Photochemical Model PM2.5 Source Apportionment of 2011 National Emission Inventory Based Residential Fuel Combustion Emissions

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Air Quality Analysis Division
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Background

The US Environmental Protection Agency regulates emissions from residential wood heaters under the Clean Air Act through new source performance standards (NSPS). Additionally, the Agency has a public outreach partnership program where EPA works with Federal partners, States, Tribes, local air agencies, device manufacturers, retailers, and chimney sweeps to promote best practices about burning wood in home appliances. This program, Burn Wise, also provides information to communities about appliance change-out programs and educational material (http://www.epa.gov/burnwise/). Air quality modeling can provide useful information about the contribution of this sector to ambient particulate matter less than 2.5 microns in diameter (PM2.5) to support public outreach efforts.

Photochemical grid models use state of the science numerical algorithms to estimate pollutant formation, transport, and deposition over a variety of spatial scales that range from urban to continental. Emissions of precursor species are injected into the model where they react to form secondary species such as PM2.5 and then undergo transport before ultimately being removed by deposition or chemical reaction. Photochemical model source apportionment estimates source specific contribution from primarily emitted PM2.5 and from precursors through the formation and transport of secondary formed particulate matter. This type of emissions apportionment is useful to understand what types of sources or regions are contributing to PM2.5 estimated by photochemical grid models.

Photochemical transport model source apportionment is used to estimate the contribution of emissions from the residential fuel combustion sector to model estimated primary and secondary PM2.5. This is a national scale assessment done using emissions from the 2011 National Emissions Inventory version 1.5, which contains estimates of multiple residential fuel combustion groups: fireplaces, woodstoves, outdoor hydronic heaters, and outdoor recreational devices. Photochemical transport model estimates have a 12 km grid resolution, represent the year 2011, and do not include any projections to future years or quantify the effects of any specific control programs.

Methods

Photochemical Model

The Comprehensive Air Quality Model with Extensions (CAMx) version 6.01 is a state of the science three-dimensional Eularian "one-atmosphere" photochemical transport model that treats the physical processes and chemistry that form ozone and PM2.5 (Nobel et al., 2002; Chen et al., 2003; Baker and Scheff, 2007). CAMx is applied with ISORROPIA inorganic chemistry (Nenes et al., 1998), a semi-volatile equilibrium scheme to partition condensable organic gases between gas and particle phase (Strader et al., 1999), regional acid deposition model (RADM) aqueous phase chemistry (Chang et al., 1987), and Carbon Bond (CB6) gas-phase chemistry module (ENVIRON, 2013).

Particulate matter source apportionment technology (PSAT) has been implemented into the most recent version of the CAMx model and is publicly available (ENVIRON, 2008; Wagstrom et al, 2008). PSAT estimates the contribution from specific emissions source groups, emissions source regions, initial conditions, and boundary conditions to PM2.5 using reactive tracers. The tracer species are estimated with source apportionment algorithms rather than by the host model routines. PSAT has the capability to track contribution to PM sulfate, nitrate, ammonium, secondary organic aerosol, and inert primarily emitted species. Non-linear processes like gas and aqueous phase chemistry are solved for bulk species and then apportioned to the tagged species.

Particulate source apportionment tracks contributions to particulate species from pre-cursor emissions. Emissions of nitrogen oxides are tracked through all intermediate nitrogen species to particulate nitrate ion. Ammonia emissions are tracked to particulate ammonium ion. Even though ammonium nitrate is chemically coupled, the apportionment schemes do not attempt to determine which species is limiting the formation, but directly attributes precursor gases to specific particulate ions (Table 1).

Table 1. Emissions precursor species (left) tracked for contribution to PM2.5 species (right).

NOX → $NO3^-$
SOX → $SO4^=$
NH3 → $NH4^+$
POC → POC
PEC → PEC
SOIL → SOIL

The CAMx photochemical model was applied for the entire calendar year of 2011 to track source group emissions. PSAT is used to track source contribution to model estimated PM2.5 sulfate, nitrate, ammonium, and primary emitted species. Contribution is not tracked to model estimated secondary organic aerosol. All model domains are applied with a Lambert projection centered at (-97, 40) and true latitudes at 33 and 45. The specifications for the model domain are given in Table 2. The model domain is applied with square 12 km sized grid cells. The vertical atmosphere up to approximately 15 km above ground level is resolved with 25 layers. The layers are smaller inside the planetary boundary layer (mixing layer) to capture the important diurnal variations in mixing height.

Meteorological Inputs

Meteorological inputs are generated with version 3.4 of the WRF model, Advanced Research WRF (ARW) core (Skamarock, 2008). Selected physics options include Pleim-Xiu land surface model, Asymmetric Convective Model version 2 planetary boundary layer scheme, Kain-Fritsch cumulus parameterization utilizing the moisture-advection trigger (Ma and Tan, 2009), Morrison double moment microphysics, and RRTMG longwave and shortwave radiation schemes (Gilliam and Pleim, 2010). More details on the meteorological modeling are available elsewhere (US Environmental Protection Agency, 2011).

Emissions

The emissions used for the photochemical modeling are based on the 2011 National Emission Inventory version 1.5 for stationary point, area, and mobile sources (emissions scenario 2011ec_rwc_v6_11f). Residential fuel combustion emissions are based on data extracted from the residential fuel sector emission inventory tool on January 17, 2014 (2011NEIv1_5_nonpoint_20140117). The residential wood combustion inventory tool combines information from residential surveys about appliance profiles and burn rates with relevant emission factors to estimate County total emissions from 12 different wood burning appliance types (Cooley et al, 2014). More details about the anthropogenic emissions used for this analysis are provided elsewhere (http://www.epa.gov/ttn/chief/emch/index.html#2011). Biogenic emissions are estimated using hourly gridded day-specific meteorology with the BEIS v3.14 model. All emissions were processed using the latest version of the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (UNC, 2007).

Total emissions of PM2.5 precursors from the residential wood combustion sector are presented in Table 2 for each SCC group tracked for contribution estimates. Emissions are presented in tons per year. Primarily emitted PM2.5 from each residential fuel combustion sector is based on profile 91105 (see Table 3).

Table 2. Precursor emissions (TPY) used for source apportionment tracking

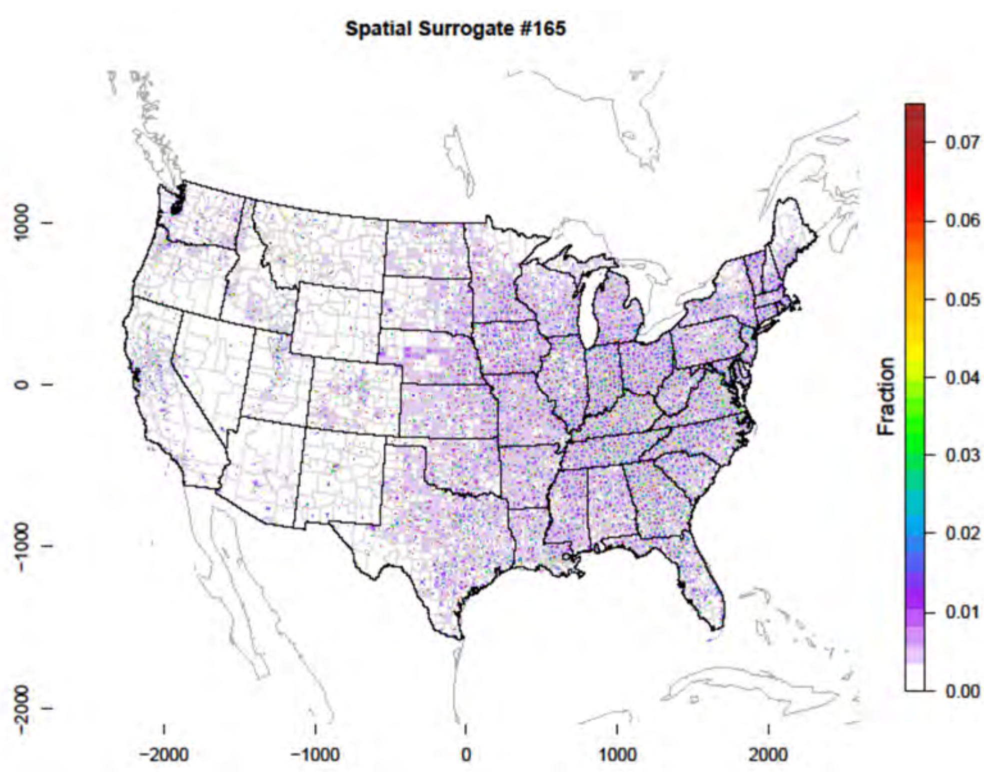
Group	SCC Description	SCC	POM	PEC	Other PM2.5	NOX	NH3	SO2
2	Fireplace: general	2104008100	58,997	3,666	3,044	7,223	4,987	1,111
3	Woodstove: fireplace inserts; non-EPA certified	2104008210	46,671	2,900	2,408	4,756	2,888	679
4	Woodstove: fireplace inserts; EPA certified; non-catalytic	2104008220	10,870	675	561	1,408	556	247
5	Woodstove: fireplace inserts; EPA certified; catalytic	2104008230	4,267	265	220	466	210	93
6	Woodstove: freestanding, non-EPA certified	2104008310	79,212	4,923	4,087	8,082	4,901	1,155
7	Woodstove: freestanding, EPA certified, non-catalytic	2104008320	17,700	1,100	913	2,290	904	401
8	Woodstove: freestanding, EPA certified, catalytic	2104008330	10,187	633	526	1,112	501	223
9	Woodstove: pellet-fired, general (freestanding or FP insert)	2104008400	1,763	110	91	2,438	192	205
9	Woodstove: pellet-fired, EPA certified (freestanding or FP insert)	2104008420	15	1	1	56	-	2
10	Furnace: Indoor, cordwood-fired, non-EPA certified	2104008510	23,835	1,481	1,230	1,767	1,731	1,952
11	Hydronic heater: outdoor	2104008610	70,768	4,398	3,651	2,280	2,236	2,521
12	Outdoor wood burning device, NEC (fire-pits, chimeas, etc)	2104008700	18,514	1,151	955	2,272	1,573	349
13	Firelog;Total: All Combustor Types	2104009000	6,438	400	332	1,940	-	-
	Total		349,236	21,703	18,020	36,089	20,678	8,939

Table 3. Speciation of primary PM2.5 emissions from the residential wood combustion sector.

Profile	Specie	Fraction of PM25
91105	POC	0.528
91105	PNCOM	0.370
91105	PEC	0.056
91105	PMOTHR	0.025
91105	PK	0.010
91105	PSO4	0.004
91105	PCL	0.003
91105	PNO3	0.002
91105	PNH4	0.002
91105	PNA	0.001
91105	PSI	0.000
91105	PMG	0.000
91105	PAL	0.000
91105	PCA	0.000
91105	PFE	0.000
	Total	1.000

All residential fuel combustion emissions are spatially allocated based on the same surrogate (165), which is comprised of 50% low intensity residential land and 50% residential heating-wood. This surrogate is shown in Figure 1.

Figure 1. Spatial surrogate (percentage) used for residential wood combustion sector.



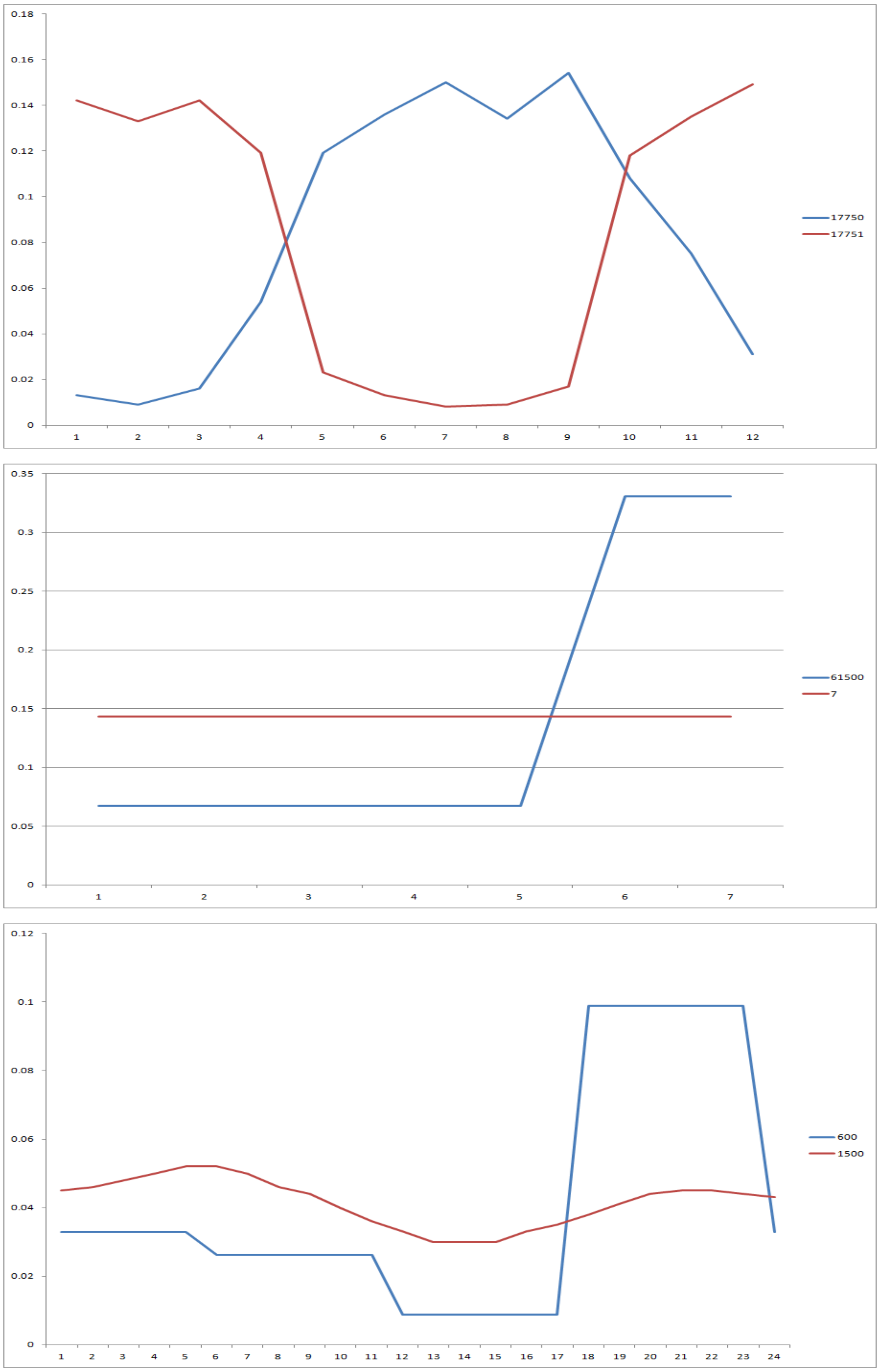
Temporal allocation of annual total emissions varies among SCC categories for the residential fuel combustion sector (see Table 4). Outdoor hydronic heaters (2104008610) are allocated from year to month using profile 17751 which puts most of the mass in the colder months and profile 7 to allocate emissions equally to each day of the week. Outdoor recreational devices (2104008700) use profile 17750 to allocate annual emissions to month, which allocates most of the mass to the warmer months. Week to day allocation for this category is based on 61500 which allocates most of the mass to weekend days.

Table 4. Temporal allocation approach for each SCC of the residential wood combustion sector.

Group	SCC Description	SCC	Year to Month Allocation	Week to Day Allocation	Diurnal Allocation
2	Fireplace: general	2104008100	County based meteorology		600
3	Woodstove: fireplace inserts; non-EPA certified	2104008210	County based meteorology		600
4	Woodstove: fireplace inserts; EPA certified; non-catalytic	2104008220	County based meteorology		600
5	Woodstove: fireplace inserts; EPA certified; catalytic	2104008230	County based meteorology		600
6	Woodstove: freestanding, non-EPA certified	2104008310	County based meteorology		600
7	Woodstove: freestanding, EPA certified, non-catalytic	2104008320	County based meteorology		600
8	Woodstove: freestanding, EPA certified, catalytic	2104008330	County based meteorology		600
9	Woodstove: pellet-fired, general (freestanding or FP insert)	2104008400	County based meteorology		600
9	Woodstove: pellet-fired, EPA certified (freestanding or FP insert)	2104008420	County based meteorology		600
10	Furnace: Indoor, cordwood-fired, non-EPA certified	2104008510	County based meteorology		600
11	Hydronic heater: outdoor	2104008610	17751	7	1500
12	Outdoor wood burning device, NEC (fire-pits, chimeas, etc)	2104008700	17750	61500	600
13	Firelog;Total: All Combustor Types	2104009000	County based meteorology		600

All other categories allocate annual emissions to specific days based on meteorology, where colder days are allocated more emissions mass (Adelman et al, 2010). All categories use profile 600 for diurnal allocation except outdoor hydronic heaters (profile 1500). Both profiles generally put more emissions mass into the early morning and evening hours. Temporal profiles for year to month, day of week, and hourly allocation are shown in Figure 2.

Figure 2. Non-meteorologically based temporal allocation used for some SCCs in the residential wood combustion sector: year to month (top), month to day of the week (middle), and diurnal (bottom).



Selection of Sources

The sources selected for tracking with source apportionment include emissions from specific SCC categories that make up the residential fuel combustion sector (see Table 2). Source group 1 contains all non-residential wood combustion emissions and is not included in this analysis.

Identifying Monitors (Receptors)

Receptors are defined as individual model grid cells that contain a monitor of interest for regulatory analysis. PM2.5 design values are estimated using methods described in 40 CFR part 50 Appendix N.

Post Processing Contributions

Modeled PM2.5 source contribution estimates are expressed as a percentage of bulk modeled PM2.5 to estimate daily relative response factors, which are averaged over all modeled elevated air quality days (U.S. Environmental Protection Agency 2007). The Modeled Attainment Test Software (MATS) (U.S. Environmental Protection Agency 2010) matches model estimates with weighted observed design values by the grid cell where the monitor is located. For this assessment the weighted design value is an average of 2009-2011 and 2010-2012 values.

We developed and applied several post-processing steps to transform the PSAT modeling outputs to PM2.5 contributions. The approach involved processing the PSAT model outputs using MATS along with other post-processing software to calculate the contribution of each category to each FRM monitor. The following is a description of the procedures for calculating contributions for annual PM2.5. These procedures were applied separately for each source group shown in Table 4.

1. Receptor sites include all FRM sites in the modeling domain.
2. Contributions for each of the PM2.5 species from each source group, as predicted by PSAT, are subtracted from the standard 2011 base case model output to generate a new set of model output files for each source group.
3. Daily, 24-hr average PM2.5 species are calculated for the “standard” model output files and newly generated source contribution output files.

4. The relative response factors (RRFs) for each of the PM2.5 species is calculated for each source group at all receptors using the MATS model attainment software. In this approach, the MATS "baseline" model file is defined as the standard base case model output file and the "future case" model file is defined as the source group contribution model output file (from step 3).
5. The species-specific annual average RRFs (generated by MATS in step 4) for each source group are multiplied by the annual average observed species concentrations to estimate PM2.5 species contributions in ug/m^3 from each species for each source group.
6. The annual average contributions of organic carbon, elemental carbon, sulfate ion, nitrate ion, ammonium, and water for each source group are combined to calculate the total PM2.5 contribution.
7. Annual PM2.5 (i.e., nitrate plus sulfate) contributions are expressed in units of μg/m^3. Values of annual PM2.5 contribution are truncated after two places to the right of the decimal (e.g. a contribution of 0.149 μg/m^3 is truncated to 0.14 μg/m^3).

The 24-hour PM2.5 contributions were calculated in a manner similar to the procedures for annual PM2.5. However, there are several more steps in the 24-hour calculations which are designed to retain the contributions in each quarter through most of the post-processing. For 24-hour PM2.5, the contributions are calculated as the multi-year average contributions to the "high" concentration quarters at each site. The following is a description of the procedures for calculating contributions for 24-hour PM2.5. These procedures were applied separately for each source group.

1. Receptor sites include all FRM sites in the modeling domain.
2. Contributions for each of the PM2.5 species from each source group, as predicted by PSAT, are subtracted from the standard 2011 base case model output to generate a new set of model output files for each source group.
3. Daily, 24-hr average PM2.5 species are calculated for the "standard" model output files and newly generated source contribution output files.
4. Relative response factors (RRFs) are calculated for each of the PM2.5 species for each source group at all receptors using the MATS model attainment software. Quarterly RRFs are calculated using the "high" concentration model days in each quarter. The high concentration days are based on the highest 10% of modeled PM2.5 days (in each grid cell) in the base case. The MATS

"baseline" model file is defined as the standard base case model output file and the "future case" model file is defined as the source group contribution model output file (from step 3).

5. The species "high days" quarterly average RRFs (generated by MATS in step 4) for each source group are multiplied by the high days quarterly average observed species concentrations from the 24-hr PM2.5 base case. This calculation is done for (up to) 5 years of data for each quarter (for a total of up to 20 quarters). The result of this calculation is the contribution of each of the species from the source group to each of the 20 quarters.
6. For each receptor, the contributions during the high quarters for each year are identified and selected for use in the analysis. The high quarter for each year (based on the 2011 base case) is already known from the base case 24-hr PM2.5 design value calculations. The contributions of each species for the (up to) 5 high quarters are averaged together. This represents the species contributions to 24-hour PM2.5 concentrations.
7. The 24-hour contributions of organic carbon, elemental carbon, sulfate ion, nitrate ion, ammonium, and water for each source group are combined to calculate the total PM2.5 contribution.
8. 24-hour PM2.5 contributions are expressed in units of $\mu g/m^3$. Values of 24-hour PM2.5 contribution are truncated after two places to the right of the decimal (e.g. a contribution of 0.349 $\mu g/m^3$ is truncated to 0.34 $\mu g/m^3$).

RESULTS

Limitations

Source apportionment estimates are as good as the inputs to the photochemical model. Any deficiencies with the emissions or meteorological inputs may lead to source contribution estimates that may not fully characterize the source contribution mix at a receptor location. Some contribution from the residential fuel combustion sector may be overstated to some degree in certain locations on certain days where burn restrictions may have been in place.

This application used a minimum of a complete year of meteorology to capture the variety of PM2.5 formation regimes. However, it is possible that the meteorology used for these model applications may not represent all PM2.5 formation regimes at every individual receptor location in the continental

United States. Additionally, the meteorology used may not capture local scale features such as persistent near-surface inversions coupled with nearby terrain features that result in pollutant accumulation.

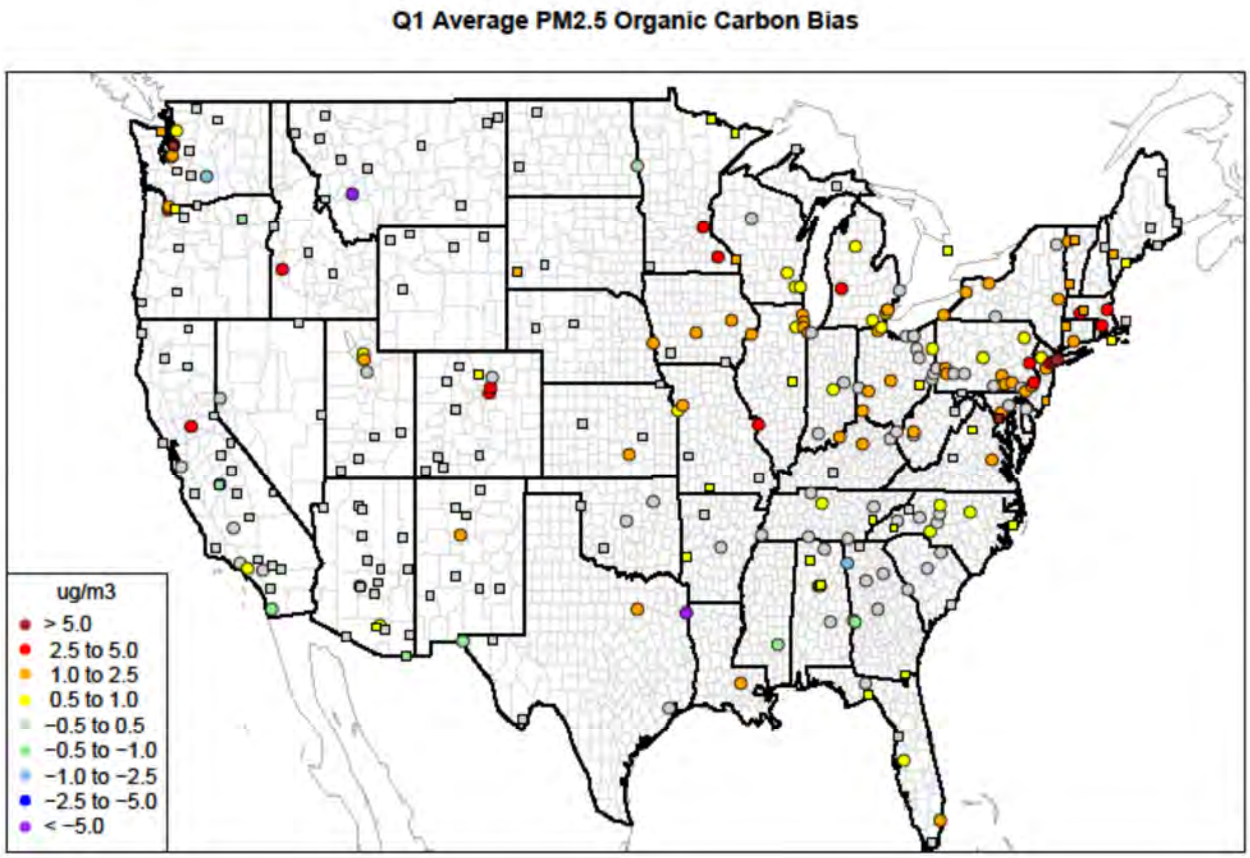
Contribution was not estimated for secondary organic aerosol. However, total anthropogenic secondary organic aerosol estimated by CAMx was examined to provide an upper bound for residential fuel combustion impacts based on current SOA model formulations. Total anthropogenic SOA estimates from all sectors were very low compared to contributions from this sector alone to secondary inorganic PM2.5 and primary PM2.5.

Operational Model Performance Description

Speciated PM2.5 data from the IMPROVE and STN networks are compared to model predictions to estimate operational model performance. Model estimates are compared to observations collected during 2011. Metrics used to describe model performance include mean bias and gross error (Boylan et al., 2006). The bias and error metrics describe performance in terms of measured concentration. The best possible performance is when the metrics approach 0.

Model performance is shown for PM2.5 organic in Figure 3, which shows the average bias for 24-hr average model-observed pairs during Quarter 1 (January, February, and March) for all monitors in the modeling domain. Warm colors indicate the modeling system is overpredicting and cool colors indicate the modeling system is underestimating observations. The model tends to overpredict PM2.5 organic carbon in the northeast and in certain urban areas of the Midwest including St. Louis, Kansas City, and Louisville.

Figure 3. Quarter 1 average PM2.5 organic carbon bias at all CSN/STN and IMPROVE monitors.



Source Contribution Estimates

The Quarter 1 average contribution estimated by the model for the residential fuel combustion sector is shown in Figure 4. These results are not adjusted to account for differences between model and observation data. Figure 5 shows the same information but for SCC level sub-categories of the sector.

Figure 4. Average of quarter 1 (Jan, Feb, Mar) contribution from the residential wood combustion sector to PM2.5.

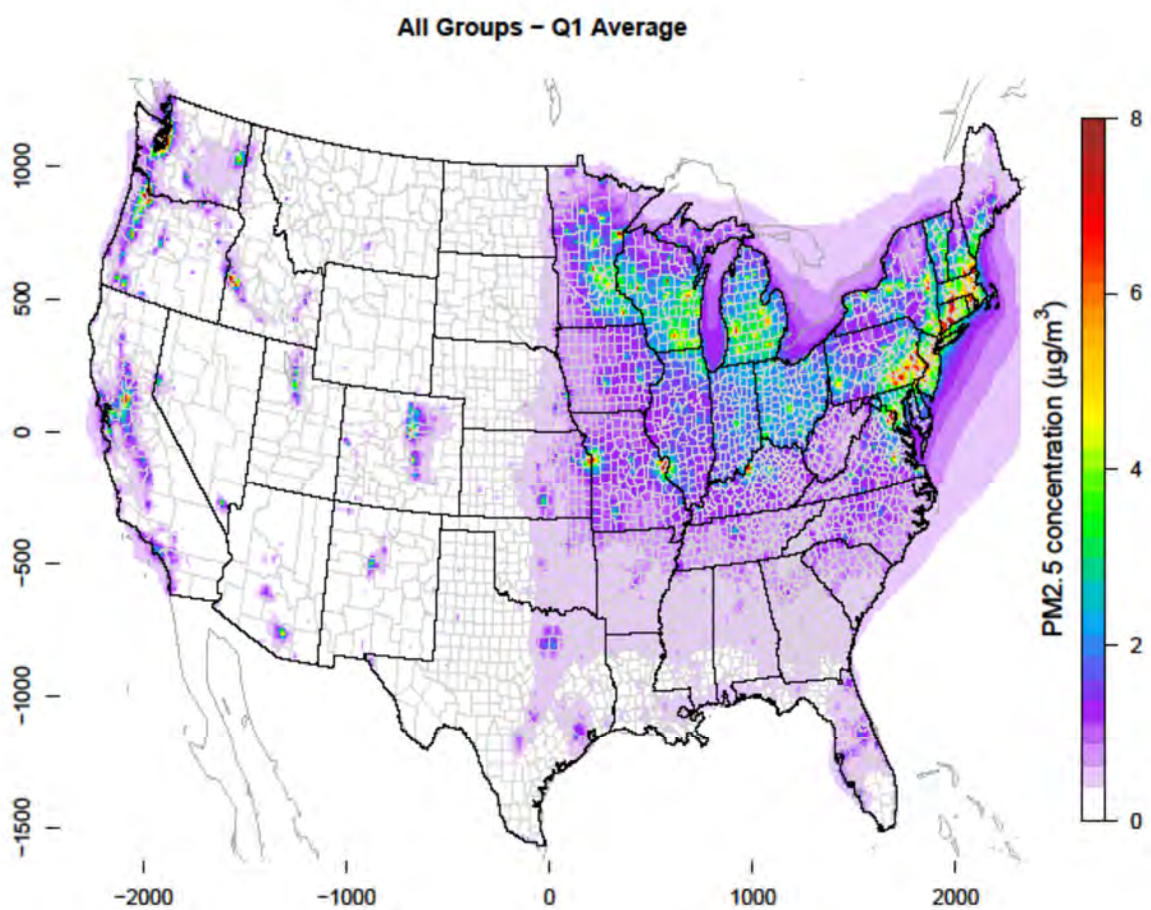
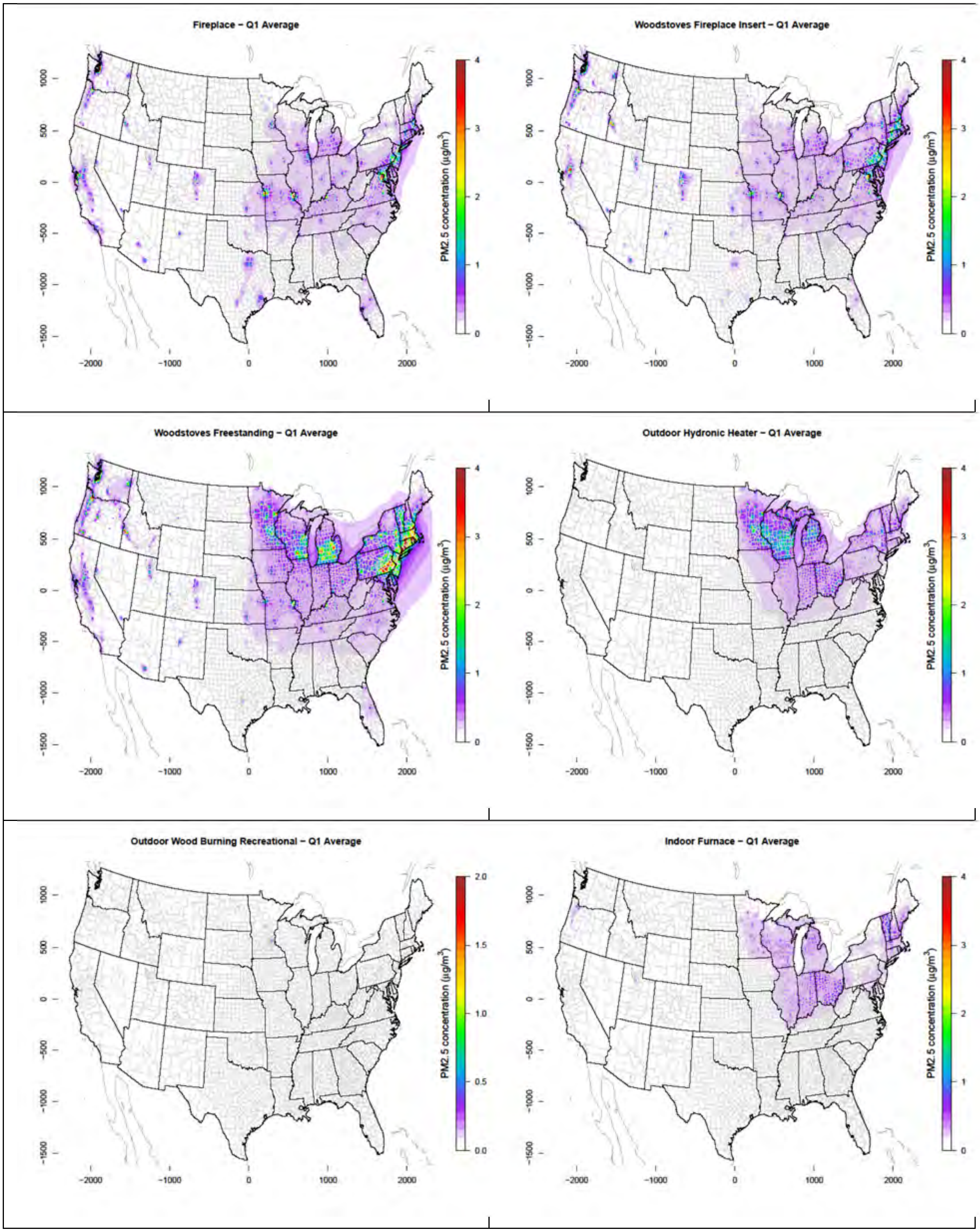
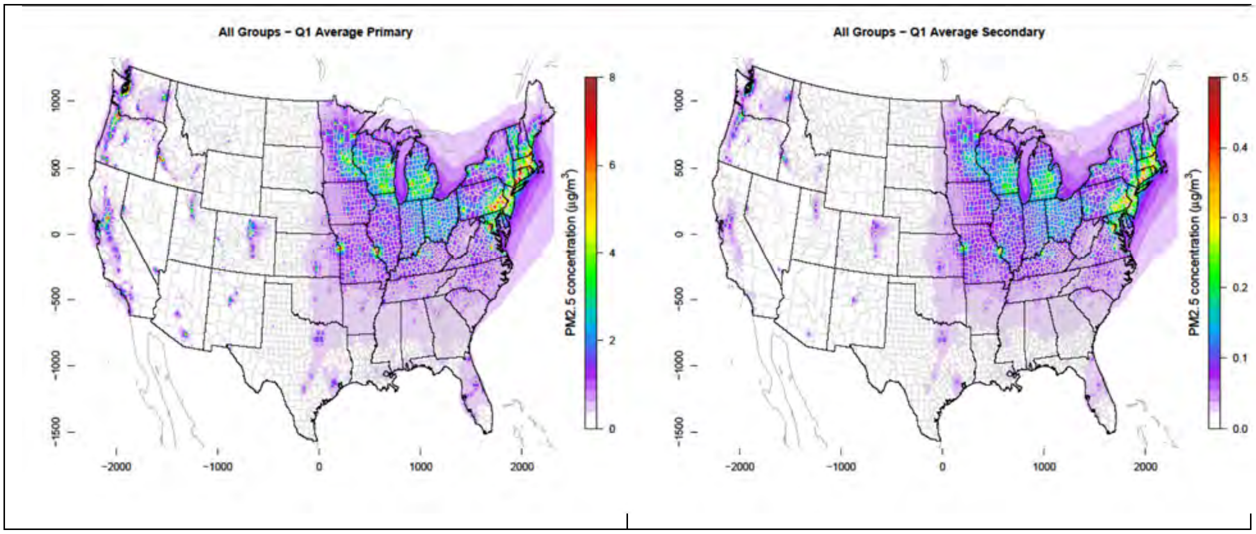


Figure 5. Contributions to quarter 1 average PM2.5 are further broken out by sub-categories including fireplaces, wood stoves, outdoor hydronic heaters, outdoor recreational devices, and indoor furnaces.



Quarter 1 average contribution is largely based on emissions of primarily emitted PM2.5 rather than secondarily formed inorganic PM2.5 (see Figure 6).

Figure 6. Quarter 1 average PM2.5 from the residential wood combustion sector. Contribution is shown from primarily emitted PM2.5 (left) and from precursor emissions NOX, SO2, and NH3 (right).



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