refueling (5%). The NMOG emissions were speciated using profiles 8774 for tailpipe and 4547 for refueling.

Off-road diesel: Off-road diesel emissions were only from tailpipe sources. All off-road diesel NMOG were speciated using profile 4674 except for agricultural tractors (SCC 2770005015) which were speciated using profile 3161. Agricultural tractors accounted for 28% of off-road diesel NMOG emissions.

Residential wood combustion: NMOG from residential wood combustion was speciated using one of two profiles in the emissions inventory: 4642 or 1084. Only EPA certified catalytic woodstoves (SCC 2104008230 and 2104008330) used profile 1084 and accounted for about 3% of residential wood combustion NMOG.

Biomass burning: Biomass burning included agricultural field burning, prescribed fires, and wildfires. The NMOG from wildfires and prescribed burns was speciated using profile 5560 while agricultural fires were speciated using profile 8746.

The speciation profiles used in this work are available in Table S7 and the actual speciation is in Table S9.

Tables

Table S1: Comparison of the 25th to 75th percentile of POA, SOA and NMOG data between this work and literature. Our estimates compare reasonably well with those in the literature, except for our on-road gasoline and on-road diesel SOA, which are somewhat lower than previous estimates. SOA production depends on extent of oxidation (OH exposure), total mass of NMOG emissions, VOC-to-NO_x ratios and OA concentrations. For example, higher SOA formation is measured from higher emitting vehicles.

Source	Reference	POA (mg kg-fuel ⁻¹)	SOA (mg kg-fuel ⁻¹)	NMOG (mg kg-fuel ⁻¹)
On-road gasoline	This work	3.2 - 6.7	20 - 67	507 - 1810
	Platt et al. (20) - Euro 5	4.8 - 44	345	840 - 1750
On-road diesel	This work	43 - 70	9.5 - 85	551 - 1530
	Chirico et al. (21) - Light duty diesels	81 - 147	19 - 254	NA
Biomass burning	This work	310 - 3310	124 - 1650	5760 - 35900
	Grieshop et al. (22), Heringa et al. (23) - Woodstoves	250 - 701	324 - 2050	NA

Species	9/19/09	9/20/09	9/21/09	9/22/09	9/23/09	9/24/09	9/25/09	9/26/09	9/28/09	9/29/09	9/30/09	10/1/09	10/2/09	10/4/09	10/5/09	10/6/09	10/7/09	10/8/09
	Lodgepole Pine	Lodgepole Pine	Ponderosa Pine	Wire Grass	Saw Grass	Turkey Oak	Gallberry	Sage	Alaskan Duff	Sage	White Spruce	Ponderosa Pine	Chamise	Lodgepole Pine	Pocosin	Gallberry	Black Spruce	Wire Grass
methacrolein	5.7E-03	5.7E-03	0.0E+00	2.9E-03	3.7E-03	3.8E-03	6.0E-03	6.1E-03	3.3E-03	7.3E-03	3.5E-03	4.5E-03	4.0E-03	6.5E-03	4.4E-03	4.1E-03	2.1E-03	3.3E-03
methanol	1.2E-01	1.2E-01	3.4E-01	3.3E-01	6.8E-02	9.9E-02	1.1E-01	9.7E-02	1.3E-01	1.2E-01	1.9E-01	1.8E-01	1.7E-01	1.6E-01	1.7E-01	1.5E-01	4.5E-02	8.5E-03
methyl ethyl ketone	8.2E-03	8.2E-03	8.3E-03	8.4E-03	8.7E-03	1.1E-02	1.0E-02	8.1E-03	1.6E-02	7.2E-03	1.2E-02	1.0E-02	1.4E-02	1.1E-02	1.8E-02	2.0E-02	2.6E-02	3.9E-02
methyl vinyl ketone	1.7E-02	1.7E-02	5.6E-03	1.3E-02	2.2E-03	1.0E-02	1.5E-02	1.1E-02	8.5E-03	8.0E-03	1.1E-02	1.2E-02	1.0E-02	2.1E-02	1.8E-02	8.2E-03	7.7E-03	1.1E-02
n-butane	3.3E-03	3.3E-03	5.4E-03	3.1E-03	2.4E-03	4.4E-03	9.5E-03	4.7E-03	7.1E-03	2.9E-03	4.6E-03	6.0E-03	3.5E-03	6.0E-03	1.2E-02	6.5E-02	1.4E-01	1.9E-01
n-pentane	2.0E-03	2.0E-03	3.9E-03	3.4E-03	2.0E-03	4.3E-03	6.1E-03	3.4E-03	3.9E-03	1.7E-03	2.7E-03	2.9E-03	1.9E-03	3.3E-03	6.9E-03	6.9E-03	5.8E-03	6.1E-03
propanal	1.2E-02	1.2E-02	1.0E-02	1.7E-02	3.3E-02	1.1E-02	1.3E-02	1.0E-02	1.5E-02	8.9E-03	1.1E-02	1.0E-02	8.0E-03	1.2E-02	1.3E-02	2.1E-02	2.4E-02	3.2E-02
propane	1.3E-02	1.3E-02	3.3E-02	1.9E-02	2.4E-02	2.2E-02	3.6E-02	2.6E-02	4.0E-02	2.0E-02	2.9E-02	3.3E-02	2.3E-02	2.8E-02	4.4E-02	6.0E-02	6.8E-02	0.0E+00
propene	1.0E-01	1.0E-01	5.9E-02	3.7E-02	5.5E-02	7.3E-02	8.5E-02	7.8E-02	7.7E-02	6.5E-02	6.4E-02	7.8E-02	7.0E-02	9.9E-02	9.7E-02	6.1E-02	4.9E-02	4.6E-02
propyne	5.6E-03	5.6E-03	2.7E-03	4.3E-03	4.1E-03	8.7E-03	5.7E-03	7.2E-03	1.6E-03	6.7E-03	2.9E-03	4.0E-03	7.1E-03	6.2E-03	5.7E-03	2.6E-03	1.2E-03	2.4E-03
t-2-butene	8.1E-03	8.1E-03	2.4E-03	7.8E-04	1.3E-03	4.4E-04	3.0E-03	1.7E-03	2.7E-03	1.2E-03	2.5E-03	2.6E-03	1.5E-03	2.2E-03	4.4E-03	2.4E-03	3.9E-03	0.0E+00
1-hexene	8.7E-04	8.7E-04	1.2E-03	4.7E-04	4.9E-04	5.7E-04	1.9E-03	7.2E-04	1.3E-03	5.7E-04	1.0E-03	1.4E-03	5.1E-04	2.0E-03	1.3E-03	1.5E-03	9.9E-04	0.0E+00
1-pentene	4.7E-03	4.7E-03	3.9E-03	1.6E-03	1.7E-03	6.1E-04	5.4E-03	2.7E-03	4.8E-03	2.0E-03	3.3E-03	4.0E-03	9.8E-04	1.2E-03	1.9E-03	4.1E-03	2.6E-03	0.0E+00
2-methyl-1-butene	1.7E-03	1.7E-03	2.0E-03	7.1E-04	8.8E-04	1.0E-03	2.3E-03	1.3E-03	1.6E-03	9.9E-04	1.8E-03	2.0E-03	8.5E-04	6.6E-04	1.2E-03	1.7E-03	9.6E-04	0.0E+00
t-2-pentene	1.8E-03	1.8E-03	1.6E-03	8.1E-04	1.1E-03	1.2E-03	2.4E-03	1.5E-03	1.6E-03	1.1E-03	1.7E-03	1.9E-03	9.5E-04	9.2E-04	1.6E-03	1.5E-03	1.3E-03	0.0E+00
styrene	3.4E-03	3.4E-03	1.3E-03	1.3E-03	5.9E-04	3.4E-03	2.4E-03	3.0E-03	1.5E-03	2.8E-03	2.5E-03	3.1E-03	5.1E-03	7.1E-03	3.4E-03	1.5E-04	1.6E-04	0.0E+00
o-xylene	1.0E-03	1.0E-03	9.3E-04	3.7E-04	4.2E-04	7.3E-04	1.4E-03	8.4E-04	9.7E-04	6.8E-04	1.2E-03	1.1E-03	1.2E-03	2.2E-03	2.3E-03	9.3E-04	9.5E-04	0.0E+00
decane	2.3E-04	2.3E-04	5.7E-04	2.5E-04	2.2E-04	2.6E-04	3.8E-04	4.2E-04	4.0E-04	3.6E-04	4.9E-04	4.3E-04	2.6E-04	6.1E-04	8.5E-04	1.4E-03	9.7E-04	0.0E+00
camphene	1.4E-04	1.4E-04	5.8E-04	0.0E+00	3.2E-04	2.8E-04	0.0E+00	1.4E-03	1.6E-04	2.6E-03	7.5E-02	6.1E-04	6.6E-04	4.7E-04	3.0E-04	0.0E+00	1.3E-02	0.0E+00
toluene	1.9E-02	1.9E-02	1.6E-02	5.9E-03	7.8E-03	1.4E-02	1.5E-02	1.5E-02	2.0E-02	1.3E-02	1.6E-02	2.1E-02	2.3E-02	2.9E-02	2.4E-02	9.1E-03	8.0E-03	6.6E-03

Table S3: SOA surrogates and VBS SOA mass yields (24) used for unspciated organics from different source categories.

Source	SOA surrogate	C* ($\mu\text{g m}^{-3}$)			
		0.1	1	10	100
Biomass burning/Wood burning	<i>n</i> -pentadecane (C ₁₅)	0.044	0.071	0.41	0.30
On- and off-road gasoline	<i>n</i> -tridecane (C ₁₃)	0.014	0.059	0.22	0.4
On- and off-road diesel	<i>n</i> -pentadecane (C ₁₅)	0.044	0.071	0.41	0.30

Table S4: VBS SOA mass yields used in this work for the SAPRC07 species. The yields are the same as those in Carlton et al. (10) but fit to a five-product VBS.

SAPRC07 species	High NO _x (Low Yield)					Low NO _x (High Yield)
	C* ($\mu\text{g/m}^3$)					C* ($\mu\text{g/m}^3$)
	0.1	1	10	100	1000	non-volatile
ALK5	0.0736	0.0000	0.0000	0.0000	0.0000	-
BENZ	0.0534	0.0402	0.0000	1.0551	0.2386	0.4840
ARO1	0.0002	0.0279	0.1606	0.0409	0.0000	0.4710
ARO2	0.0055	0.0121	0.0760	0.0652	0.0000	0.3730
ISOP	0.0067	0.0218	0.0000	0.2025	0.0654	-
SESQ	0.0000	0.0000	0.7811	0.9518	0.0000	-
TERP	0.0000	0.0000	0.0926	0.4488	0.0880	-

Table S5: NMOG totals (ton/yr) from the 2008 NEI version 3 for the United States. Obtained on September 23, 2013 from the EPA website: <http://www.epa.gov/ttn/chief/net/2008inventory.html>

Sector	NMOG emission (ton/yr)
Biogenics - Vegetation and Soil	38,909,250
Fires - Wildfires	2,846,634
Mobile - On-Road Gasoline Light Duty Vehicles	2,659,763
Mobile - Non-Road Equipment - Gasoline	2,242,472
Fires - Prescribed Fires	1,693,249
Industrial Processes - Oil & Gas Production	1,688,384
Solvent - Consumer & Commercial Solvent Use	1,619,338
Solvent - Industrial Surface Coating & Solvent Use	647,913
Gas Stations	643,143
Solvent - Non-Industrial Surface Coating	429,183
Fuel Comb - Residential - Wood	374,612
Solvent - Graphic Arts	356,044
Industrial Processes - Storage and Transfer	240,178
Miscellaneous Non-Industrial NEC	226,996
Industrial Processes - NEC	214,754
Mobile - On-Road Diesel Heavy Duty Vehicles	212,658
Solvent - Degreasing	198,260
Waste Disposal	180,883
Mobile - On-Road Gasoline Heavy Duty Vehicles	169,193

Sector	NMOG emission (ton/yr)
Mobile - Non-Road Equipment - Diesel	165,198
Industrial Processes - Pulp & Paper	129,910
Industrial Processes - Chemical Manuf	100,311
Bulk Gasoline Terminals	92,800
Agriculture - Livestock Waste	92,535
Industrial Processes - Petroleum Refineries	68,070
Fuel Comb - Industrial Boilers, ICEs - Natural Gas	59,463
Fires - Agricultural Field Burning	55,088
Solvent - Dry Cleaning	48,939
Mobile - Non-Road Equipment - Other	46,480
Mobile – Locomotives	44,198
Mobile – Aircraft	33,165
Fuel Comb - Electric Generation - Coal	28,676
Industrial Processes - Ferrous Metals	19,333
Industrial Processes - Non-ferrous Metals	16,213
Mobile - Commercial Marine Vessels	14,406
Fuel Comb - Residential - Natural Gas	12,706
Commercial Cooking	12,261
Mobile - On-Road Diesel Light Duty Vehicles	10,190
Fuel Comb - Electric Generation - Natural Gas	9,335
Industrial Processes - Cement Manuf	9,237
Fuel Comb - Comm/Institutional - Natural Gas	9,177
Fuel Comb - Industrial Boilers, ICEs - Biomass	8,380
Fuel Comb - Industrial Boilers, ICEs - Other	6,358
Fuel Comb - Industrial Boilers, ICEs - Oil	3,838
Fuel Comb - Residential – Other	2,904
Fuel Comb - Comm/Institutional - Oil	2,687
Fuel Comb - Electric Generation - Oil	2,568
Fuel Comb - Industrial Boilers, ICEs - Coal	2,121
Industrial Processes – Mining	2,012
Fuel Comb - Electric Generation - Other	1,989
Fuel Comb - Residential – Oil	1,743
Fuel Comb - Electric Generation - Biomass	1,039
Fuel Comb - Comm/Institutional - Other	894
Fuel Comb - Comm/Institutional - Biomass	535
Fuel Comb - Comm/Institutional - Coal	423
Dust - Construction Dust	17
Total	56,668,106

Table S6: Primary PM_{2.5} totals (filterable and condensable in ton/yr) from the 2008 NEI version 3 for the United States. Obtained on September 23, 2013 from the EPA website: <http://www.epa.gov/ttn/chief/net/2008inventory.html>

Sector	PM_{2.5} emissions (ton/yr)
Fires – Wildfires	998,604
Agriculture - Crops & Livestock Dust	923,495
Dust - Unpaved Road Dust	812,438
Fires - Prescribed Fires	696,365
Fuel Comb - Residential - Wood	352,997
Dust - Paved Road Dust	280,419
Fuel Comb - Electric Generation - Coal	275,267
Dust - Construction Dust	219,923
Waste Disposal	207,737
Mobile - On-Road Diesel Heavy Duty Vehicles	160,256
Mobile - Non-Road Equipment - Diesel	122,637
Industrial Processes - NEC	117,237
Industrial Processes - Mining	106,638
Mobile - On-Road Gasoline Light Duty Vehicles	83,307
Commercial Cooking	78,162
Fires - Agricultural Field Burning	67,943
Mobile - Non-Road Equipment - Gasoline	57,156
Industrial Processes - Pulp & Paper	40,388
Industrial Processes - Ferrous Metals	35,542
Fuel Comb - Industrial Boilers, ICEs - Biomass	32,059
Fuel Comb - Industrial Boilers, ICEs - Other	31,233
Fuel Comb - Industrial Boilers, ICEs - Natural Gas	29,175
Mobile – Locomotives	25,435
Mobile - Commercial Marine Vessels	25,374
Industrial Processes - Storage and Transfer	24,928
Fuel Comb - Industrial Boilers, ICEs - Coal	24,223
Industrial Processes - Petroleum Refineries	23,471
Industrial Processes - Chemical Manuf	22,357
Industrial Processes - Non-ferrous Metals	20,088
Fuel Comb - Electric Generation - Natural Gas	19,891
Industrial Processes - Cement Manuf	13,082
Fuel Comb - Electric Generation - Oil	10,912
Agriculture - Livestock Waste	7,580
Fuel Comb - Industrial Boilers, ICEs - Oil	7,276
Industrial Processes - Oil & Gas Production	7,107
Fuel Comb - Comm/Institutional - Natural Gas	5,922
Fuel Comb - Residential – Oil	5,866
Fuel Comb - Residential - Natural Gas	5,056

Sector	PM_{2.5} emissions (ton/yr)
Mobile - On-Road Diesel Light Duty Vehicles	4,825
Fuel Comb - Comm/Institutional - Oil	4,741
Mobile - On-Road Gasoline Heavy Duty Vehicles	4,215
Solvent - Industrial Surface Coating & Solvent Use	3,828
Mobile – Aircraft	3,656
Miscellaneous Non-Industrial NEC	3,181
Fuel Comb - Comm/Institutional - Biomass	2,513
Fuel Comb - Comm/Institutional - Coal	2,210
Mobile - Non-Road Equipment - Other	2,102
Fuel Comb - Electric Generation – Other	1,813
Fuel Comb - Electric Generation - Biomass	1,433
Fuel Comb - Residential – Other	1,071
Fuel Comb - Comm/Institutional – Other	557
Solvent - Graphic Arts	255
Bulk Gasoline Terminals	88
Solvent – Degreasing	69
Solvent - Dry Cleaning	16
Solvent - Consumer & Commercial Solvent Use	13
Gas Stations	11
Solvent - Non-Industrial Surface Coating	0
Total	6,014,143

Table S7: NMOG and PM_{2.5} totals and speciation profile numbers used in SMOKE-MOVES/CMAQ for the sources and its sub-categories considered in this study.

Source	NMOG Profile #	NMOG Emissions (Tg yr ⁻¹)	PM _{2.5} Profile #	POA Emissions (Tg yr ⁻¹)
Biomass burning				
wild fires/prescribed fires	5560	4.1	92102	0.99
agricultural fires	8746	0.04	92000	0.04
Wood burning				
residential wood combustion	4642	0.3	92068	0.24
catalytic woodstoves	1084	0.01		
On-road gasoline				
Tailpipe	8750/51	1.7	92050	0.05
evaporative/refueling	8753/54, 8869/70, 8766/69	0.9	NA	NA
brake/tire wear	NA	NA	92009, 92087	0.005
Off-road gasoline				
Tailpipe	8750/51	1.5	92113	0.035
evaporative/refueling	8753/54, 8869/70	0.5	NA	NA
On-road diesel				
Tailpipe	8774	0.2	92035/42	0.05
Refueling	4547	0.0	NA	NA
brake/tire wear	NA	NA	92009, 92087	0.002
Off-road diesel				
Tractors	3161	0.04	92035	0.03
Others	4674	0.11		

Table S8: PM_{2.5} speciation profiles from the SMOKE-MOVES for the AERO6 aerosol module in CMAQ (19).

Profile	Species	Weight Fraction
92000	POC	0.388
92000	PEC	0.109
92000	PNH4	0.018
92000	PNO3	0.0035
92000	PSO4	0.0165
92000	PNCOM	0.272
92000	PNA	0.00655
92000	PMG	8.06E-04

Profile	Species	Weight Fraction
92000	PAL	3.00E-04
92000	PSI	1.50E-04
92000	PCL	0.0905
92000	PK	0.0704
92000	PCA	3.30E-04
92000	PTI	1.00E-05
92000	PFE	1.00E-04
92000	PMOTHR	0.023854

Profile	Species	Weight Fraction
92000	PMFINE	0.483
92009	POC	0.107
92009	PEC	0.0261
92009	PNH4	3.00E-05
92009	PNO3	0.0016
92009	PSO4	0.0334
92009	PNCOM	0.0428
92009	PH2O	0.00802
92009	PNA	1.60E-04
92009	PMG	0.111
92009	PAL	0.00124
92009	PSI	0.088
92009	PCL	0.00148
92009	PK	1.90E-04
92009	PCA	0.01
92009	PTI	0.0036
92009	PMN	0.00107
92009	PFE	0.115
92009	PMOTHR	0.44931
92009	PMFINE	0.8319
92035	POC	0.176
92035	PEC	0.771
92035	PNO3	0.00114
92035	PSO4	0.00295
92035	PNCOM	0.0439
92035	PCL	2.05E-04
92035	PK	3.80E-05
92035	PCA	5.83E-04
92035	PTI	4.00E-06
92035	PFE	2.62E-04
92035	PMOTHR	0.003918
92035	PMFINE	0.04891
92042	POC	0.355
92042	PEC	0.514
92042	PNH4	0.0073
92042	PNO3	0.0023
92042	PSO4	0.0086
92042	PNCOM	0.0888
92042	PNA	3.83E-04
92042	PMG	1.28E-04
92042	PAL	2.10E-04

Profile	Species	Weight Fraction
92042	PSI	0.00299
92042	PCL	2.04E-04
92042	PK	2.41E-05
92042	PCA	2.83E-04
92042	PMN	2.97E-06
92042	PFE	8.80E-04
92042	PMOTHR	0.018895
92042	PMFINE	0.1201
92049	POC	0.475
92049	PEC	0.122
92049	PNO3	7.00E-04
92049	PSO4	5.00E-04
92049	PNCOM	0.119
92049	PSI	0.0012
92049	PCA	3.00E-04
92049	PFE	1.00E-04
92049	PMOTHR	0.2812
92049	PMFINE	0.4018
92050	POC	0.549
92050	PEC	0.19
92050	PNH4	0.0167
92050	PNO3	0.00151
92050	PSO4	0.00808
92050	PNCOM	0.137
92050	PNA	0.00108
92050	PMG	5.29E-04
92050	PAL	0.00147
92050	PSI	0.00475
92050	PCL	7.96E-04
92050	PK	1.81E-04
92050	PCA	0.00288
92050	PTI	5.54E-05
92050	PMN	4.03E-05
92050	PFE	0.00404
92050	PMOTHR	0.081888
92050	PMFINE	0.25141
92068	POC	0.528
92068	PEC	0.0558
92068	PNH4	0.0015
92068	PNO3	0.0019
92068	PSO4	0.0041

Profile	Species	Weight Fraction
92068	PNCOM	0.37
92068	PNA	9.40E-04
92068	PMG	1.13E-04
92068	PAL	1.10E-04
92068	PSI	3.40E-04
92068	PCL	0.00297
92068	PK	0.00967
92068	PCA	1.00E-04
92068	PFE	9.00E-05
92068	PMOTHR	0.024367
92068	PMFINE	0.4102
92087	POC	0.472
92087	PEC	0.22
92087	PNH4	1.90E-04
92087	PNO3	0.0015
92087	PSO4	0.0311
92087	PNCOM	0.189
92087	PH2O	0.00751
92087	PNA	6.10E-04
92087	PMG	3.75E-04
92087	PAL	6.05E-04
92087	PSI	0.00115
92087	PCL	0.0078
92087	PK	3.80E-04

Profile	Species	Weight Fraction
92087	PCA	0.00112
92087	PTI	3.60E-04
92087	PMN	1.00E-04
92087	PFE	0.0046
92087	PMOTHR	0.0616
92087	PMFINE	0.2754
92090	POC	0.462
92090	PEC	0.0949
92090	PNH4	0.00879
92090	PNO3	0.00132
92090	PSO4	0.0126
92090	PNCOM	0.323
92090	PNA	0.00573
92090	PMG	3.14E-04
92090	PAL	6.08E-04
92090	PSI	0.00182
92090	PCL	0.0415
92090	PK	0.0294
92090	PCA	0.00386
92090	PTI	5.15E-05
92090	PMN	1.60E-05
92090	PFE	4.34E-04
92090	PMOTHR	0.013657
92090	PMFINE	0.42918

Table S9: TOG speciation profiles from SMOKE-MOVES for the SAPRC07 chemical mechanism in CMAQ (9, 25). In this work, we use an NMOG profile by removing methane from the TOG profile.

Profile	Species	SAPRC07 mechanism species	Weight fraction
1084	TOG	ALK1	0.0521
1084	TOG	ALK2	0.0137
1084	TOG	ALK3	9.09E-04
1084	TOG	ALK4	3.72E-04
1084	TOG	ALK5	0.0037817
1084	TOG	APIN	2.21E-04
1084	TOG	ARO1	4.07E-04
1084	TOG	ARO2	6.53E-04
1084	TOG	B124	4.50E-05
1084	TOG	BENZ	0.1891

Profile	Species	SAPRC07 mechanism species	Weight fraction
1084	TOG	CH4	0.3839
1084	TOG	CRES	3.65E-05
1084	TOG	ETHE	0.1638
1084	TOG	ETOH	0.1647
1084	TOG	MVK	5.00E-06
1084	TOG	NROG	3.18E-04
1084	TOG	OLE1	0.0053113
1084	TOG	OLE2	1.80E-04
1084	TOG	PRD2	2.69E-04
1084	TOG	PRPE	0.02

Profile	Species	SAPRC07 mechanism species	Weight fraction
1084	TOG	TERP	1.90E-04
3161	TOG	13BDE	0.0019002
3161	TOG	ACET	0.0751
3161	TOG	ACYE	0.0425
3161	TOG	ALK1	0.0056999
3161	TOG	ALK2	0.0019001
3161	TOG	ALK3	0.017
3161	TOG	ALK4	0.0267
3161	TOG	ALK5	0.1047
3161	TOG	APIN	0.0049709
3161	TOG	ARO1	0.0182
3161	TOG	ARO2	0.0305
3161	TOG	B124	0.0070559
3161	TOG	BALD	0.0069998
3161	TOG	BENZ	0.02
3161	TOG	CCHO	0.0735
3161	TOG	CH4	0.0408
3161	TOG	CRES	8.34E-04
3161	TOG	ETHE	0.1438
3161	TOG	ETOH	1.00E-04
3161	TOG	HCHO	0.1471
3161	TOG	MEK	0.0148
3161	TOG	MEOH	3.00E-04
3161	TOG	MVK	1.11E-04
3161	TOG	MXYL	0.0061002
3161	TOG	NROG	0.0071509
3161	TOG	OLE1	0.0521
3161	TOG	OLE2	0.0183
3161	TOG	OXYL	0.0034005
3161	TOG	PRD2	0.015
3161	TOG	PRPE	0.026
3161	TOG	PXYL	0.0010001
3161	TOG	RCHO	0.0675
3161	TOG	TERP	0.0042605
3161	TOG	TOLU	0.0147
4547	TOG	ALK3	0.3108
4547	TOG	ALK4	0.2669
4547	TOG	ALK5	0.2171
4547	TOG	APIN	0.004673

Profile	Species	SAPRC07 mechanism species	Weight fraction
4547	TOG	ARO1	0.0188
4547	TOG	ARO2	0.0477
4547	TOG	B124	0.007888
4547	TOG	BENZ	0.0040993
4547	TOG	CRES	7.85E-04
4547	TOG	ISOP	5.12E-04
4547	TOG	MVK	1.04E-04
4547	TOG	MXYL	0.0043145
4547	TOG	NROG	0.0067217
4547	TOG	OLE1	0.0264
4547	TOG	OLE2	0.0515
4547	TOG	OXYL	0.0035873
4547	TOG	PRD2	0.0056763
4547	TOG	PXYL	0.0043145
4547	TOG	TERP	0.0040042
4547	TOG	TOLU	0.0142
4642	TOG	13BDE	0.0061431
4642	TOG	ACET	0.0393
4642	TOG	ACRO	0.0033077
4642	TOG	ACYE	0.0369
4642	TOG	ALK1	0.0225
4642	TOG	ALK2	0.0088734
4642	TOG	ALK3	0.0013666
4642	TOG	ALK4	0.0027618
4642	TOG	ALK5	0.0034923
4642	TOG	APIN	1.50E-04
4642	TOG	ARO1	0.01
4642	TOG	ARO2	0.0184
4642	TOG	B124	3.00E-05
4642	TOG	BACL	0.0173
4642	TOG	BALD	0.0169
4642	TOG	BENZ	0.0201
4642	TOG	CCHO	0.0895
4642	TOG	CH4	0.2163
4642	TOG	CRES	0.1291
4642	TOG	ETHE	0.0588
4642	TOG	GLY	0.0352
4642	TOG	HCHO	0.0612
4642	TOG	IPRD	0.0145

Profile	Species	SAPRC07 mechanism species	Weight fraction
4642	TOG	ISOP	0.0021525
4642	TOG	MACR	0.0012076
4642	TOG	MEK	0.0113
4642	TOG	MGLY	0.0495
4642	TOG	MVK	3.33E-06
4642	TOG	MXYL	0.0015755
4642	TOG	NROG	2.15E-04
4642	TOG	NVOL	1.40E-04
4642	TOG	OLE1	0.0058554
4642	TOG	OLE2	0.0178
4642	TOG	OXYL	9.50E-04
4642	TOG	PACD	5.46E-04
4642	TOG	PRD2	1.81E-04
4642	TOG	PRPE	0.0225
4642	TOG	PXYL	0.0015755
4642	TOG	RCHO	0.0641
4642	TOG	TERP	1.27E-04
4642	TOG	TOLU	0.0082961
4662	TOG	RCHO	5.46E-04
4674	TOG	13BDE	0.001184
4674	TOG	ACRO	0.013
4674	TOG	ACYE	0.0176
4674	TOG	ALK3	0.0208
4674	TOG	ALK4	0.0454
4674	TOG	ALK5	0.1504
4674	TOG	APIN	0.0065917
4674	TOG	ARO1	0.0354
4674	TOG	ARO2	0.0363
4674	TOG	B124	0.0047105
4674	TOG	BACL	0.0034375
4674	TOG	BALD	0.0302
4674	TOG	BENZ	0.0105
4674	TOG	CCHO	0.1596
4674	TOG	CRES	0.0059201
4674	TOG	ETHE	0.0327
4674	TOG	GLY	0.00802
4674	TOG	HCHO	0.0852
4674	TOG	IPRD	0.0512
4674	TOG	MACR	0.0153

Profile	Species	SAPRC07 mechanism species	Weight fraction
4674	TOG	MEK	0.0286
4674	TOG	MGLY	0.0064928
4674	TOG	MVK	1.46E-04
4674	TOG	MXYL	0.0044494
4674	TOG	NROG	0.0094813
4674	TOG	NVOL	0.00293
4674	TOG	OLE1	0.0129
4674	TOG	OLE2	0.0165
4674	TOG	OXYL	0.0031701
4674	TOG	PACD	0.002949
4674	TOG	PRD2	0.0080063
4674	TOG	PRPE	0.0029788
4674	TOG	PXYL	0.0044494
4674	TOG	RCHO	0.1405
4674	TOG	TERP	0.005648
4674	TOG	TOLU	0.0152
4674	TOG	UNK	0.0021625
5560	TOG	13BDE	0.0026493
5560	TOG	AACD	0.1438
5560	TOG	ACET	0.021
5560	TOG	ACRO	0.0090828
5560	TOG	ACYE	0.0102
5560	TOG	ALK1	0.0237
5560	TOG	ALK2	0.0134
5560	TOG	ALK3	0.0034437
5560	TOG	ALK4	0.0124
5560	TOG	ALK5	0.0011737
5560	TOG	APIN	0.0033309
5560	TOG	ARO1	0.0221
5560	TOG	ARO2	9.46E-04
5560	TOG	BACL	0.035
5560	TOG	BALD	0.0013626
5560	TOG	BENZ	0.0185
5560	TOG	CCHO	0.0189
5560	TOG	CH4	0.1779
5560	TOG	CRES	1.89E-04
5560	TOG	ETHE	0.0424
5560	TOG	ETOH	6.81E-04

Profile	Species	SAPRC07 mechanism species	Weight fraction
5560	TOG	FACD	0.1098
5560	TOG	HCHO	0.0833
5560	TOG	ISOP	0.0037846
5560	TOG	MEK	0.0206
5560	TOG	MEOH	0.0757
5560	TOG	MXYL	0.0018919
5560	TOG	NROG	0.0092044
5560	TOG	OLE1	0.0142
5560	TOG	OLE2	0.0135
5560	TOG	OXYL	0.0018919
5560	TOG	PRPE	0.0223
5560	TOG	PXYL	0.0018919
5560	TOG	RCHO	0.0179
5560	TOG	TERP	0.0049957
5560	TOG	TOLU	0.0151
5560	TOG	UNK	0.0417
8746	TOG	13BDE	0.0132
8746	TOG	ACYE	0.0142
8746	TOG	ALK2	0.0237
8746	TOG	ALK3	0.0107
8746	TOG	ALK4	0.0337
8746	TOG	ALK5	0.03
8746	TOG	APIN	2.79E-04
8746	TOG	ARO1	0.0031532
8746	TOG	ARO2	0.004808
8746	TOG	B124	0.0016043
8746	TOG	BACL	0.0196
8746	TOG	BALD	0.0168
8746	TOG	BENZ	0.0186
8746	TOG	CCHO	0.1243
8746	TOG	CRES	2.63E-05
8746	TOG	ETHE	0.0949
8746	TOG	GLY	0.1974
8746	TOG	HCHO	0.0843
8746	TOG	IPRD	0.0066782
8746	TOG	ISOP	0.0065631
8746	TOG	MACR	0.0052014
8746	TOG	MEK	0.019
8746	TOG	MGLY	0.0828

Profile	Species	SAPRC07 mechanism species	Weight fraction
8746	TOG	MVK	3.33E-06
8746	TOG	MXYL	0.0019248
8746	TOG	NROG	2.30E-04
8746	TOG	OLE1	0.0136
8746	TOG	OLE2	0.0344
8746	TOG	OXYL	0.0015521
8746	TOG	PRD2	1.94E-04
8746	TOG	PRPE	0.0543
8746	TOG	PXYL	0.0019248
8746	TOG	RCHO	0.0662
8746	TOG	TERP	1.57E-04
8746	TOG	TOLU	0.0139
8750	TOG	13BDE	0.0053598
8750	TOG	ACET	0.0013596
8750	TOG	ACRO	6.40E-04
8750	TOG	ACYE	0.0301
8750	TOG	ALK1	0.0215
8750	TOG	ALK2	0.0026109
8750	TOG	ALK3	0.0297
8750	TOG	ALK4	0.1059
8750	TOG	ALK5	0.0896
8750	TOG	ARO1	0.0293
8750	TOG	ARO2	0.0553
8750	TOG	B124	0.0148
8750	TOG	BALD	0.0052486
8750	TOG	BENZ	0.0437
8750	TOG	CCHO	0.0104
8750	TOG	CH4	0.1404
8750	TOG	ETHE	0.0548
8750	TOG	GLY	1.23E-04
8750	TOG	HCHO	0.0133
8750	TOG	IPRD	2.27E-04
8750	TOG	MACR	5.79E-04
8750	TOG	MEK	9.55E-04
8750	TOG	MXYL	0.0281
8750	TOG	OLE1	0.0876
8750	TOG	OLE2	0.0372
8750	TOG	OXYL	0.0219
8750	TOG	PRPE	0.037

Profile	Species	SAPRC07 mechanism species	Weight fraction
8750	TOG	PXYL	0.0281
8750	TOG	RCHO	0.0067678
8750	TOG	TOLU	0.0962
8750	TOG	UNK	0.0013221
8751	TOG	13BDE	0.00521
8751	TOG	ACET	0.0013794
8751	TOG	ACRO	6.40E-04
8751	TOG	ACYE	0.0305
8751	TOG	ALK1	0.0218
8751	TOG	ALK2	0.0026484
8751	TOG	ALK3	0.0302
8751	TOG	ALK4	0.1074
8751	TOG	ALK5	0.0909
8751	TOG	ARO1	0.0263
8751	TOG	ARO2	0.0499
8751	TOG	B124	0.0133
8751	TOG	BALD	0.0047227
8751	TOG	BENZ	0.0441
8751	TOG	CCHO	0.0127
8751	TOG	CH4	0.1424
8751	TOG	ETHE	0.0556
8751	TOG	ETOH	0.0157
8751	TOG	GLY	1.25E-04
8751	TOG	HCHO	0.0136
8751	TOG	IPRD	2.31E-04
8751	TOG	MACR	5.87E-04
8751	TOG	MEK	9.68E-04
8751	TOG	MXYL	0.0253
8751	TOG	OLE1	0.0889
8751	TOG	OLE2	0.0376
8751	TOG	OXYL	0.0197
8751	TOG	PRPE	0.0375
8751	TOG	PXYL	0.0253
8751	TOG	RCHO	0.0068648
8751	TOG	TOLU	0.0866
8751	TOG	UNK	0.0011905
8753	TOG	13BDE	0.0029571
8753	TOG	ALK2	0.0029847
8753	TOG	ALK3	0.3066

Profile	Species	SAPRC07 mechanism species	Weight fraction
8753	TOG	ALK4	0.2946
8753	TOG	ALK5	0.0468
8753	TOG	ARO1	0.0291
8753	TOG	ARO2	0.0294
8753	TOG	B124	0.0133
8753	TOG	BENZ	0.0336
8753	TOG	ISOP	2.93E-05
8753	TOG	MXYL	0.0301
8753	TOG	OLE1	0.0042276
8753	TOG	OLE2	0.0574
8753	TOG	OXYL	0.0208
8753	TOG	PXYL	0.0301
8753	TOG	TOLU	0.0978
8754	TOG	ALK2	0.0042451
8754	TOG	ALK3	0.2095
8754	TOG	ALK4	0.3039
8754	TOG	ALK5	0.038
8754	TOG	ARO1	0.0189
8754	TOG	ARO2	0.0175
8754	TOG	B124	0.0093785
8754	TOG	BENZ	0.0323
8754	TOG	ETOH	0.1205
8754	TOG	ISOP	1.29E-05
8754	TOG	MXYL	0.0236
8754	TOG	OLE1	0.0022588
8754	TOG	OLE2	0.0332
8754	TOG	OXYL	0.018
8754	TOG	PXYL	0.0236
8754	TOG	TOLU	0.1453
8757	TOG	13BDE	0.0089141
8757	TOG	ACET	0.0034627
8757	TOG	ACRO	7.23E-04
8757	TOG	ACYE	0.0165
8757	TOG	ALK1	0.027
8757	TOG	ALK2	0.0015098
8757	TOG	ALK3	0.052
8757	TOG	ALK4	0.0882
8757	TOG	ALK5	0.1078
8757	TOG	ARO1	0.0331

Profile	Species	SAPRC07 mechanism species	Weight fraction
8757	TOG	ARO2	0.0429
8757	TOG	B124	0.0114
8757	TOG	BALD	0.0023709
8757	TOG	BENZ	0.0379
8757	TOG	CCHO	0.0161
8757	TOG	CH4	0.1736
8757	TOG	ETHE	0.1005
8757	TOG	ETOH	0.0381
8757	TOG	HCHO	0.008737
8757	TOG	ISOP	0.0010211
8757	TOG	MEK	1.38E-04
8757	TOG	MEOH	0.0053635
8757	TOG	MXYL	0.0109
8757	TOG	OLE1	0.0129
8757	TOG	OLE2	0.0443
8757	TOG	OXYL	0.0278
8757	TOG	PRPE	0.0399
8757	TOG	PXYL	0.0109
8757	TOG	RCHO	0.0014931
8757	TOG	TOLU	0.0744
8762	TOG	13BDE	9.95E-05
8762	TOG	ALK2	0.001462
8762	TOG	ALK3	0.104
8762	TOG	ALK4	0.6841
8762	TOG	ALK5	0.0766
8762	TOG	APIN	5.27E-04
8762	TOG	ARO1	0.0036274
8762	TOG	ARO2	0.0060772
8762	TOG	B124	0.0020088
8762	TOG	BENZ	0.0034322
8762	TOG	CRES	8.90E-05
8762	TOG	ISOP	1.79E-04
8762	TOG	MVK	1.17E-05
8762	TOG	MXYL	0.0023877
8762	TOG	NROG	7.60E-04
8762	TOG	OLE1	0.0097976
8762	TOG	OLE2	0.0674
8762	TOG	OXYL	9.85E-04
8762	TOG	PRD2	6.41E-04

Profile	Species	SAPRC07 mechanism species	Weight fraction
8762	TOG	PXYL	0.0023877
8762	TOG	TERP	4.52E-04
8762	TOG	TOLU	0.0329
8766	TOG	13BDE	1.71E-04
8766	TOG	ALK2	1.74E-04
8766	TOG	ALK3	0.1561
8766	TOG	ALK4	0.36
8766	TOG	ALK5	0.1494
8766	TOG	APIN	1.48E-04
8766	TOG	ARO1	0.0068652
8766	TOG	ARO2	0.0149
8766	TOG	B124	0.0017609
8766	TOG	BENZ	0.025
8766	TOG	CH4	7.84E-04
8766	TOG	CRES	2.48E-05
8766	TOG	ETHE	3.00E-04
8766	TOG	ISOP	3.04E-04
8766	TOG	MEOH	6.33E-04
8766	TOG	MVK	3.33E-06
8766	TOG	MXYL	0.0066449
8766	TOG	NROG	2.14E-04
8766	TOG	OLE1	0.0128
8766	TOG	OLE2	0.1389
8766	TOG	OXYL	0.00341
8766	TOG	PRD2	1.81E-04
8766	TOG	PXYL	0.0066449
8766	TOG	TERP	1.27E-04
8766	TOG	TOLU	0.1146
8769	TOG	13BDE	0.00449
8769	TOG	ALK1	1.07E-04
8769	TOG	ALK2	0.0020963
8769	TOG	ALK3	0.1008
8769	TOG	ALK4	0.2819
8769	TOG	ALK5	0.0822
8769	TOG	APIN	2.15E-04
8769	TOG	ARO1	0.0104
8769	TOG	ARO2	0.0139
8769	TOG	B124	0.0019905

Profile	Species	SAPRC07 mechanism species	Weight fraction
8769	TOG	BENZ	0.0227
8769	TOG	CRES	3.65E-05
8769	TOG	ETHE	1.14E-04
8769	TOG	ETOH	0.2024
8769	TOG	ISOP	0.0023664
8769	TOG	MEOH	0.0044968
8769	TOG	MVK	5.00E-06
8769	TOG	MXYL	0.0044112
8769	TOG	NROG	3.09E-04
8769	TOG	OLE1	0.0168
8769	TOG	OLE2	0.1395
8769	TOG	OXYL	0.0025373
8769	TOG	PRD2	2.61E-04
8769	TOG	PXYL	0.0044112
8769	TOG	TERP	1.83E-04
8769	TOG	TOLU	0.1012
8774	TOG	13BDE	0.0085998
8774	TOG	ACYE	0.0669
8774	TOG	ALK1	0.0185
8774	TOG	ALK2	0.0223
8774	TOG	ALK3	0.0296
8774	TOG	ALK4	0.1182
8774	TOG	ALK5	0.0521
8774	TOG	APIN	8.87E-04
8774	TOG	ARO1	0.0051793
8774	TOG	ARO2	0.0148
8774	TOG	B124	3.29E-06
8774	TOG	BALD	0.0061508
8774	TOG	BENZ	0.032
8774	TOG	CCHO	0.0557
8774	TOG	CRES	2.56E-06
8774	TOG	ETHE	0.2045
8774	TOG	GLY	0.0073897
8774	TOG	HCHO	0.0889
8774	TOG	IPRD	0.0032003
8774	TOG	ISOP	0.0015299
8774	TOG	MEK	0.03
8774	TOG	MVK	7.30E-07
8774	TOG	MXYL	0.0031499

Profile	Species	SAPRC07 mechanism species	Weight fraction
8774	TOG	NROG	2.41E-05
8774	TOG	OLE1	0.0327
8774	TOG	OLE2	0.0957
8774	TOG	OXYL	0.0026095
8774	TOG	PRD2	1.97E-05
8774	TOG	PRPE	0.0587
8774	TOG	PXYL	0.0031499
8774	TOG	RCHO	0.0198
8774	TOG	TERP	3.35E-04
8774	TOG	TOLU	0.0147
8774	TOG	UNK	0.0028092
8869	TOG	ALK2	0.0011806
8869	TOG	ALK3	0.1246
8869	TOG	ALK4	0.5431
8869	TOG	ALK5	0.0729
8869	TOG	APIN	5.83E-04
8869	TOG	ARO1	0.0063553
8869	TOG	ARO2	0.0133
8869	TOG	B124	0.0034155
8869	TOG	BENZ	0.0077198
8869	TOG	CRES	9.77E-05
8869	TOG	ISOP	2.57E-04
8869	TOG	MVK	1.33E-05
8869	TOG	MXYL	0.0072086
8869	TOG	NROG	8.39E-04
8869	TOG	OLE1	0.0183
8869	TOG	OLE2	0.1202
8869	TOG	OXYL	0.002945
8869	TOG	PRD2	7.09E-04
8869	TOG	PXYL	0.0072086
8869	TOG	TERP	5.00E-04
8869	TOG	TOLU	0.0687
8870	TOG	ALK2	4.45E-04
8870	TOG	ALK3	0.1066
8870	TOG	ALK4	0.4469
8870	TOG	ALK5	0.0802
8870	TOG	APIN	0.0013146
8870	TOG	ARO1	0.0121
8870	TOG	ARO2	0.0339

Profile	Species	SAPRC07 mechanism species	Weight fraction
8870	TOG	B124	0.0094559
8870	TOG	BENZ	0.0087095
8870	TOG	CRES	2.20E-04
8870	TOG	ETOH	0.0645
8870	TOG	ISOP	2.70E-04
8870	TOG	MVK	2.92E-05
8870	TOG	MXYL	0.0121
8870	TOG	NROG	0.00189

Profile	Species	SAPRC07 mechanism species	Weight fraction
8870	TOG	OLE1	0.0298
8870	TOG	OLE2	0.1194
8870	TOG	OXYL	0.0052891
8870	TOG	PRD2	0.0015963
8870	TOG	PXYL	0.0121
8870	TOG	TERP	0.0011271
8870	TOG	TOLU	0.0521

Figures

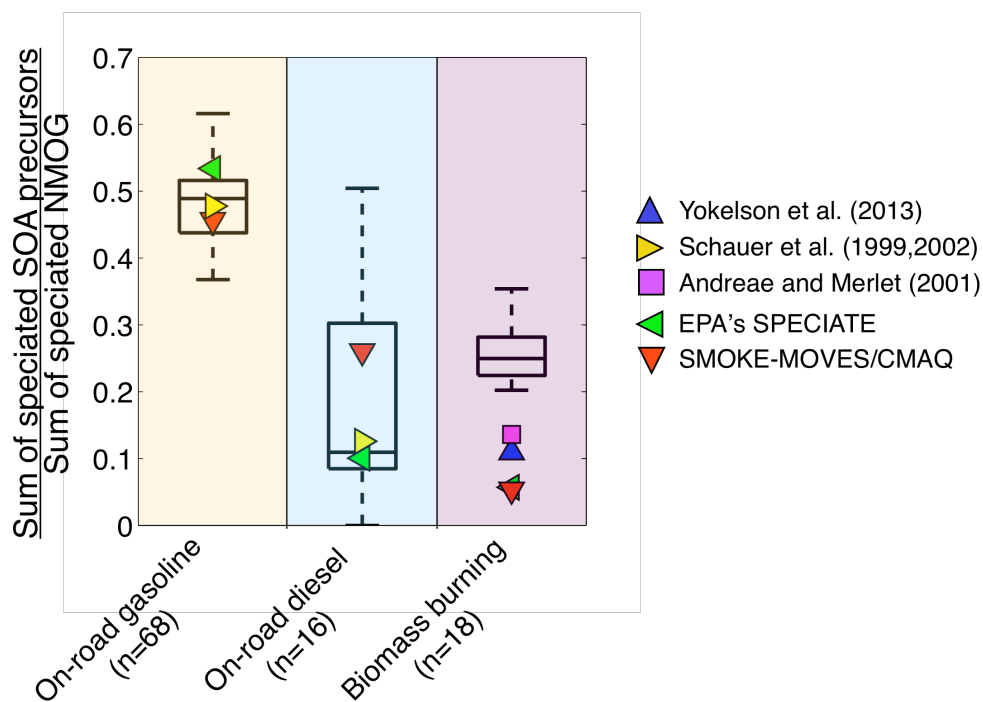


Figure S1: Box-plots for the ratio of speciated SOA precursors found in the emissions to the sum of all speciated compounds. Ratios from literature are shown using symbols (18, 26-29).

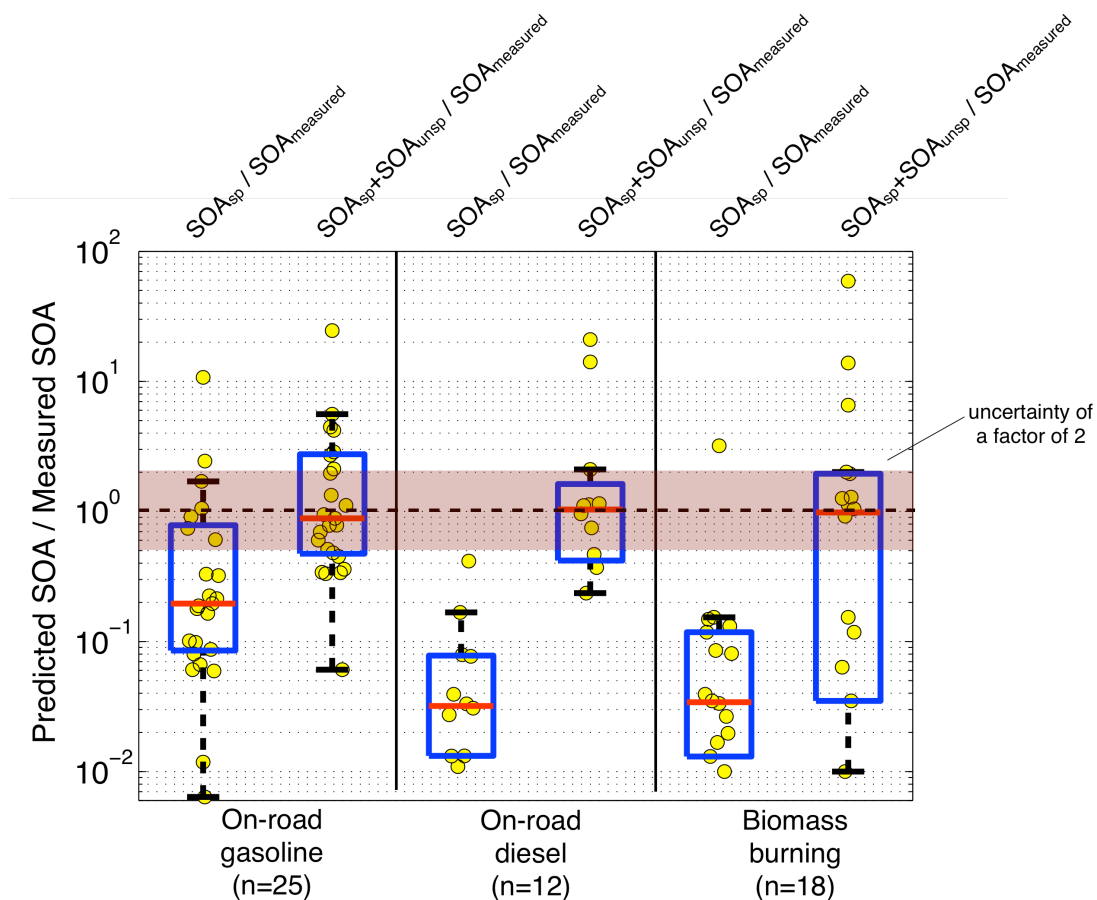


Figure S2: Ratio of predicted SOA to measured SOA for the model that included SOA formation from speciated organics only and for the model that included SOA formation from both speciated and unspeciated organics. The median model-measurement agreement is substantially improved (i.e., the bias observed for the $SOA_{sp}/SOA_{measured}$ is eliminated) for each of the three source classes when the SOA contributions from the unspeciated precursors ($SOA_{sp}+SOA_{unsp}/SOA_{measured}$) are included.

PROCESS FOR CONVERTING A SPECIATED EMISSION PROFILE TO MODEL-READY EMISSION INPUTS

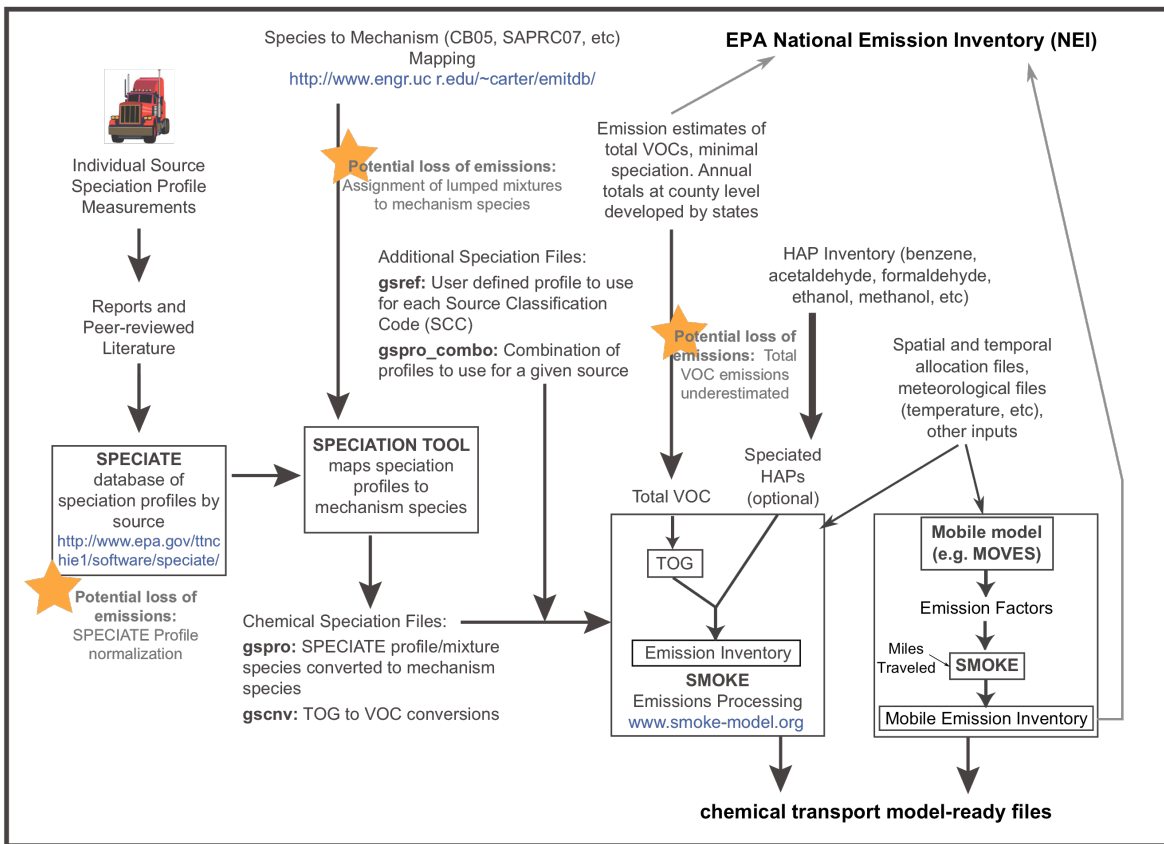


Figure S3: Schematic of emissions processing for chemical transport model-ready emissions for a model like CMAQ. Three potential losses of unspciated NMOG mass are highlighted: normalization of EPA’s SPECIATE profiles, species to mechanism mapping, and underestimation of total NMOG. Hazardous air pollutants (HAP) may either come from the HAP inventory or by speciating total NMOG from the NEI.

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