# Supporting information for: Emissions Inventory of $PM_{2.5}$ Trace Elements across the United States

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64 pages, 23 figures, 6 tables

# S1 Introduction

Development of the speciation profiles for each source category involved deliberating over numerous details regarding the data and meta-data of the SPECIATE profiles, the source-specificity of SCCs in the NEI, the profile compositing process, the evaluation of the speciated $PM_{2.5}$ inventory against the HAP inventory, and the computation of derived chemical species (e.g. metal-bound oxygen and non-carbon organic matter). In addition, the resulting emissions inventory contains a great amount of spatial-, chemical- and source-specific information that can be subsetted in various ways for further study. This Supporting Information is intended to house all of the details that are too numerous to fit in the Methods and Results sections of the main paper, but which are potentially useful for readers who wish to conduct similar analyses and see a more complete set of results.

# S2 Source Categorization Scheme

The 66 $PM_{2.5}$ source categories used for speciation of the 2001 CAIR NEI (see Table S1 by Bhave et al. (2007) (*1*)) were used as our starting point because a cross-reference table that maps all 3497 SCCs to one of those 66 source categories had been developed and reviewed extensively prior to our study. The steps below were then taken to develop the expanded set of 84 source categories used in this work.

First, we assessed the underlying speciation data for the original 66 source categories. On this basis, 6 of the categories (liquid waste combustion, jet fuel combustion, carbon black manufacturing, basic oxygen furnace, organic liquid, and nonroad diesel exhaust) were deleted because no source-specific measured profiles were available to speciate their emissions. In addition, 3 of the original 66 source categories were renamed to reflect better the emission source from which their underlying speciation data were collected: “residential heating” was renamed as “Residential Natural Gas Combustion”, “organic chemical manufacture” was renamed as “Urea Fertilizer”, and “paint manufacture” was renamed as “Surface Coating”.

Second, we identified source categories that could be subdivided due to the availability of source-specific profiles in SPECIATE v4.0 and the presence of SCCs with comparable specificity. The coal-combustion category was split into industrial-scale combustion of bituminous, subbituminous, and lignite coals, and a fourth category was created for residential coal combustion. The soil-dust category was subdivided into Unpaved Road Dust, Agricultural Soil, Construction Dust, Dairy Soil, Limestone Dust, Crustal Material, and Industrial Soil. The charcoal broiling category was divided into Charbroiling and Meat Frying. By subdividing 3 of the original categories into 13 new categories, 10 additional source categories were added to the list.

Third, we identified SCCs for which source-specific profiles are in SPECIATE 4.0 but the existing source categories were too generic. To improve the speciation of these SCCs, 13 new source categories were created: Auto Body Shredding, Boric Acid Manufacturing, Calcium Carbide Furnace, Cast Iron Cupola, Coke Calciner, Inorganic Fertilizer, Lime Kiln, Potato Deep-Frying, Prescribed Burning, Process Gas Combustion, Slash Burning, Sludge Combustion, and Steel Desulfurization. In the process of adding these source categories, the original petroleum-heater category was rendered obsolete and deleted from the list.

Fourth, new source categories were created for tire dust and brake lining dust emissions because the 2001 $PM_{2.5}$ NEI does not itemize those components of motor vehicle emissions. Contributions from each of the motor vehicle processes were therefore disaggregated by applying emission factors obtained from a base run of the MOBILE 6 model (`http://www.epa.gov/otaq/m6.htm`). The brake/tire/exhaust fractions used are shown in Table S1. These fractional contributions were multiplied by emissions from the corresponding SCCs as shown in Table S1. The resulting emissions were then put into the Brake Lining Dust, Tire Dust, and Exhaust source categories (the latter specific to vehicle type) for subsequent speciation.

**Table S1.** Fractions used to subdivide $PM_{2.5}$ emissions from motor vehicle exhaust SCCs into the Brake Lining Dust, Tire Dust, and Exhaust source categories.

| SCC Type | Source Category | Brake | Tire | Exhaust |
|---|---|---|---|---|
| Heavy Duty Diesel Vehicles | HDDV | 0.0097 | 0.0119 | 0.978 |
| Light Duty Diesel Vehicles | LDDV | 0.019 | 0.0072 | 0.974 |
| Light Duty Diesel Trucks | LDDV | 0.0196 | 0.0074 | 0.973 |
| Light Duty Gasoline Vehicles | Onroad Gasoline | 0.329 | 0.124 | 0.547 |
| Light Duty Gasoline Trucks | Onroad Gasoline | 0.256 | 0.097 | 0.647 |
| Buses | Onroad Gasoline | 0.0097 | 0.0119 | 0.978 |
| Motorcycles | Onroad Gasoline | 0.245 | 0.046 | 0.708 |

Overall, 25 new source categories were added and 7 were deleted from the original list. The final set of 84 source categories is shown in Figure 2 of the main paper and the cross-reference table mentioned above was revised to map each SCC to one of these 84 categories.

# S3 Speciation Profiles for Emission Source Categories

We began developing speciation profiles for each source category in November 2005 using an in-house pre-release version of the SPECIATE database. From March - May 2006, our preliminary set of profiles was reviewed by a large number of emission experts and source-testing specialists from various Offices within the EPA. In December 2006, those profiles were included in the public release of SPECIATE v4.0. In 2007-2009, further analyses of those profiles were conducted by the authors of this paper, resulting in a number of refinements and enhancements. A complete description of the methods and procedures we followed to arrive at the final profiles is provided in this section. The final set of profiles is given in Table S6 and will be incorporated into the public release of SPECIATE v4.3.

## S3.1 Unused SPECIATE Profiles

Numerous profiles from SPECIATE v4.0 were excluded from use in this study for a variety of reasons. Over 3000 profiles were found to be derived from measurements of $PM_{10}$ or larger particles, and 88 were from $PM_1$ or smaller. We also found that 268 of the $PM_{2.5}$ profiles were

collected outside of the U.S. (e.g. emissions from a grilled chicken shop in Mexico City) and did not appear to be appropriate for matching to any SCCs in the NEI. Many of the SPECIATE profiles that we selected are composites of other profiles that are also in SPECIATE, so the latter were excluded to prevent double weighting. The database also includes roadside and tunnel samples that we excluded because those are aggregates of numerous fuel and vehicle types whose emissions are separately classified in the NEI. Finally, a small number profiles were excluded due to outdated sampling methodologies, obsolete source technologies, and erroneous data (i.e. sum of measured species was $> 1$). After excluding these profiles, we were left with about 300 high-quality profiles from which to develop the final set of source-category-specific emission profiles.

## S3.2 Source Categories with One Speciation Profile

For some source categories, only 1 raw profile from SPECIATE v4.0 was found and used directly in the present study. These source categories are listed in Table S2, along with the corresponding profile numbers from SPECIATE v4.0.

**Table S2.** Source categories for which only one raw SPECIATE v4.0 profile was available.

| Source Category | SPECIATE Profile # |
|---|---|
| Aluminum Production | 291012.5 |
| Ammonium Nitrate Production | 254092.5 |
| Ammonium Sulfate Production | 254102.5 |
| Asphalt Manufacturing | 4083 |
| Asphalt Roofing | 255002.5 |
| Auto Body Shredding | 171092.5 |
| Boric Acid Manufacturing | 254052.5 |
| Brick Grinding and Screening | 4079 |
| Calcium Carbide Furnace | 252012.5 |
| Charcoal Manufacturing | 253022.5 |
| Coke Calciner | 262062.5 |
| Dairy Soil | 3458 |
| Distillate Oil Combustion | 4736[3] |
| Fiberglass Manufacturing | 4708[1] |
| Food & Ag-Drying | 4706[1] |
| Glass Furnace | 271022.5 |
| Gypsum Manufacture | 275012.5 |
| HDDV Exhaust | 3914 |
| Heat Treating | 286012.5[1] |
| Inorganic Chemical Manufacturing | 900022.5 (OC and EC = 0)[2] |
| Kraft Recovery Furnace | 231042.5 |
| Lead Processing | 204012.5 |
| Lead Production | 293302.5 |
| Meat Frying | 160012.5 |
| Nonroad Gasoline Exhaust | 4559[5] |
| Open Hearth Furnace | 283022.5[1] |
| Potato Deep-Frying | 4655 |
| Residential Natural Gas Combustion | 421072.5 |
| Residual Oil Combustion | 4737[4] |
| Sea Salt | 431012.5 |
| Steel Desulfurization | 283062.5 |
| Urea Fertilizer | 254042.5[1] |
| Wood Products - Drying | 221012.5 |
| Wood Products - Sanding | 222012.5[1] |
| Wood Fired Boiler | 4704 |

[1]Volatile Carbon (VC) in these profiles was treated as a synonym for OC
[2]Trace elements were removed from this profile (see Section S3.5).
[3]Trace-elemental data were added to this profile based on Appendix M (medians of data measured by EDXRF in Table M-6) of an EPA case study report (*2*).
[4] Trace elements were added from Table 2 of Hays et al. (*3*).
[5] Denuded measurements of OC and EC from the primary reference were used rather than the non-denuded values provided in SPECIATE v4.0.

## S3.3 Compositing Methodology

In cases where multiple high-quality profiles were available for a single source category, profiles were averaged together to create a *composite profile*. The general procedure for creating composite profiles was to compute the median of SPECIATE profiles that were grouped into the given source category. The median was chosen over the mean to help mitigate possible large errors stemming from the presence of outlier samples and measurements. Missing values were excluded from the

median calculations, whereas zeros were included. In cases where multiple measurements of a single element were available (e.g. Cl by XRF and $Cl^-$ by IC), the XRF-based values were used because those are assumed to represent the total concentration of all forms of the given element. Table S3 lists the individual profiles from SPECIATE v4.0 that were subjected to this compositing process.

**Table S3.** Source categories for which multiple raw SPECIATE v4.0 profiles were used to develop a composite profile.

| Source Category | SPECIATE Profile # |
|---|---|
| Agricultural Burning | 3243, 3258, 3448, 3453, 423222.5, 423232.5, 423242.5, 4390, 1 subcomposite |
| Agricultural Soil | 3196, 3298, 3308, 3313, 3333, 3338, 3358, 3363, 3393, 3428, 3443, 3488, 3497, 4071 |
| Aluminum Processing | 201012.5[1], 201022.5, 201032.5 |
| BituminousCombustion | 3690, 3694, 3700 |
| Brake Lining Dust | 340042.5, 340062.5, 340072.5 |
| Catalytic Cracking | 262092.5, 3231, 4375, 4411 |
| Cement Production | 272012.5, 4378 |
| Charbroiling | 160002.5, 3915, 4383, 4554 |
| Construction Dust | 272042.5, 3491, 441012.5 |
| Copper Production | 292022.5, 292032.5, 292042.5, 292052.5, 292062.5, 292072.5, 292082.5, 292092.5, 292102.5, 292112.5 |
| Crustal Material | 3553, 3717, 4075, 413502.5, 413512.5, 413532.5, 433052.5, 433062.5, 433072.5, 433092.5, 4351, 4353 |
| Electric Arc Furnace | 283052.5, 3989, 3997 |
| Ferromanganese Furnace | 284012.5, 3993 |
| Fly Ash | 3547, 3985, 433032.5, 4374 |
| Food & Ag - Handling | 214012.5, 4073 |
| Industrial Soil | 3538, 3550, 3556, 3965, 3973, 3979, 413312.5 |
| Inorganic Fertilizer | 254082.5, 254152.5, 254182.5 |
| LDDV Exhaust | 321042.5, 3912, 3963, 4675 |
| Lignite Combustion | 4367, 4368, 4369, 4370 |
| Lime Kiln | 232022.5 ,276022.5, 3999 |
| Limestone Dust | 211012.5, 433042.5 |
| Natural Gas Combustion[3] | 4403, 4398 |
| Onroad Gasoline Exhaust | 311072.5, 3517, 3884, 3892, 3904, 3947, 3951, 3955, 3959, 4558 |
| Paved Road Dust | 3197, 3201, 3541, 3559, 3713, 3938, 4067, 411302.5, 411372.5, 411382.5, 4347, 3 subcomposites |
| Phosphate Manuf | 254072.5, 254112.5, 254122.5, 254132.5, 254142.5, 254162.5, 254172.5, 254192.5, 254202.5, 254212.5, 254222.5 |
| Prescribed Burning | 423212.5, 4463, 4464, 4465, 4466, 4467, 4468 |
| Process Gas Combustion | 4398, 4407, 4415 |
| Pulp & Paper Mills | 4733, 4734, 4735 |
| Residential Coal Combustion | 3761, 432012.5 |
| Residential Wood Combustion | 3770, 3920, 421032.5, 422022.5, 422042.5, 4384, 4644, 3931, 421012.5, 422032.5, 4387, 4645 |
| Sand & Gravel | 3202 , 3318, 3981, 4087, 441032.5 |
| Sandblast | 257022.5[2], 441042.5, 4707 |
| Sintering Furnace | 283012.5, 3991 |
| Solid Waste Combustion | 113012.5, 171082.5, 3288 |
| Sub-Bituminous Combustion | 3191, 3192 |
| Surface Coating | 4663, 4664 |
| Tire Dust | 340032.5, 340082.5 |
| Unpaved Road Dust | 3204, 3515, 3719, 4069, 412202.5, 4349, 441022.5, 4 subcomposites |
| Wildfires | 3766, 4366 |
| Wood Products - Sawing | 222022.5, 4709[1] |

[1] Volatile Carbon (VC) in these profiles was treated as a synonym for OC

[2] This profile contains neither OC nor EC, but it does contain total carbon (TC). OC was copied from the TC value, assuming EC from this source is negligible.

[3] Data was adjusted by source experts (Ron Myers and Barrett Parker of EPA) to account for $PM_{2.5}$ loss on the walls of the sampling train using test reports at `http://www.nyserda.org/programs/Environment/EMEP/finalreports.asp`.

Different analytical methods can yield very different results for OC and EC in a sample, but total carbon (TC) results are fairly consistent across the methods (*4*). To ensure that 1) the sum of OC and EC in each composite is in agreement with median TC values in the individual profiles and 2) the composite values of OC and EC are reasonably consistent with measurements taken by a thermal-optical reflectance (TOR) method (*5*), the raw OC and EC weight fractions were modified in the following manner prior to compositing. The OC and EC fractions were summed to calculate TC in each source profile with non-missing OC and EC values; profiles with missing values were not considered in this compositing process. If any SPECIATE profiles in a source category measured carbon using a TOR method, those profiles were used to compute $median(OC)/median(TC)$ ratios for the source category. These ratios were multiplied by the TC values from all non-TOR profiles in the same source category to estimate OC values that might have been measured if the TOR method were used. EC was then re-computed as TC minus the estimated OC value for each non-TOR profile.

If no SPECIATE profiles in a source category included TOR measurements of carbon, then the $median(OC)/median(TC)$ ratio computed using all profiles in that category were multiplied by TC to obtain OC values. An exception to this rule was made for three source categories (Wildfires, Wood Fired Boiler, and Nonroad Gasoline Exhaust) because they have no TOR data and they are among the ten largest carbon-emitting source categories in the U.S. For the Wildfires category, OC and EC were redistributed using the $OC/TC$ ratio from our Prescribed Burning profile. For the Wood Fired Boiler category, the $OC/TC$ ratio from our Residential Wood Combustion profile was used. For the Nonroad Gasoline Exhaust category, the $OC/TC$ ratio from a high-emitting gasoline exhaust profile (# 3955) was used because the carbon distribution from high-emitting vehicles might resemble those of a nonroad gasoline engine.

During our survey of the SPECIATE data, a number of raw profiles were found to be repeat samples from a single study. To prevent these samples from over-weighting the composite profile for their assigned source category, sub-composites of the similar profiles were created prior to their

inclusion in the final composite for the given source category. Sub-composites were created in the same manner as the main composites (e.g. median of $PM_{2.5}$ species weight fractions, similar treatment of OC/EC data as described above). Information about the creation of the sub-composites is shown in Table S4.

**Table S4.** Sub-composites made during the compositing of SPECIATE profiles.

| Source Category | Sub-composite Description | SPECIATE Profile # |
|---|---|---|
| Agricultural Burning | Rice & Straw Burning | 4391, 4392 |
| Paved Road Dust | Central CA | 3303, 3328, 3348, 3383, 3388, 3423, 3433 |
| | Phoenix | 3500, 3503 |
| | Robbins, IL | 3971, 3975, 3977 |
| Unpaved Road Dust | Central CA | 3323, 3343, 3353, 3368, 3373, 3378, 3438 |
| | Ohio | 3532, 3535 |
| | Phoenix | 3506, 3509 |
| | Robbins, IL | 3967, 3969 |

## S3.4 Hybrid Profiles

For some categories, it was deemed appropriate to combine a subset of species values from one profile with a different subset of species values from another profile to make a *hybrid profile*. For example, several of the raw profiles in SPECIATE v4.0 contain data on the trace-elemental composition but they are missing values for OC, EC, and $NO_3^-$ . In such cases, the mass fractions of OC, EC, and $NO_3^-$ are copied from the most comparable profile in the database. Source categories for which a hybrid profile was developed are discussed below.

1. Cast Iron Cupola: Hybrid was created from elements in the Cast Iron Cupola raw profile (# 282022.5) and EC, OC, and $NO_3^-$ values from the Heat Treating profile (# 286012.5).

2. Copper Processing: OC, EC, and $SO_4^{2-}$, are from profile 205032.5; S is computed from $SO_4^{2-}$ (see Section S3.7.5); and the remaining trace elements are composited from 295012.5 and 295022.5 (see Section S3.6).

3. Sludge Combustion: OC, EC, and $NO_3^-$ were copied from the Solid Waste Combustion composite profile. Trace elements are taken from profile # 171202.5, which sampled from sewage sludge incineration.

4. Slash Burning: $NO_3^-$ value was copied from the Wildfires composite profile, other species were composited from twelve profiles in the Slash Burning category (profiles 423052.5, 423062.5, 423072.5, 423082.5, 423092.5, 423102.5, 423112.5, 423122.5, 423132.5, 423142.5, 423152.5, 423162.5)

## S3.5 "AVG" Profiles

The original 66 speciation profiles that were the starting point for our source categorization scheme contained five profiles named "Chemical Manufacturing - Avg" (SPECIATE v4.0 profile # 900022.5),"Industrial Manufacturing - Avg" (900162.5), "Mineral Products - Avg" (900132.5), "Petroleum Industry - Avg" (900142.5), and "Overall Average-Default" (000002.5; renamed in this work to "Misc. Sources"). The data in these profiles were deemed to be of low quality due to 1) the lack of source specificity, and 2) the fact that many of the values for trace elements appeared to be imputed using a single value because measurements were below detection limits. For many of the SCCs to which these profiles were originally mapped, no newer or more source-specific profiles were available in SPECIATE v4.0.

Our initial inclination was to remove these profiles entirely and map all of their emissions to PMO, but this resulted in an unacceptable decrease in air quality model predictions of OC and EC. As a compromise solution, we therefore decided to remove the trace elemental values and flag them as missing to indicate the need for their future research; the OC, EC, $SO_4^{2-}$, $NO_3^-$, and $NH_4^+$, values were retained due to their historical usage in SMOKE. Following that rationale, we also removed all trace elements from Inorganic Chemical Manufacturing, which was derived from the Chemical Manufacturing - Avg. profile (see Table S2).

## S3.6 Comparison to 2002 HAP Inventory

EPA's emissions inventories, including those of the HAPs, are typically built "bottom-up" using population activity data, emissions factors, and emissions reported by the state and regional agencies (`http://www.epa.gov/ttn/chief/index.html`). In the HAP inventory, raw emissions of the 10 toxic metals (As, Cd, Co, Cr, Hg, Mn, Ni, Pb, Sb, Se) are estimated in their compound forms (e.g. ammonium dichromate instead of Cr) in total suspended particulate matter; to convert the HAP emissions data to $PM_{2.5}$ elemental emissions for comparison with our speciated inventory, the following formula was applied:

$$E_{El} = \sum_{i}^{n} E_i \cdot St_i \cdot F_i \tag{S1}$$

where $E_i$ is the emission of compound $i$ reported in the HAP inventory, $St_i$ is the stoichiometric ratio of the element's mass to the compound's mass, $F_i$ (obtained from the "Toxicity Weighting Factors" database located at `ftp://ftp.epa.gov/EmisInventory/2002finalnei/documentation/point/augmentation_point/tox_wt_factors_2002nei_011007.mdb`) is the mass fraction of compound $i$ that is emitted in the $PM_{2.5}$ size range, $n$ is the number of compounds in the HAP inventory that contain element $El$, and $E_{El}$ is the $PM_{2.5}$ emission of trace element $El$ calculated from the HAP inventory. $F_i$ values were not available for Hg-containing compounds; instead, fractions of the amount of Hg in the particulate divalent form were applied (*6*) to non-specific Hg emissions, and all particulate Hg was assumed to be present in the fine fraction of PM emissions (*7*). In addition, any ligands on Hg atoms were assumed to have an insignificant mass relative to that of the Hg atom, so that a $St_i$ value of 1 was used for those compounds.

Equation (S1) was applied to calculate emissions of each of the HAP elements from each SCC in the 2002 HAP inventory. This resulted in non-zero emissions for at least one of the 10 HAP metals for 1393 SCCs. This count is much smaller than the number of SCCs in the 2001 $PM_{2.5}$ NEI (3497) in part because the HAP NEI does not contain certain major PM-emitting source categories

such as fugitive dust and wildfires.

Data by which to map metal emissions from the HAP inventory to the metal emissions in our speciated $PM_{2.5}$ inventory at both the SCC and county levels were not available. Large differences in source category totals were therefore used to qualitatively flag possible errors in our speciation profiles. Source-category specific metal emissions that were found to be substantially different (leading to a difference that was at least 20% of the domain-wide total emission of the given metal in the HAP inventory) between the two inventories are discussed below. The results of making the adjustments discussed below are illustrated in Figure S1. Figure S1 shows that at the national level, all metal emissions in our speciated $PM_{2.5}$ NEI were initially greater than the HAP NEI totals. The adjustments described below achieved some reduction of all 10 HAP metals in our inventory. In the cases of As, Co, Mn, and Pb, the adjustments brought our inventory to a level lower than (but closer to) that of the HAP NEI.

1. Aluminum Processing: Sb values in our speciated $PM_{2.5}$ inventory are substantially higher than in the HAP inventory (104 vs 1.75 ton/yr). All 3 profiles in this category were collected prior to 1990. One profile (201012.5) is missing a Sb value, but the other two profiles (201022.5 & 201032.5) have similar mass fractions for Sb (0.00597 & 0.0057). Figure S15(c) shows emissions from this source category in numerous states, so our inventory value of 103 ton/year might be more reasonable than the (State reported) 2002 value of 1.75 ton/year. Therefore, no adjustment was made to the profiles in this source category.

2. Bituminous Combustion: Se emissions are much lower in the HAP NEI than in our speciated $PM_{2.5}$ inventory (315 vs. 1334 ton/year). Figure S10(e) shows that over 400,000 tons of $PM_{2.5}$ were emitted from this source category in 2001, so our inventory total for Se from Bituminous Combustion appears reasonable. Arsenic has a value of 0 in all three SPECIATE profiles in this source category and thus in our speciated inventory, but the HAP inventory reports 74 ton/year, suggesting that analytical methods with lower detection limits (e.g. ICP-MS) need to be used for measuring As emissions from Bituminous Combustion in the future.

In the present study, no adjustments were made to the Se and As mass fractions in our composite profile.

3. Cast Iron Cupola: Sb emissions are substantially larger (16.81 ton/yr) in our speciated inventory than in the HAP inventory (0.032 ton/yr). This might be due to incomplete reporting of emissions to the HAP NEI, since the Air Emissions Regulatory Rule does not require reporting of Sb emissions (*8*). No adjustment was made to the HAP metals in our profile for this source category.

4. Copper Processing: Sb emissions were initally much higher in our speciated $PM_{2.5}$ inventory than the HAP inventory because an antimony roasting profile (205032.5) was being used to develop the original composite profile for this category. Upon inspection of the other available profiles in SPECIATE v4.0, we decided to use only the OC, EC, and $SO_4^{2-}$ values from the antimony roasting profile and obtain the remaining speciation fractions from two Copper Processing profiles (see Section S3.4). Initially, Pb emissions from this source category were also much higher in our inventory than the HAP NEI. Since all the SPECIATE profiles in this category pre-date the 1990 Clean Air Act amendments, the Pb values are removed from our final profile and treated as missing (see Table S6).

5. Copper Production: The mass fraction of As in profile #292022.5 is very high (0.92449) and comes from a 1978 EPA report. This mass fraction was set to missing since the profile is likely outdated due to the 1990 Clean Air Act amendments. Pb values were also found to be very high, reflecting the outdated measurements, so the composite Pb value was set to missing. Sb from this source was also flagged by our screening method, but was not changed in our composite profile because Sb emissions from this source were not inventoried in the HAP NEI.

6. Electric Arc Furnace: Large Cr emissions versus 16 ton/yr in the HAP NEI were being generated in the our inventory (128 ton/yr) due to one profile (283052.5) in this category. Large emissions of Cr seem to be unlikely since concentrations are strictly limited by the

1990 Clean Air Act amendments, so this profile's Cr value was changed to missing in our final inventory.

7. Ferromanganese Furnace: The emission of Mn from this source category was found to be much higher in our speciated $PM_{2.5}$ inventory than in the HAP inventory (1664 ton/yr vs. 2.26 ton/yr). One profile (284012.5) is from a 1979 reference, and the other (3993) is from a poor quality (based on the Quality rating in the SPECIATE database) dust sample in South Africa, neither of which is likely to be representative of modern U.S. emissions. There are also very few locations where $PM_{2.5}$ from this source category is emitted (see Figure S18(d)). To prevent over-prediction of Mn from this source category, the Mn composite weight fraction was regarded as missing in our final inventory.

8. Heat Treating: Ni and Cr emissions are much greater in our speciated $PM_{2.5}$ inventory (541 and 149 ton/year) than in the HAP inventory (6.1 and 7.3 ton/yr). The spatial map of Heat Treating shows widespread emissions (see Figure S13(f)), making incomplete reporting from the states a likely possibility. We therefore deemed our emissions to be reasonable, and the profile was not adjusted.

9. Lead Processing: Profile #204012.5 is from a 1979 source in which the Pb weight fraction is 0.5. The Lead Processing spatial map only has a few sources (see Figure S21(c)), so the HAP inventory value (27.5 ton/yr) seemed more reasonable than our initial inventory (683 ton/yr). Controls are also likely to have been updated for Pb since it is a criteria pollutant, so the Pb weight fraction in Lead Processing was regarded as missing in our final inventory to prevent gross over-predictions.

10. Nonroad gasoline exhaust: Pb and Mn emissions were initially very high in our speciated $PM_{2.5}$ inventory vs. the HAP inventory for this source category. Profile #311082.5 was subsequently excluded from our analysis, because we discovered that it originated from a sample of leaded gasoline exhaust (*9*), which is not representative of current emissions.

11. Open Hearth Furnace: Only one SPECIATE profile was put into this category (283022.5), which is from a 1979 report. The Cr value is 0.02, leading to 297 ton Cr/yr in our speciated inventory (vs 8.9 in the HAP NEI). The spatial map of Open Hearth Furnace (Figure S15(d)) shows a number of points around the country with emissions so, despite the large discrepancies in the national total, our profile was left as is.

12. Prescribed Burning: Cd was found to have a substantial weight fraction in our composite profile for this source category. The reference for profile 4467 states that heavy metals can hyper-accumulate in vegetation (*10*). Therefore, Cd and other metals in our Prescribed Burning profile were not changed due to the fact that prescribed burning is spread through areas where anthropogenic combustion occurs (compare Figure S11(d) with Figures S10(e) and S11(a)), giving ample opportunity for the hyper-accumulation process to take place.

13. Residential Coal Combustion: Similar to Cast Iron Cupola, Sb emissions are much larger in our speciated inventory (10.62 ton/yr) than in the HAP inventory (0.035 ton/yr), which again may be due to optional reporting practices. Thus, this profile was left unchanged.

14. Residual Oil Combustion: Ni (25.6 ton/yr), Cr (no data), Se (2.66 ton/yr) and As (no data) were much smaller in our speciated inventory than in the HAP NEI (269, 106, 609 ton/yr and 109 ton/yr), and might be underestimated in our speciation profile for this source category. More emissions measurements of this source category (only one profile was used in our speciated inventory) may help reconcile these differences. In the present study, our profile for Residual Oil Combustion was left unchanged.

15. Sintering Furnace: Cr, Pb, and Cd emissions were found to be higher in our initial speciated $PM_{2.5}$ inventory (296, 552, and 54 tons/yr) than in the HAP inventory (1.2, 16.9, and 1.0 tons/yr). One of the profiles (283012.5) was collected in the 1970s and is expected to be outdated with regards to HAP metals. Therefore, mass fractions of Cr, Pb, and Cd were removed from that profile prior to compositing with the other available profile for this source

category (3991). As a result, all three elements in our final inventory match the HAP NEI values within about 1 ton/yr.

16. Solid Waste Combustion: In our initial inventory, Pb and Hg appeared questionable. The composite contains 3 profiles, all of which were collected in the 1980s. Only one of the individual profiles contains Hg (0.0135), whereas all of them contain Pb (median = 0.058). A large region of the eastern U.S. has emissions from Solid Waste Combustion (see Figure S14(c)), so uncertain state reporting could mean that the large emissions of Hg and Pb in our inventory (1391 and 324 ton/yr) are more reasonable than the HAP NEI values (0 and 86 ton/yr). On the other hand, no details could be found about the sources sampled in our composite profile. Given the time period when the sampling was conducted (1980s), batteries might have been burned as part of the solid waste. The amount of mercury in batteries has been declining and is believed to be totally absent in present-day solid waste (*6*). Emissions from lead-acid batteries are expected to come from recycling facilties rather than general waste disposal facilities (*11*). Based on these considerations, the Hg and Pb values in our original Solid Waste Combustion composite profile were therefore removed when preparing the final inventory.

17. Sub-Bituminous Combustion: The emissions of As in our speciated inventory are 0 (vs 84 ton/yr in the HAP inventory), indicating that As could not be detected in either of the source samples used to develop our composite profile. A measurement technique with lower detection limits than XRF seems likely to yield a small non-zero value, which would result in an emission total that is comparable to the HAP NEI. As discussed above for Bituminous Combustion, this discrepancy with the HAP NEI reveals a need for future measurments, but no change was made to our composite profile for Sub-Bituminous Combustion.

18. Surface Coating: Sb emissions were higher in our speciated inventory (153 ton/yr) than in the HAP inventory (8.5 ton/yr). Only one of the 2 profiles (4663) has a non-zero Sb value (0.0142). A number of metallic compounds (especially $TiO_2$) are in paint, and Sb might

be an impurity from extracting or making these compounds. Surface coating operations are widespread (see Figure S14(f)), and are state-reported point sources, so the value in our inventory seem reasonable. Therefore, no change was made to this profile.

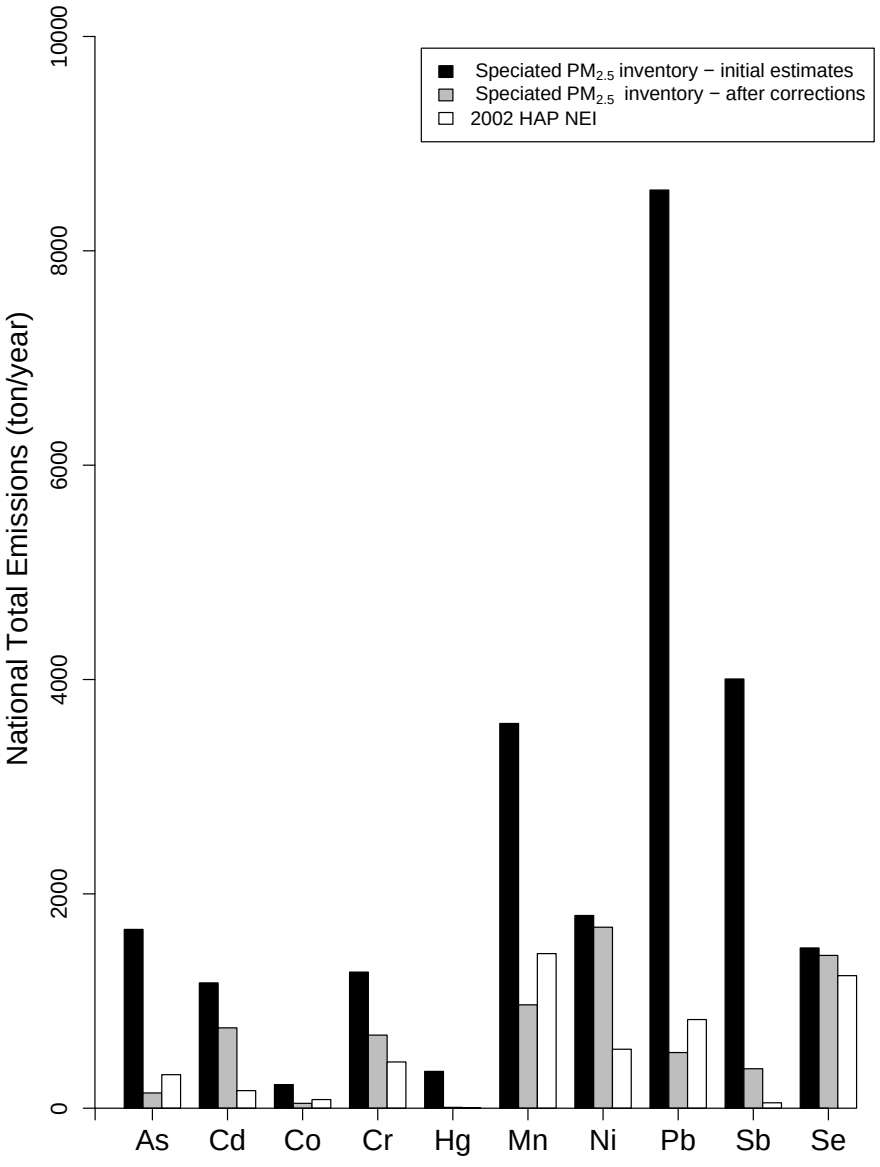



**Figure S1.** Total emissions of HAP metals from our 2001 speciated $PM_{2.5}$ inventory and from the 2002 inventory of HAP compounds. Data aggregated in the white bars were compared to data in the black bars, and a subset of our profiles were updated based on differences highlighted in the comparison. Data in the gray bars are the result of those updates. Note that only emissions from the 1393 SCCs common to both inventories (which included no fugitive dust and wildire SCCs) were used when performing these comparisons.

## S3.7 Mass Fractions of Unmeasured Species

To account for as much of the emitted $PM_{2.5}$ mass as possible, we calculated additional species that were not in the original raw measured profiles in SPECIATE v4.0. Details about these calculations are provided below.

### S3.7.1 Particulate Water

Particulate water ($H_2O$) emissions were calculated for each composite profile as 24% of the sum of $SO_4^{2-}$ and $NH_4^+$ emissions (*12*). $H_2O$ emissions from combustion and other high-temperature sources were forced to be 0 with the expectation that the water emitted from such environments is likely to be in the vapor phase. Sources considered to have no particulate $H_2O$ emissions are: Agricultural Burning, Bituminous Combustion, Calcium Carbide Furnace, Charbroiling, Charcoal Manufacturing, Distillate Oil Combustion, Electric Arc Furnace, Ferromanganese Furnace, Glass Furnace, HDDV Exhaust, Heat Treating, Kraft Recovery Furnace, LDDV Exhaust, Lignite Combustion, Lime Kiln, Meat Frying, Natural Gas Combustion, Nonroad Gasoline Exhaust, Onroad Gasoline Exhaust, Open Hearth Furnace, Prescribed Burning, Process Gas Combustion, Pulp & Paper Mills, Residential Coal Combustion, Residential Natural Gas Combustion, Residential Wood Combustion, Residual Oil Combustion, Sintering Furnace, Slash Burning, Sludge Combustion, Solid Waste Combustion, Sub-Bituminous Combustion, Wildfires, and Wood Fired Boiler.

### S3.7.2 Metal-Bound Oxygen

Metal-bound oxygen (MO) was calculated by multiplying most of the trace elemental emissions by an oxygen-to-metal ratio . These ratios were based on the expected oxidation states of the metals in the atmosphere. Table S5 shows the expected oxide forms of each metal, which are based on the most common oxidation states of the metals (*13*). Total MO was then calculated for each source category using the following equation:

$$MO = \sum_{El}^{n} Ox_{El} \cdot E_{El} \tag{S2}$$

where $Ox_{El}$ is the oxygen-to-metal ratio for metal $El$, and $E_{El}$ is the emission of metal $El$ after accounting for bonding with $SO_4^{2-}$ (explained in Section S3.8). For metals with more than one common oxidation state, the mean of the oxygen-to-metal ratios was used for the $Ox_{El}$ value (see Table S5). This is an extension of the assumption described by Malm et al. (*14*), where 2 common forms of Fe are assumed to exist in ambient particulate matter in equal quantities. The list of metal oxides in Table S5 is inclusive of metal oxide forms used in some previous studies of particulate matter (*14–16*). In the Sea Salt profile, MO is forced to be zero because the Na, Mg, Ca, and K ions are assumed to be neutralized by $Cl^-$ and $SO_4^{2-}$ rather than oxygen (*17*). In the Agricultural Burning profile, we assumed all K to be in the form of KCl rather than $K_2O$.

**Table S5.** Assumed oxide forms of each metal and resulting mean oxygen-to-metal ratio used in Equation S2 to calculate the emissions of Metal-bound Oxygen (MO).

| Species | Oxide Form 1 | Oxide Form 2 | Oxide Form 3 | Oxygen/Metal Ratio |
|---|---|---|---|---|
| Na | $Na_2O$ | | | 0.348 |
| Mg | $MgO$ | | | 0.658 |
| Al | $Al_2O_3$ | | | 0.889 |
| Si | $SiO_2$ | | | 1.139 |
| P | $P_2O_3$ | $P_2O_5$ | | 1.033 |
| K | $K_2O$ | | | 0.205 |
| Ca | $CaO$ | | | 0.399 |
| Ti | $TiO_2$ | | | 0.669 |
| V | $V_2O_5$ | | | 0.785 |
| Cr | $Cr_2O_3$ | $CrO_3$ | | 0.692 |
| Mn | $MnO$ | $MnO_2$ | $Mn_2O_7$ | 0.631 |
| Fe | $FeO$ | $Fe_2O_3$ | | 0.358 |
| Co | $CoO$ | $Co_2O_3$ | | 0.339 |
| Ni | $NiO$ | | | 0.273 |
| Cu | $CuO$ | | | 0.252 |
| Zn | $ZnO$ | | | 0.245 |
| Ga | $Ga_2O_3$ | | | 0.344 |
| As | $As_2O_3$ | $As_2O_5$ | | 0.427 |
| Se | $SeO$ | $SeO_2$ | $SeO_3$ | 0.405 |
| Rb | $Rb_2O$ | | | 0.094 |
| Sr | $SrO$ | | | 0.183 |
| Zr | $ZrO_2$ | | | 0.351 |
| Mo | $MoO_2$ | $MoO_3$ | | 0.417 |
| Pd | $PdO$ | $PdO_2$ | | 0.226 |
| Ag | $Ag_2O$ | | | 0.074 |
| Cd | $CdO$ | | | 0.142 |
| In | $In_2O_3$ | | | 0.209 |
| Sn | $SnO$ | $SnO_2$ | | 0.202 |
| Sb | $Sb_2O_3$ | $Sb_2O_5$ | | 0.263 |
| Ba | $BaO$ | | | 0.117 |
| La | $La_2O_3$ | | | 0.173 |
| Ce | $Ce_2O_3$ | $CeO_2$ | | 0.200 |
| Hg | $Hg_2O$ | $HgO$ | | 0.060 |
| Pb | $PbO$ | $PbO_2$ | | 0.116 |

### S3.7.3 Non-Carbon Organic Matter

Non-Carbon Organic Matter (NCOM) was calculated for each source category by multiplying OC emissions by a source-category specific OM/OC (OM = organic matter) ratio to calculate an OM emission, and subtracting OC from OM. An OM/OC ratio of 1.25 was used for all motor-vehicle exhaust sources (the HDDV Exhaust, Nonroad Gasoline Exhaust, Onroad Gasoline Exhaust, and LDDV Exhaust source categories), which is a median of the values from Aiken et al. (*18*) (1.22, 1.25), Lipsky and Robinson (*19*) with artifact correction (1.4), Russell (*20*) (1.2, 1.3, 1.1), and Japar et al (*21*) (1.43). This ratio is also fairly consistent with the value of 1.2 used by Kleeman et al. (*15*) and Sheesley et al. (*22*), based on the measurements by Schauer et al. (*23*, *24*). An OM/OC ratio of 1.7 was used for wood combustion sources (the Wildfires, Agricultural Burning, Residential Wood Combustion, Prescribed Burning, and Slash Burning source categories), which is a median of the values from Aiken et al. (*18*) (1.55, 1.7), Lipsky and Robinson (*19*) with artifact correction (1.8), Hays et al. (*10*) (1.2), and Turpin and Lim (*25*) (1.9) - the 1.9 was computed from the organic-molecular data of Schauer et al. (*26*). The ratio of 1.7 is in agreement with the mass-closure estimates reported by Sheesley et al. (*22*) (1.7) and Bae et al. (*27*) (1.74), and falls in the range of estimates reported by Jimenez et al. (*28*) (1.5, 1.8, and 2.0). The Wood Fired Boiler category was originally assigned an OM/OC ratio of 1.7, but was later revised as explained in Section S3.8. An OM/OC ratio of 1.4 was applied to the emissions from all other source categories based on the long-standing value used in numerous studies of atmospheric $PM_{2.5}$ (*25*).

### S3.7.4 Ammonium

$NH_4^+$ values were imputed stoichiometrically in the profiles for the Ammonium Sulfate Production (assuming $(NH_4)_2SO_4$) and Ammonium Nitrate Production (assuming $NH_4NO_3$) source categories.

### S3.7.5 Sulfate and Sulfur

Many of the raw profiles contained a value for either $SO_4^{2-}$ or S, but not both. In these cases, stoichiometry was used to compute the missing value from the available measurement (assuming all S was present in the form of $SO_4^{2-}$ ). In profiles of the Ammonium Sulfate Production, Copper Processing, Lime Kiln, and Catalytic Cracking categories, both $SO_4^{2-}$ and S values were given in the data but they were not stoichiometrically consistent. In these cases S, was computed from $SO_4^{2-}$ due to the higher accuracy of ion chromatography vs. XRF (*29*).

## S3.8 Profiles In Which the Sum of Species $>$ 100% $PM_{2.5}$ Mass

After the calculation of $H_2O$, MO and NCOM, we found that a number of source categories' emissions summed to greater than 100% of the $PM_{2.5}$ emissions. To correct these profiles, the following steps were taken after all the calculations above were performed.

1. All $SO_4^{2-}$ not neutralized by $NH_4^+$ was assumed to be bonded to the metals in Table S5 and only the remaining available metallic mass was used in Equation (S2).

2. Residential Natural Gas Combustion, Prescribed Burning, Slash Burning, Surface Coating: OC measurements did not correct for artifacts. OC was scaled down until the sum of species in the profile equaled the total $PM_{2.5}$ mass.

3. Charbroiling: No OC artifact correction was done for three out of the four profiles used in this composite. OC in the composite was scaled down until the sum of species in the profile equaled the total $PM_{2.5}$ mass.

4. Residential Coal Combustion, Wildfires: No information about OC artifact correction was found in the raw profiles' references. We therefore assumed that no artifact correction was done and scaled OC in the composites until the sum of species in the profile equaled $PM_{2.5}$.

5. Process Gas Combustion, Lignite Combustion: Due to OC artifacts, the sum of species in some profiles in these categories was greater than the gravimetric $PM_{2.5}$ measurements.

Therefore, the original investigators normalized the mass fractions based on the sum of species, but they did not consider NCOM or MO during this adjustment. The raw, unnormalized data from these profiles could not be found, so all species were scaled down equally until 100% mass was reached.

6. Wood-fired boiler: The OM/OC ratio was changed to from 1.7 to 1.4 because a wood-fired boiler should not have as much oxygen as an open fire. This did not completely remove the mass overestimate so, after adjusting the OM/OC ratio, all species were scaled down equally until the sum of species equaled the total $PM_{2.5}$ mass.

7. Ammonium Sulfate Production and Ammonium Nitrate Production: The original profiles were based on stoichiometry of $(NH_4)_2SO_4$ and $(NH_4)NO_3$, but did not account for $H_2O$. Therefore, all components were scaled down until the sum of species was equal to 100% of the $PM_{2.5}$ mass.

8. Cast Iron Cupola: The sum of species in the hybridized profile (see Section S3.4) was 109.9% of $PM_{2.5}$. No information about OC artifact correction could be found in the reference of the Heat Treating profile. The Cast Iron Cupola profile is the mean of five source tests, and so is expected to have some uncertainty. Therefore, all components were scaled down until the sum of species was equal to 100% of the $PM_{2.5}$ mass.

9. Lead Production: The sum of species was 108.9% of $PM_{2.5}$, based on a single profile (#293302.5), which has a poor quality rating. All species were thus scaled back equally until the sum of species was equal to 100% of $PM_{2.5}$.

10. Cement Production: The sum of species was 100.2% of $PM_{2.5}$. All species were scaled back since the raw profiles used in this composite were originally normalized by the sum of species rather than the gravimetric $PM_{2.5}$ mass.

11. Sea Salt: The sum of species was 100.82% of $PM_{2.5}$. All species were scaled back since the raw profile seems to be an estimate rather than measurement-based, and the extra 0.82% is

likely to be within the uncertainty of our $H_2O$ estimation formula.

## S3.9 Final Composite Speciation Profiles

**Table S6.** Mass fractions of all species for each source category determined in this work.

| Species | Unpaved Road Dust | Agricultural Soil | Wildfires | Agricultural Burning | Bituminous Combustion | Residential Wood Combustion | HDDV Exhaust | Construction Dust |
|---|---|---|---|---|---|---|---|---|
| OC | 5.46e-02 | 3.08e-02 | 4.62e-01 | 3.88e-01 | 2.63e-02 | 5.28e-01 | 1.76e-01 | 4.62e-02 |
| EC | 9.75e-04 | 2.00e-04 | 9.49e-02 | 1.09e-01 | 1.70e-02 | 5.58e-02 | 7.71e-01 | 0 |
| $NH_4^+$ | 5.25e-04 | 5.50e-04 | 8.79e-03 | 1.80e-02 | 2.82e-02 | 1.50e-03 | | 6.72e-04 |
| $NO_3^-$ | 1.27e-03 | 5.66e-04 | 1.32e-03 | 3.50e-03 | 2.08e-03 | 1.90e-03 | 1.14e-03 | 3.65e-04 |
| $SO_4^{2-}$ | 7.50e-03 | 1.10e-03 | 1.26e-02 | 1.65e-02 | 1.27e-01 | 4.10e-03 | 2.95e-03 | 1.05e-02 |
| NCOM | 2.18e-02 | 1.23e-02 | 3.23e-01 | 2.72e-01 | 1.05e-02 | 3.70e-01 | 4.39e-02 | 1.85e-02 |
| MO | 2.57e-01 | 4.00e-01 | 1.28e-02 | 3.51e-03 | 1.49e-01 | 3.02e-03 | 1.39e-04 | 2.18e-01 |
| $H_2O$ | 1.93e-03 | 3.96e-04 | 0 | 0 | 0 | 0 | 0 | 2.68e-03 |
| Na | 3.85e-04 | 1.40e-03 | 5.73e-03 | 6.55e-03 | 1.29e-03 | 9.40e-04 | | |
| Mg | 6.70e-03 | 7.66e-04 | 3.14e-04 | 8.06e-04 | 4.58e-03 | 1.13e-04 | | |
| Al | 4.45e-02 | 8.94e-02 | 6.08e-04 | 3.00e-04 | 4.16e-02 | 1.10e-04 | | 3.68e-02 |
| Si | 1.44e-01 | 2.44e-01 | 1.82e-03 | 1.50e-04 | 7.98e-02 | 3.40e-04 | | 1.15e-01 |
| P | 5.47e-04 | 1.75e-03 | 5.40e-05 | 0 | 1.48e-03 | 4.00e-05 | | 3.30e-04 |
| S | 2.50e-03 | 1.48e-03 | 4.32e-03 | 6.50e-03 | 5.64e-02 | 1.82e-03 | 2.52e-03 | 3.50e-03 |
| Cl | 1.30e-03 | 1.35e-03 | 4.15e-02 | 9.05e-02 | 7.02e-03 | 2.97e-03 | 2.05e-04 | 4.85e-04 |
| K | 1.59e-02 | 2.05e-02 | 2.94e-02 | 7.04e-02 | 4.19e-03 | 9.67e-03 | 3.80e-05 | 1.49e-02 |
| Ca | 7.01e-02 | 2.45e-02 | 3.86e-03 | 3.30e-04 | 3.47e-02 | 1.00e-04 | 5.83e-04 | 9.10e-02 |
| Ti | 3.46e-03 | 5.20e-03 | 5.15e-05 | 1.00e-05 | 1.96e-03 | 0 | 4.00e-06 | 3.55e-03 |
| V | 2.24e-04 | 3.00e-04 | 3.50e-06 | 0 | 1.04e-04 | 0 | 6.00e-06 | 2.44e-04 |
| Cr | 2.00e-04 | 2.67e-04 | 8.00e-06 | 0 | 6.90e-05 | 0 | 0 | 1.86e-04 |
| Mn | 1.15e-03 | 1.30e-03 | 1.60e-05 | 0 | 1.57e-04 | 0 | 0 | 1.05e-03 |
| Fe | 4.13e-02 | 5.77e-02 | 4.34e-04 | 1.00e-04 | 1.90e-02 | 9.00e-05 | 2.62e-04 | 3.51e-02 |
| Co | 0 | 9.80e-05 | 5.00e-07 | 0 | 5.00e-06 | 0 | 2.00e-06 | 1.06e-04 |
| Ni | 4.40e-05 | 1.00e-04 | 5.50e-06 | 0 | 3.30e-05 | 0 | 2.00e-06 | 7.20e-05 |
| Cu | 1.10e-04 | 1.00e-04 | 1.60e-05 | 0 | 1.15e-04 | 0 | 0 | 9.45e-05 |
| Zn | 7.18e-04 | 2.00e-04 | 1.67e-04 | 1.00e-04 | 3.39e-04 | 2.20e-04 | 6.82e-04 | 2.30e-04 |
| Ga | 0 | 0 | 5.00e-07 | 0 | 0 | 0 | 0 | 5.00e-06 |
| As | 1.00e-05 | 0 | 1.23e-04 | 0 | 0 | 2.00e-06 | 3.00e-06 | 4.80e-05 |
| Se | 0 | 0 | 6.00e-06 | 0 | 3.30e-03 | 0 | 0 | 0 |
| Br | 1.65e-05 | 0 | 4.20e-04 | 2.80e-04 | 3.29e-04 | 2.00e-05 | 1.50e-05 | 9.00e-06 |
| Rb | 1.00e-04 | 1.41e-04 | 1.90e-05 | 1.00e-04 | 3.00e-05 | 6.00e-06 | 1.00e-06 | 5.05e-05 |
| Sr | 3.00e-04 | 3.00e-04 | 3.85e-05 | 0 | 1.13e-03 | 0 | 1.00e-06 | 4.61e-04 |
| Zr | 1.36e-04 | 1.00e-04 | 6.00e-06 | 0 | 7.40e-05 | 0 | 0 | 1.56e-04 |
| Mo | 9.50e-06 | 0 | 5.00e-07 | 0 | 0 | 0 | 2.00e-06 | 6.00e-06 |
| Pd | 3.10e-05 | 0 | 5.00e-07 | 5.15e-05 | 5.80e-05 | 1.00e-06 | 2.60e-05 | 1.10e-05 |
| Ag | 0 | 0 | 2.50e-06 | 1.55e-05 | 8.90e-05 | 7.00e-06 | 6.10e-05 | 5.00e-06 |
| Cd | 2.35e-05 | 0 | 5.50e-06 | 0 | 2.70e-05 | 7.00e-06 | 4.40e-05 | 2.00e-05 |
| In | 0 | 0 | 1.45e-05 | 5.00e-05 | 4.00e-05 | 0 | 1.80e-05 | 5.00e-06 |
| Sn | 0 | 0 | 1.45e-05 | 0 | 1.50e-04 | 1.00e-05 | 2.50e-05 | 0 |
| Sb | 2.25e-05 | 0 | 7.00e-06 | 0 | 0 | 0 | 1.90e-05 | 2.03e-04 |
| Ba | 7.00e-04 | 8.00e-04 | 2.25e-05 | 2.50e-04 | 2.24e-03 | 0 | 3.50e-04 | 4.76e-04 |
| La | 1.65e-04 | 1.00e-04 | 2.00e-06 | 0 | 2.00e-05 | 1.00e-05 | 2.72e-04 | 0 |
| Ce | | | | | | | | |
| Hg | 0 | 0 | 2.50e-06 | 0 | 1.00e-05 | 0 | 0 | 1.80e-05 |
| Pb | 2.88e-04 | 9.55e-05 | 1.60e-05 | 1.00e-05 | 4.00e-05 | 6.00e-06 | 1.00e-06 | 1.38e-04 |
| PMO | 3.22e-01 | 1.03e-01 | 0 | 1.96e-02 | 4.36e-01 | 2.11e-02 | 2.42e-03 | 4.02e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Paved Road Dust | Prescribed Burning | Sub-Bituminous Combustion | Sand&Gravel | Natural Gas Combustion | Nonroad Gasoline Exhaust | Wood Fired Boiler | Distillate Oil Combustion |
|---|---|---|---|---|---|---|---|---|
| OC | 9.74e-02 | 5.02e-01 | 3.16e-02 | 0 | 2.47e-01 | 4.75e-01 | 3.51e-01 | 2.50e-01 |
| EC | 1.04e-02 | 1.09e-01 | 4.28e-02 | 0 | 3.84e-01 | 1.22e-01 | 3.71e-02 | 1.00e-01 |
| $NH_4^+$ | 6.88e-04 | 3.41e-03 | 3.48e-03 | 0 | | | | |
| $NO_3^-$ | 4.00e-04 | 1.07e-02 | 5.71e-04 | 5.15e-04 | 2.10e-02 | 7.00e-04 | 0 | 0 |
| $SO_4^{2-}$ | 6.46e-03 | 3.30e-03 | 1.02e-01 | 2.87e-03 | 8.60e-02 | 5.00e-04 | 6.53e-02 | 1.90e-01 |
| NCOM | 3.90e-02 | 3.51e-01 | 1.27e-02 | 0 | 9.88e-02 | 1.19e-01 | 1.40e-01 | 1.00e-01 |
| MO | 2.87e-01 | 4.20e-03 | 1.81e-01 | 3.18e-01 | 0 | 1.95e-03 | 1.66e-01 | 0 |
| $H_2O$ | 1.72e-03 | 0 | 0 | 6.89e-04 | 0 | 0 | 0 | 0 |
| Na | 2.55e-03 | 1.35e-03 | | 5.00e-03 | | | 1.51e-03 | |
| Mg | 6.92e-03 | | | 4.14e-03 | | | 1.42e-03 | |
| Al | 5.36e-02 | 4.60e-04 | 5.97e-02 | 8.09e-02 | | | | |
| Si | 1.69e-01 | 1.00e-04 | 9.01e-02 | 1.91e-01 | | 1.20e-03 | 1.36e-01 | 8.00e-03 |
| P | 8.12e-04 | 6.00e-04 | 9.37e-03 | 3.67e-04 | | 3.00e-04 | 7.53e-04 | |
| S | 5.13e-03 | 1.40e-03 | 2.95e-02 | 1.80e-03 | 2.87e-02 | 1.67e-04 | 3.10e-02 | 2.74e-02 |
| Cl | 2.18e-03 | 2.39e-03 | 6.29e-04 | 2.70e-03 | | | 5.35e-03 | |
| K | 1.64e-02 | 1.35e-03 | 4.64e-03 | 1.41e-02 | | | 8.87e-02 | 4.00e-04 |
| Ca | 4.65e-02 | 7.20e-04 | 3.45e-02 | 1.90e-02 | | 3.00e-04 | 6.36e-03 | 2.00e-04 |
| Ti | 3.66e-03 | 6.70e-04 | 4.32e-03 | 2.40e-03 | | | | |
| V | 2.16e-04 | 1.00e-05 | 0 | 2.85e-04 | | | | |
| Cr | 2.52e-04 | 2.00e-05 | 1.76e-04 | 2.00e-04 | | | | 2.50e-04 |
| Mn | 9.20e-04 | 1.10e-04 | 2.83e-04 | 1.30e-03 | | | | |
| Fe | 4.18e-02 | 4.45e-04 | 2.92e-02 | 2.96e-02 | | 1.00e-04 | | 9.00e-04 |
| Co | 0 | | 0 | 5.00e-05 | | | | |
| Ni | 8.25e-05 | 2.00e-05 | 7.20e-05 | 6.20e-05 | | | | |
| Cu | 2.28e-04 | 2.00e-05 | 1.78e-04 | 7.00e-05 | | | | 2.00e-04 |
| Zn | 1.27e-03 | 2.28e-03 | 7.97e-04 | 2.30e-04 | | 3.00e-04 | | 1.55e-03 |
| Ga | 5.00e-07 | | 1.33e-04 | 0 | | | | |
| As | 4.50e-06 | | 0 | 1.00e-04 | | | | |
| Se | 1.00e-06 | | 4.06e-04 | 0 | | | | |
| Br | 2.47e-05 | 9.00e-05 | 1.47e-04 | 0 | | | | |
| Rb | 1.02e-04 | | 2.75e-05 | 6.00e-05 | | | | |
| Sr | 3.55e-04 | | 1.96e-03 | 2.15e-04 | | | | |
| Zr | 1.30e-04 | | 2.48e-04 | 1.45e-04 | | | | |
| Mo | 3.50e-06 | | 5.90e-05 | 0 | | | | |
| Pd | 4.00e-06 | | 0 | 5.00e-05 | | | | |
| Ag | 1.00e-05 | 1.90e-04 | 0 | 2.85e-05 | | | | |
| Cd | 7.50e-06 | 4.86e-03 | 0 | 0 | | 1.00e-04 | | |
| In | 0 | | 0 | 1.00e-04 | | 1.00e-04 | | |
| Sn | 3.80e-05 | 1.80e-04 | 0 | 3.89e-04 | | | | |
| Sb | 0 | | 0 | 0 | | 2.00e-04 | | |
| Ba | 8.50e-04 | | 1.33e-02 | 1.62e-03 | | | | |
| La | 1.25e-04 | | 0 | 0 | | | | |
| Ce | | | | | | | | |
| Hg | 2.00e-06 | | 0 | 0 | | | | |
| Pb | 6.67e-04 | 1.00e-04 | 6.81e-04 | 3.80e-05 | | 3.00e-04 | | |
| PMO | 2.08e-01 | 0 | 3.75e-01 | 3.24e-01 | 1.63e-01 | 2.78e-01 | 0 | 3.49e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Charbroiling | Residual Oil Combustion | Dairy Soil | Kraft Recovery Furnace | Mineral Products-Avg | Industrial Manufacturing-Avg | Onroad Gasoline Exhaust | Heat Treating |
|---|---|---|---|---|---|---|---|---|
| OC | 6.70e-01 | 1.00e-02 | 3.18e-01 | 5.23e-02 | 5.25e-02 | 7.36e-02 | 5.49e-01 | 7.00e-02 |
| EC | 4.06e-02 | 1.00e-02 | 5.16e-02 | 1.53e-02 | 1.47e-02 | 8.94e-03 | 1.90e-01 | 1.00e-02 |
| $NH_4^+$ | 0 | | 3.33e-02 | 1.06e-02 | | | 1.67e-02 | |
| $NO_3^-$ | 1.45e-03 | 0 | 9.26e-02 | 3.51e-03 | 2.71e-03 | 2.97e-03 | 1.51e-03 | 5.50e-03 |
| $SO_4^{2-}$ | 1.35e-03 | 4.40e-01 | 4.46e-02 | 5.48e-01 | 1.41e-01 | 9.86e-02 | 8.08e-03 | 3.20e-01 |
| NCOM | 2.68e-01 | 4.00e-03 | 1.27e-01 | 2.09e-02 | 2.10e-02 | 2.94e-02 | 1.37e-01 | 2.80e-02 |
| MO | 4.52e-03 | 0 | 8.52e-02 | 1.56e-02 | 0 | 0 | 1.30e-02 | 0 |
| $H_2O$ | 0 | 0 | 1.87e-02 | 0 | 3.37e-02 | 2.37e-02 | 0 | 0 |
| Na | 3.00e-03 | | 5.00e-03 | 2.58e-01 | | | 1.08e-03 | |
| Mg | 9.56e-04 | | | 1.80e-04 | | | 5.29e-04 | |
| Al | 5.21e-04 | | 1.29e-02 | 9.20e-04 | | | 1.47e-03 | |
| Si | 9.60e-04 | | 3.99e-02 | 3.03e-03 | | | 4.75e-03 | |
| P | 4.78e-04 | | 8.10e-03 | | | | 1.73e-03 | |
| S | 1.66e-03 | 1.26e-01 | 2.61e-02 | 1.30e-01 | 4.69e-02 | 3.29e-02 | 5.38e-03 | 1.07e-01 |
| Cl | 3.70e-03 | 3.13e-03 | 3.90e-03 | 3.06e-02 | | | 7.96e-04 | 3.00e-01 |
| K | 2.47e-03 | | 2.05e-02 | 2.49e-02 | | | 1.81e-04 | 5.50e-03 |
| Ca | 3.35e-04 | 3.51e-04 | 2.14e-02 | 6.21e-03 | | | 2.88e-03 | 5.50e-03 |
| Ti | 3.35e-05 | 6.43e-04 | 5.00e-04 | 4.00e-05 | | | 5.54e-05 | |
| V | 2.00e-06 | 8.19e-04 | 0 | 3.00e-05 | | | 1.99e-05 | |
| Cr | 0 | | 1.00e-04 | 3.00e-05 | | | 4.84e-05 | 5.50e-03 |
| Mn | 5.00e-07 | 3.70e-05 | 4.00e-04 | 3.00e-05 | | | 4.03e-05 | |
| Fe | 4.57e-04 | 2.93e-04 | 1.19e-02 | 6.70e-04 | | | 4.04e-03 | 9.00e-02 |
| Co | 8.00e-06 | | 0 | 1.00e-05 | | | 7.52e-06 | |
| Ni | 6.25e-05 | 4.83e-04 | 0 | 1.00e-05 | | | 6.40e-05 | 2.00e-02 |
| Cu | 1.43e-04 | | 1.00e-04 | 6.00e-05 | | | 2.10e-04 | 5.00e-04 |
| Zn | 3.08e-04 | 7.38e-05 | 3.00e-04 | 1.40e-04 | | | 2.65e-03 | 5.00e-04 |
| Ga | 0 | | 0 | 0 | | | 8.72e-06 | |
| As | 3.50e-06 | | 1.00e-04 | 1.00e-05 | | | 5.21e-06 | |
| Se | 5.00e-07 | 5.02e-05 | 1.00e-04 | | | | 6.09e-06 | |
| Br | 3.85e-05 | | 2.00e-04 | 7.20e-04 | | | 5.90e-05 | 5.00e-04 |
| Rb | 1.50e-06 | | 0 | 6.00e-05 | | | 1.81e-06 | |
| Sr | 5.50e-06 | | 2.00e-04 | 3.00e-05 | | | 1.09e-05 | |
| Zr | 0 | | 0 | | | | 9.00e-06 | |
| Mo | 0 | | 0 | 0 | | | 2.30e-05 | 5.50e-03 |
| Pd | 0 | | 1.00e-04 | | | | 5.90e-05 | |
| Ag | 4.40e-05 | | 1.00e-04 | | | | 5.80e-05 | |
| Cd | 2.00e-06 | | 1.00e-04 | 4.00e-05 | | | 7.25e-05 | 5.00e-04 |
| In | 2.00e-06 | | 0 | | | | 3.50e-05 | |
| Sn | 1.40e-05 | | 0 | 0 | | | 1.32e-04 | |
| Sb | 2.70e-05 | 2.35e-04 | 0 | 2.00e-05 | | | 1.89e-04 | |
| Ba | 2.10e-04 | | 2.00e-03 | 0 | | | 5.87e-04 | 5.50e-03 |
| La | 1.00e-04 | 5.04e-04 | 1.00e-04 | | | | 3.34e-04 | |
| Ce | | | | | | | | |
| Hg | 1.00e-06 | | 0 | 1.00e-05 | | | 1.15e-05 | |
| Pb | 6.60e-05 | | 0 | 0 | | | 4.75e-04 | 5.00e-04 |
| PMO | 0 | 5.29e-01 | 1.00e-01 | 7.22e-03 | 7.35e-01 | 7.63e-01 | 6.15e-02 | 1.27e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Chemical Manufacturing-Avg | Lignite Combustion | Solid Waste Combustion | Cement Production | Wood Products-Drying | Surface Coating | Food&Ag-Handling | Wood Products-Sawing |
|---|---|---|---|---|---|---|---|---|
| OC | 9.17e-02 | 2.84e-01 | 8.41e-02 | 1.27e-01 | 5.00e-01 | 2.08e-01 | 4.05e-02 | 4.45e-01 |
| EC | 1.83e-02 | 1.43e-02 | 1.52e-02 | 2.93e-02 | 8.00e-02 | 7.00e-03 | 1.76e-03 | 3.80e-02 |
| $NH_4^+$ | | 4.07e-03 | 8.22e-02 | 2.34e-02 | | | 1.13e-03 | |
| $NO_3^-$ | 3.50e-03 | 3.89e-03 | 1.42e-02 | 4.69e-02 | 1.25e-02 | | 0 | 5.50e-03 |
| $SO_4^{2-}$ | 3.13e-02 | 7.56e-02 | 6.80e-02 | 1.77e-01 | 3.65e-02 | 0 | 3.56e-03 | 5.50e-03 |
| NCOM | 3.67e-02 | 1.14e-01 | 3.36e-02 | 5.07e-02 | 2.00e-01 | 8.30e-02 | 1.62e-02 | 1.78e-01 |
| MO | 0 | 1.83e-01 | 1.28e-01 | 1.35e-01 | 2.09e-02 | 2.72e-01 | 1.09e-01 | 9.00e-03 |
| $H_2O$ | 7.50e-03 | 0 | 0 | 4.80e-02 | 8.76e-03 | 0 | 1.12e-03 | 1.32e-03 |
| Na | | 1.95e-03 | 8.09e-03 | 2.29e-02 | 3.70e-02 | | 6.14e-04 | 2.40e-03 |
| Mg | | 9.94e-03 | | 4.71e-04 | 0 | | 2.59e-03 | |
| Al | | 3.79e-02 | 1.99e-02 | 1.05e-02 | | 3.70e-02 | 2.69e-03 | 1.80e-03 |
| Si | | 6.39e-02 | 6.87e-02 | 4.23e-02 | 2.50e-03 | 3.81e-02 | 8.14e-02 | 3.00e-03 |
| P | | 2.85e-03 | 3.35e-03 | 0 | | 0 | 5.38e-03 | |
| S | 1.04e-02 | 3.31e-02 | 2.90e-02 | 6.56e-02 | 1.40e-02 | 0 | 3.59e-03 | 1.83e-03 |
| Cl | | 9.04e-04 | 1.38e-01 | 3.13e-02 | 4.50e-02 | 2.45e-03 | 9.27e-04 | 3.00e-04 |
| K | | 4.27e-03 | 2.16e-02 | 6.94e-02 | 4.00e-02 | 2.95e-03 | 8.79e-03 | 3.00e-04 |
| Ca | | 1.45e-01 | 2.19e-02 | 1.67e-01 | 5.00e-03 | 3.79e-02 | 9.01e-03 | 2.40e-03 |
| Ti | | 6.86e-03 | 2.40e-03 | 3.47e-03 | | 2.55e-01 | 0 | 5.00e-04 |
| V | | 4.20e-04 | 1.05e-04 | 1.63e-04 | | 0 | 0 | |
| Cr | | 1.27e-04 | 3.00e-04 | 6.40e-05 | | 0 | 0 | 5.00e-04 |
| Mn | | 2.66e-04 | 6.00e-04 | 9.64e-04 | 9.00e-04 | 0 | 1.97e-04 | 3.00e-04 |
| Fe | | 2.77e-02 | 2.38e-02 | 6.65e-03 | 1.00e-03 | 5.00e-04 | 1.76e-03 | 3.80e-03 |
| Co | | 1.34e-05 | 0 | 2.88e-05 | | 1.00e-04 | 8.80e-05 | |
| Ni | | 1.29e-04 | 1.80e-04 | 2.41e-04 | | 0 | 1.10e-05 | 5.00e-04 |
| Cu | | 6.69e-04 | 5.00e-04 | 2.40e-04 | | 0 | 2.66e-04 | 5.00e-04 |
| Zn | | 2.96e-03 | 4.96e-03 | 4.06e-03 | 6.00e-04 | 0 | 1.38e-04 | 5.00e-04 |
| Ga | | 1.04e-04 | 7.50e-05 | 1.98e-06 | | 0 | 0 | |
| As | | 2.14e-05 | 7.00e-05 | 8.93e-06 | | 0 | 0 | |
| Se | | 9.27e-05 | 1.00e-05 | 6.05e-05 | | 0 | 0 | |
| Br | | 5.43e-06 | 2.00e-04 | 1.10e-03 | | 1.50e-04 | 1.21e-04 | 5.50e-03 |
| Rb | | 2.10e-05 | 1.00e-04 | 7.74e-04 | | 1.00e-04 | 0 | |
| Sr | | 3.70e-03 | 2.00e-04 | 8.50e-04 | | 5.00e-05 | 5.30e-05 | |
| Zr | | 1.88e-04 | 1.95e-04 | 1.21e-04 | | 6.50e-04 | 0 | |
| Mo | | 6.01e-05 | 1.90e-04 | 3.97e-05 | | 5.00e-05 | 2.00e-06 | 5.00e-04 |
| Pd | | 2.93e-05 | 1.00e-04 | 1.98e-06 | | 0 | 1.18e-04 | |
| Ag | | 3.08e-05 | 1.35e-04 | 4.36e-05 | | 0 | 1.43e-04 | |
| Cd | | 2.39e-05 | 1.00e-04 | 5.36e-05 | | 7.00e-04 | 2.50e-05 | |
| In | | 6.81e-05 | 5.50e-05 | 8.23e-05 | | 0 | 0 | |
| Sn | | 8.55e-05 | 1.00e-04 | 5.36e-05 | | 0 | 8.70e-05 | 5.00e-04 |
| Sb | | 8.55e-05 | 7.00e-05 | 3.77e-05 | | 7.10e-03 | 1.53e-04 | |
| Ba | | 1.07e-02 | 8.30e-04 | 3.92e-04 | | 1.89e-02 | 1.02e-03 | 5.00e-04 |
| La | | 1.45e-06 | 9.00e-05 | 1.09e-05 | | 2.82e-02 | 1.59e-03 | |
| Ce | | | 7.50e-05 | | | | | |
| Hg | | 1.96e-05 | | 1.79e-05 | | 0 | 0 | |
| Pb | | 1.78e-04 | | 6.30e-04 | | 4.50e-04 | 1.19e-04 | 5.00e-04 |
| PMO | 8.11e-01 | 0 | 2.57e-01 | 0 | 9.35e-03 | 0 | 7.09e-01 | 2.93e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Aluminum Processing | Open Hearth Furnace | Brake Lining Dust | Meat Frying | Process Gas Combustion | Aluminum Production | Lime Kiln | Sintering Furnace |
|---|---|---|---|---|---|---|---|---|
| OC | 1.50e-02 | 2.00e-01 | 1.07e-01 | 5.74e-01 | 3.01e-01 | 3.90e-02 | 6.65e-02 | 2.73e-02 |
| EC | 1.90e-03 | | 2.61e-02 | 0 | 1.46e-01 | 2.30e-02 | 2.32e-02 | 1.72e-03 |
| $NH_4^+$ | | | 3.00e-05 | 0 | 7.60e-02 | | 0 | |
| $NO_3^-$ | 1.38e-02 | 5.50e-03 | 1.60e-03 | 2.08e-02 | 2.37e-02 | 4.10e-03 | 5.56e-03 | |
| $SO_4^{2-}$ | 8.65e-02 | 4.00e-01 | 3.34e-02 | 9.10e-03 | 1.66e-01 | 4.40e-02 | 3.69e-01 | 1.03e-01 |
| NCOM | 6.00e-03 | 8.00e-02 | 4.28e-02 | 2.30e-01 | 1.21e-01 | 1.56e-02 | 2.66e-02 | 1.09e-02 |
| MO | 1.38e-01 | 3.24e-03 | 2.28e-01 | 4.57e-03 | 5.67e-02 | 2.74e-01 | 8.49e-02 | 1.25e-01 |
| $H_2O$ | 2.08e-02 | 0 | 8.02e-03 | 0 | 0 | 1.06e-02 | 0 | 0 |
| Na | 1.55e-01 | | 1.60e-04 | 4.50e-03 | 6.47e-04 | 4.10e-02 | 2.70e-01 | 1.30e-01 |
| Mg | | | 1.11e-01 | 0 | 1.10e-03 | 2.80e-02 | 5.00e-03 | 1.60e-03 |
| Al | 1.70e-02 | | 1.24e-03 | 0 | 1.42e-02 | 2.70e-01 | 3.10e-03 | 9.00e-04 |
| Si | 4.76e-02 | | 8.80e-02 | 0 | 1.86e-02 | 3.40e-03 | 2.90e-03 | 1.24e-02 |
| P | 4.12e-03 | | 0 | 0 | 6.88e-05 | | 2.31e-03 | |
| S | 3.00e-02 | 1.33e-01 | 1.11e-02 | 3.03e-03 | 5.09e-02 | 1.40e-02 | 1.23e-01 | 3.42e-02 |
| Cl | 2.00e-01 | | 1.48e-03 | 3.52e-02 | 2.17e-03 | 1.33e-02 | 1.53e-02 | 1.70e-01 |
| K | 1.00e-01 | 3.00e-02 | 1.90e-04 | 3.60e-03 | 1.28e-03 | 2.20e-03 | 1.56e-02 | 2.10e-01 |
| Ca | 1.20e-03 | 5.50e-03 | 1.00e-02 | 1.50e-03 | 1.32e-02 | 3.30e-03 | 8.85e-02 | 5.83e-03 |
| Ti | 2.50e-04 | | 3.60e-03 | 0 | 0 | 4.00e-04 | 1.70e-04 | |
| V | 5.00e-05 | 5.50e-03 | 3.35e-04 | 0 | 0 | 6.40e-04 | 6.00e-05 | |
| Cr | 3.90e-04 | 2.00e-02 | 1.09e-03 | 1.50e-03 | 2.21e-05 | 0 | 9.50e-05 | |
| Mn | 1.20e-04 | 5.50e-03 | 1.07e-03 | 4.10e-04 | 2.42e-04 | 1.10e-04 | 1.00e-03 | 2.83e-03 |
| Fe | 2.90e-03 | 1.00e-01 | 1.15e-01 | 2.40e-03 | 1.65e-02 | 4.50e-03 | 1.20e-03 | 6.94e-02 |
| Co | 2.55e-04 | | 7.00e-05 | | 1.95e-05 | | | |
| Ni | 3.30e-04 | 5.50e-03 | 6.60e-04 | 4.90e-04 | 5.36e-04 | 1.90e-03 | 1.80e-04 | |
| Cu | 4.50e-04 | 5.50e-03 | 1.15e-02 | 0 | 1.03e-02 | 4.40e-04 | 6.00e-05 | 2.00e-02 |
| Zn | 7.50e-04 | | 1.69e-03 | 0 | 3.00e-02 | 1.50e-04 | 7.00e-05 | 2.95e-03 |
| Ga | | | | | 0 | | 1.00e-05 | |
| As | 6.50e-05 | 5.00e-04 | 1.00e-05 | 0 | 4.58e-05 | | 1.00e-04 | 2.77e-03 |
| Se | 1.10e-04 | | 2.00e-05 | 6.00e-05 | 1.02e-05 | | 2.50e-05 | 5.00e-04 |
| Br | 6.40e-04 | 5.00e-04 | 4.00e-05 | 8.40e-04 | 6.71e-05 | 3.70e-04 | 1.20e-04 | 5.50e-03 |
| Rb | 6.50e-05 | 5.00e-04 | 5.00e-05 | 9.00e-04 | 1.70e-05 | | 9.50e-05 | 5.50e-03 |
| Sr | 5.00e-05 | | 5.75e-04 | 1.00e-04 | 1.53e-05 | | 7.00e-05 | 5.00e-04 |
| Zr | 2.00e-05 | | | | 0 | | 9.00e-05 | 5.00e-04 |
| Mo | | | 3.70e-03 | | 1.70e-06 | | 1.00e-04 | |
| Pd | | | | | 0 | | 9.00e-05 | |
| Ag | 1.00e-04 | 5.00e-04 | | | 0 | | 1.85e-04 | 5.00e-04 |
| Cd | 0 | 5.00e-04 | | | 8.05e-04 | | 8.00e-05 | |
| In | | | | | 0 | | 6.00e-05 | |
| Sn | 5.00e-04 | 5.00e-04 | 6.60e-03 | | 8.49e-06 | | 3.20e-04 | |
| Sb | 5.84e-03 | 5.00e-04 | | | 5.09e-06 | | 1.10e-04 | |
| Ba | 3.60e-04 | | 3.45e-02 | 4.60e-03 | 1.42e-04 | | 3.20e-04 | |
| La | 2.00e-05 | | | | 3.66e-04 | | 9.90e-04 | |
| Ce | 0 | | 1.00e-05 | | | | 1.18e-03 | 5.50e-03 |
| Hg | | | | | 0 | | 4.00e-05 | |
| Pb | 7.90e-04 | 5.50e-03 | 5.00e-05 | 2.00e-03 | 2.63e-05 | 1.20e-04 | 3.10e-04 | 1.75e-03 |
| PMO | 1.79e-01 | 1.25e-01 | 1.61e-01 | 1.04e-01 | 0 | 2.20e-01 | 1.34e-02 | 8.29e-02 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Charcoal Manufacturing | Catalytic Cracking | Fiberglass Manufacturing | Glass Furnace | Pulp&Paper Mills | Petroleum Industry-Avg | Slash Burning | Misc. Sources |
|---|---|---|---|---|---|---|---|---|
| OC | 1.80e-02 | 1.37e-04 | 2.80e-01 | 7.10e-03 | 0 | 3.50e-02 | 5.22e-01 | 8.77e-02 |
| EC | 5.20e-02 | 7.03e-04 | 2.00e-02 | 6.20e-04 | 1.00e-03 | | 4.35e-02 | 4.41e-02 |
| $NH_4^+$ | | 3.28e-03 | | | | | | |
| $NO_3^-$ | 3.00e-03 | 0 | 5.50e-03 | 1.50e-04 | | 2.75e-03 | 1.32e-03 | 1.64e-03 |
| $SO_4^{2-}$ | 6.50e-02 | 3.27e-01 | 5.50e-03 | 4.89e-01 | 5.06e-01 | 2.35e-01 | 1.27e-02 | 5.51e-02 |
| NCOM | 7.20e-03 | 5.48e-05 | 1.12e-01 | 2.84e-03 | 0 | 1.40e-02 | 3.65e-01 | 3.51e-02 |
| MO | 3.18e-01 | 2.59e-01 | 3.54e-03 | 0 | 2.51e-02 | 0 | 1.17e-02 | 0 |
| $H_2O$ | 0 | 7.92e-02 | 1.32e-03 | 0 | 0 | 5.64e-02 | 0 | 1.32e-02 |
| Na | | 1.80e-05 | | 1.28e-01 | 2.93e-01 | | 1.73e-02 | |
| Mg | 1.90e-02 | 0 | | 1.83e-03 | 2.90e-03 | | | |
| Al | 7.00e-02 | 8.08e-02 | | 1.10e-04 | | | 1.03e-03 | |
| Si | 1.50e-01 | 1.98e-01 | | 3.20e-03 | | | 1.60e-03 | |
| P | | 3.45e-04 | | 2.50e-04 | | | 9.05e-04 | |
| S | 3.10e-02 | 1.43e-01 | 1.83e-03 | 1.76e-01 | 1.74e-01 | 7.83e-02 | 4.22e-03 | 1.84e-02 |
| Cl | 5.00e-02 | 9.30e-05 | 1.50e-01 | 1.60e-04 | 2.02e-02 | | 5.11e-03 | |
| K | 7.40e-02 | 8.00e-04 | 5.50e-03 | 1.80e-02 | 2.62e-02 | | 1.31e-02 | |
| Ca | 9.50e-02 | 1.34e-03 | 5.00e-04 | 4.27e-03 | | | 1.54e-03 | |
| Ti | 3.60e-03 | 6.67e-03 | | 4.00e-05 | | | 1.15e-04 | |
| V | 2.80e-04 | 2.62e-03 | | 6.00e-05 | | | 4.00e-05 | |
| Cr | 4.40e-04 | 4.21e-04 | 5.00e-04 | 2.15e-03 | | | 1.05e-04 | |
| Mn | 4.40e-03 | 1.90e-04 | | 3.00e-05 | | | 1.70e-04 | |
| Fe | 6.40e-02 | 1.16e-02 | 5.50e-03 | 2.60e-04 | | | 4.35e-04 | |
| Co | | 7.60e-05 | 5.00e-04 | 2.00e-04 | | | | |
| Ni | | 1.47e-03 | 5.00e-04 | 4.00e-05 | | | 7.00e-05 | |
| Cu | 5.40e-04 | 2.30e-04 | 5.00e-04 | 1.00e-05 | | | 5.00e-05 | |
| Zn | 3.60e-03 | 2.41e-04 | 5.00e-04 | 2.20e-04 | | | 4.70e-04 | |
| Ga | | 1.40e-05 | | | | | | |
| As | | 0 | | 2.30e-04 | | | | |
| Se | | 1.20e-05 | 5.00e-04 | 1.40e-04 | | | | |
| Br | 9.00e-04 | 1.20e-05 | 5.00e-04 | 4.00e-05 | | | 2.90e-04 | |
| Rb | | 3.00e-06 | | 1.30e-04 | | | | |
| Sr | | 8.30e-05 | | 3.00e-05 | | | | |
| Zr | | 1.18e-04 | | | | | | |
| Mo | | 7.90e-05 | | 4.20e-04 | | | | |
| Pd | | 0 | | | | | | |
| Ag | | 0 | | 1.00e-05 | | | 2.70e-04 | |
| Cd | | 0 | | 3.00e-05 | | | 3.00e-04 | |
| In | | 0 | | | | | | |
| Sn | | 5.50e-06 | | 1.50e-04 | | | 3.10e-04 | |
| Sb | | 6.68e-04 | | 2.00e-05 | | | | |
| Ba | | 1.47e-04 | | 3.00e-05 | | | | |
| La | | 5.39e-03 | | | | | | |
| Ce | | | | | | | | |
| Hg | | 0 | | | | | | |
| Pb | 9.00e-04 | 9.40e-05 | 5.00e-04 | 1.95e-03 | | | 7.30e-04 | |
| PMO | 4.77e-04 | 1.95e-02 | 4.07e-01 | 3.38e-01 | 1.26e-01 | 6.57e-01 | 0 | 7.63e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Asphalt Roofing | Inorganic Chemical Manufacturing | Tire Dust | Ferromanganese Furnace | Wood Products-Sanding | Electric Arc Furnace | Food&Ag-Drying | Residential Coal Combustion |
|---|---|---|---|---|---|---|---|---|
| OC | 6.03e-01 | 0 | 4.72e-01 | 5.14e-02 | 3.50e-01 | 3.23e-02 | 1.20e-01 | 4.48e-01 |
| EC | 1.30e-04 | 0 | 2.20e-01 | 1.01e-01 | 6.00e-02 | 3.63e-03 | | 2.40e-01 |
| $NH_4^+$ | 3.40e-03 | | 1.90e-04 | | | | | 1.41e-02 |
| $NO_3^-$ | 1.20e-04 | 3.50e-03 | 1.50e-03 | 5.70e-02 | | 2.30e-03 | 5.00e-04 | 3.15e-03 |
| $SO_4^{2-}$ | 9.10e-04 | 3.13e-02 | 3.11e-02 | 4.02e-02 | | 2.24e-02 | 5.00e-04 | 3.32e-02 |
| NCOM | 2.41e-01 | 0 | 1.89e-01 | 2.06e-02 | 1.40e-01 | 1.29e-02 | 4.80e-02 | 1.79e-01 |
| MO | 4.33e-04 | 0 | 2.50e-03 | 1.23e-01 | 4.70e-04 | 2.46e-01 | 8.88e-02 | 2.73e-02 |
| $H_2O$ | 1.03e-03 | 7.50e-03 | 7.51e-03 | 0 | | 0 | 1.20e-04 | 0 |
| Na | 1.10e-04 | | 6.10e-04 | 2.57e-02 | | 3.90e-02 | | 2.01e-04 |
| Mg | 0 | | 3.75e-04 | 2.27e-02 | | 3.11e-02 | | 2.90e-03 |
| Al | 1.80e-04 | | 6.05e-04 | 1.43e-02 | | 6.50e-03 | | 6.70e-03 |
| Si | 1.70e-04 | | 1.15e-03 | 5.49e-02 | | 2.99e-02 | 5.00e-02 | 6.86e-03 |
| P | 0 | | 1.25e-03 | 1.20e-03 | | 9.50e-04 | | 5.39e-04 |
| S | 3.03e-04 | 1.04e-02 | 1.04e-02 | 1.32e-02 | | 1.88e-02 | 1.67e-04 | 1.50e-02 |
| Cl | 7.00e-03 | | 7.80e-03 | 4.20e-03 | | 1.85e-02 | 5.50e-03 | 9.05e-04 |
| K | 1.00e-05 | | 3.80e-04 | 7.18e-02 | | 1.86e-02 | 5.50e-03 | 5.33e-03 |
| Ca | 0 | | 1.12e-03 | 1.20e-02 | 5.00e-04 | 6.20e-02 | 1.00e-02 | 1.16e-02 |
| Ti | 0 | | 3.60e-04 | 4.60e-04 | | 2.00e-03 | | 4.00e-04 |
| V | 0 | | 0 | 1.70e-04 | | 5.00e-04 | | 1.50e-04 |
| Cr | 0 | | 3.00e-05 | 4.20e-04 | | | 5.50e-03 | 3.50e-05 |
| Mn | 0 | | 1.00e-04 | | | 3.65e-02 | 5.50e-03 | 5.65e-05 |
| Fe | 1.00e-05 | | 4.60e-03 | 2.08e-02 | 5.00e-04 | 3.20e-01 | 5.00e-02 | 8.59e-03 |
| Co | | | | | | | | 3.50e-05 |
| Ni | 0 | | 5.00e-05 | | | 7.00e-03 | 5.50e-03 | 4.51e-04 |
| Cu | 7.00e-05 | | 4.90e-04 | 3.60e-04 | | 2.80e-03 | 5.00e-04 | 3.06e-04 |
| Zn | 5.00e-05 | | 5.31e-03 | 4.70e-03 | | 1.20e-02 | | 1.58e-03 |
| Ga | | | | | | | | 6.00e-06 |
| As | 0 | | 0 | 2.50e-04 | | 1.60e-04 | | 1.53e-04 |
| Se | 0 | | 2.00e-05 | | | 2.90e-04 | | 4.10e-05 |
| Br | 0 | | 1.50e-05 | 1.05e-03 | | 9.10e-03 | 5.00e-04 | 4.50e-05 |
| Rb | 0 | | 0 | | | | | 6.00e-06 |
| Sr | 0 | | 7.00e-05 | | 5.00e-04 | | | 1.93e-04 |
| Zr | | | | | | | | 2.00e-06 |
| Mo | | | | | | 3.90e-04 | | 0 |
| Pd | | | | | | 0 | | 1.90e-05 |
| Ag | | | | | | | | 4.50e-03 |
| Cd | | | | | | | | 9.00e-06 |
| In | | | | | | 2.90e-04 | | 1.80e-05 |
| Sn | | | | | | | | 5.71e-04 |
| Sb | | | | | | | | 2.04e-03 |
| Ba | 4.00e-05 | | 1.95e-04 | | | | | 5.81e-04 |
| La | | | | | | | | 1.90e-05 |
| Ce | | | | | | | | |
| Hg | | | | | | | | 3.00e-06 |
| Pb | 1.00e-05 | | 1.60e-04 | 1.28e-03 | | 7.60e-03 | | 1.15e-04 |
| PMO | 1.42e-01 | 9.58e-01 | 5.24e-02 | 3.70e-01 | 4.48e-01 | 7.53e-02 | 6.04e-01 | 0 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Residential Natural Gas Combustion | Cast Iron Cupola | Copper Processing | Asphalt Manufacturing | Fly Ash | Sandblast | LDDV Exhaust | Ammonium Nitrate Production |
|---|---|---|---|---|---|---|---|---|
| OC | 4.90e-01 | 6.37e-02 | 1.00e-02 | 4.32e-02 | 1.24e-02 | 5.00e-03 | 3.55e-01 | |
| EC | 6.70e-02 | 9.10e-03 | 9.90e-04 | 5.72e-02 | 1.70e-02 | | 5.14e-01 | |
| $NH_4^+$ | 4.40e-03 | | | 1.56e-03 | 7.82e-04 | | 7.30e-03 | 2.13e-01 |
| $NO_3^-$ | 3.41e-02 | 5.00e-03 | | 1.01e-03 | 3.65e-03 | 5.00e-04 | 2.30e-03 | 7.35e-01 |
| $SO_4^{2-}$ | 1.26e-01 | 6.28e-02 | 5.20e-04 | 6.64e-03 | 6.39e-02 | 5.50e-03 | 8.60e-03 | |
| NCOM | 1.96e-01 | 2.55e-02 | 4.00e-03 | 1.73e-02 | 4.94e-03 | 2.00e-03 | 8.88e-02 | |
| MO | 4.40e-04 | 3.40e-01 | 5.39e-02 | 2.68e-01 | 2.34e-01 | 4.14e-01 | 4.76e-03 | 0 |
| $H_2O$ | 0 | 1.51e-02 | 1.25e-04 | 1.97e-03 | 1.55e-02 | 1.32e-03 | 0 | 5.12e-02 |
| Na | 2.13e-02 | 1.18e-02 | | 1.24e-02 | 2.23e-03 | 8.00e-03 | 3.83e-04 | |
| Mg | 0 | | | 2.12e-03 | 1.55e-02 | | 1.28e-04 | |
| Al | 2.20e-03 | 1.00e-02 | | 5.95e-02 | 5.37e-02 | 5.55e-02 | 2.10e-04 | |
| Si | 2.80e-03 | 2.18e-01 | | 1.47e-01 | 6.63e-02 | 2.82e-01 | 2.99e-03 | |
| P | 3.00e-04 | | | 1.11e-03 | 2.50e-03 | 3.10e-04 | 2.60e-04 | |
| S | 4.20e-02 | 2.09e-02 | 1.73e-04 | 2.32e-03 | 2.38e-02 | 8.80e-03 | 4.69e-03 | |
| Cl | 3.88e-02 | 8.10e-03 | 1.19e-02 | 3.57e-04 | 2.49e-03 | 1.41e-02 | 2.04e-04 | |
| K | 1.70e-03 | 2.73e-02 | 2.36e-03 | 7.15e-03 | 5.75e-03 | 9.94e-03 | 2.41e-05 | |
| Ca | 1.04e-02 | 9.10e-03 | 4.34e-03 | 6.02e-02 | 2.02e-01 | 8.20e-03 | 2.83e-04 | |
| Ti | 1.00e-03 | 5.46e-04 | | 4.37e-03 | 8.45e-03 | 4.00e-03 | 0 | |
| V | 1.80e-04 | 8.19e-05 | | 0 | 4.39e-04 | 8.00e-05 | 0 | |
| Cr | 5.00e-04 | 4.73e-04 | | 0 | 3.60e-04 | 5.50e-03 | 5.20e-06 | |
| Mn | 2.00e-04 | 4.09e-02 | 7.00e-05 | 5.72e-04 | 8.98e-04 | 1.40e-03 | 2.97e-06 | |
| Fe | 9.00e-04 | 1.36e-01 | 2.44e-03 | 3.28e-02 | 4.84e-02 | 6.30e-02 | 8.80e-04 | |
| Co | | 3.64e-05 | | 0 | 0 | | 6.51e-07 | |
| Ni | 9.00e-04 | 3.18e-04 | 9.30e-04 | 1.10e-05 | 9.50e-05 | 3.20e-03 | 2.88e-06 | |
| Cu | 0 | 2.36e-03 | 1.71e-01 | 6.40e-05 | 5.64e-04 | 5.00e-04 | 3.85e-05 | |
| Zn | 0 | 7.55e-03 | 2.21e-02 | 0 | 7.02e-04 | 3.25e-03 | 5.60e-04 | |
| Ga | | | 2.70e-04 | 3.00e-06 | 0 | | 2.43e-06 | |
| As | 0 | 1.18e-04 | 7.50e-04 | 0 | 4.45e-05 | 8.00e-05 | 5.35e-06 | |
| Se | 6.10e-04 | 1.82e-05 | 1.57e-03 | 5.00e-06 | 5.00e-06 | | 0 | |
| Br | 4.00e-04 | 8.19e-05 | 2.70e-04 | 1.80e-05 | 2.30e-05 | 2.50e-05 | 6.81e-06 | |
| Rb | 2.00e-04 | 2.00e-04 | 1.00e-05 | 2.80e-05 | 3.85e-05 | 0 | 0 | |
| Sr | 0 | | | 3.49e-04 | 9.11e-04 | 3.90e-04 | 0 | |
| Zr | | | 4.00e-05 | 1.89e-04 | 1.60e-04 | 2.75e-03 | 0 | |
| Mo | | | 4.60e-04 | 3.60e-05 | 0 | 5.50e-03 | 0 | |
| Pd | | | | 4.10e-05 | 0 | | 2.00e-05 | |
| Ag | | | | 0 | 6.00e-06 | | 5.50e-05 | |
| Cd | | | | 2.80e-05 | 2.00e-05 | 5.00e-04 | 4.05e-05 | |
| In | | | | 0 | 4.80e-05 | | 3.50e-05 | |
| Sn | | | 4.40e-03 | 0 | 1.70e-05 | | 1.90e-05 | |
| Sb | | 3.37e-03 | | 0 | 1.30e-05 | | 7.28e-05 | |
| Ba | 0 | | | 2.16e-03 | 1.90e-03 | 0 | 1.73e-04 | |
| La | | | | 2.27e-03 | 2.57e-04 | | 5.40e-05 | |
| Ce | | | | | 1.50e-04 | | | |
| Hg | | | | 2.00e-06 | 5.00e-06 | 1.00e-05 | 0 | |
| Pb | 0 | 2.09e-03 | | 0 | 4.29e-04 | 5.00e-04 | 1.55e-04 | |
| PMO | 0 | 0 | 7.08e-01 | 2.71e-01 | 2.34e-01 | 1.03e-01 | 1.22e-02 | 0 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Limestone Dust | Phosphate Manufacturing | Gypsum Manufacturing | Urea Fertilizer | Lead Processing | Crustal Material | Copper Production | Brick Grinding and Screening |
|---|---|---|---|---|---|---|---|---|
| OC | | 7.84e-02 | | 3.10e-01 | | 5.38e-02 | | 2.42e-02 |
| EC | | 2.74e-02 | | 2.00e-02 | | 2.05e-03 | | 5.77e-04 |
| $NH_4^+$ | | 3.59e-02 | | | | 1.25e-03 | | 4.42e-04 |
| $NO_3^-$ | | 1.97e-03 | 5.00e-04 | 5.50e-03 | | 5.89e-04 | | 2.19e-03 |
| $SO_4^{2-}$ | 1.71e-02 | 2.61e-02 | 4.10e-01 | 4.00e-02 | 7.53e-02 | 1.80e-03 | 1.50e-03 | 9.83e-03 |
| NCOM | | 3.14e-02 | | 1.24e-01 | | 2.15e-02 | | 9.68e-03 |
| MO | 2.15e-01 | 7.49e-02 | 0 | 0 | 1.21e-02 | 3.57e-01 | 2.38e-02 | 2.23e-01 |
| $H_2O$ | 4.10e-03 | 1.49e-02 | 9.84e-02 | 9.60e-03 | 1.81e-02 | 7.32e-04 | 3.60e-04 | 2.47e-03 |
| Na | 4.00e-04 | 3.32e-03 | | | 3.00e-02 | 3.17e-03 | | 8.26e-03 |
| Mg | 4.70e-02 | 4.20e-04 | | | 1.00e-05 | 7.23e-03 | | 6.38e-03 |
| Al | 1.26e-02 | 7.57e-03 | | | 3.00e-05 | 6.79e-02 | 0 | 2.33e-02 |
| Si | 4.45e-02 | 2.79e-02 | | | 3.00e-03 | 2.21e-01 | 0 | 9.92e-02 |
| P | 8.30e-04 | 3.02e-02 | | | 5.00e-04 | 6.85e-04 | | 2.95e-04 |
| S | 5.72e-03 | 8.70e-03 | 1.37e-01 | 1.30e-02 | 2.51e-02 | 1.61e-03 | 5.00e-04 | 5.95e-03 |
| Cl | 2.35e-03 | 1.09e-03 | 5.50e-03 | 1.10e-01 | 1.93e-01 | 5.14e-04 | | 3.85e-03 |
| K | 2.17e-03 | 3.40e-03 | 5.50e-03 | 5.50e-03 | 1.00e-02 | 1.88e-02 | 3.46e-03 | 8.67e-03 |
| Ca | 2.99e-01 | 4.86e-03 | 1.30e-01 | 5.50e-03 | 3.00e-04 | 3.81e-02 | 0 | 1.83e-01 |
| Ti | 6.05e-04 | 9.50e-05 | | | | 4.62e-03 | 1.00e-04 | 1.42e-03 |
| V | 2.00e-05 | 2.70e-04 | | | 1.00e-05 | 1.40e-04 | 3.00e-05 | 0 |
| Cr | 1.00e-05 | 1.50e-04 | 5.50e-03 | | 1.00e-05 | 1.05e-04 | 1.05e-04 | 5.00e-06 |
| Mn | 8.00e-04 | 3.00e-05 | 5.00e-04 | 5.00e-04 | 3.00e-05 | 9.55e-04 | 1.00e-04 | 4.68e-04 |
| Fe | 7.12e-03 | 5.20e-04 | 5.50e-03 | 5.00e-04 | 1.00e-03 | 4.38e-02 | 2.90e-03 | 1.99e-02 |
| Co | | 1.00e-05 | | 5.00e-04 | | 0 | | 0 |
| Ni | 2.00e-05 | 4.00e-05 | 5.50e-03 | 5.00e-04 | 1.00e-05 | 7.50e-05 | 5.50e-05 | 6.30e-05 |
| Cu | 1.60e-04 | 5.50e-05 | 5.00e-04 | 5.00e-04 | 3.00e-04 | 6.60e-05 | 7.30e-03 | 5.30e-05 |
| Zn | 5.30e-04 | 8.30e-04 | 5.00e-04 | 5.00e-04 | 3.00e-03 | 1.77e-04 | 4.56e-02 | 2.43e-04 |
| Ga | | 1.00e-05 | | | | 5.00e-06 | 1.20e-04 | 7.00e-06 |
| As | 2.00e-04 | 8.50e-05 | 0 | | 3.00e-03 | 1.45e-05 | | 6.00e-06 |
| Se | | 2.00e-05 | 5.00e-04 | 5.00e-04 | | 1.00e-06 | 6.40e-03 | 6.00e-06 |
| Br | 1.40e-04 | 1.05e-04 | 5.00e-04 | 5.00e-04 | | 1.85e-05 | 2.53e-02 | 1.90e-05 |
| Rb | | 5.00e-05 | | | | 8.30e-05 | | 2.50e-05 |
| Sr | 4.25e-04 | 9.50e-05 | 5.00e-04 | 0 | | 2.46e-04 | 4.00e-05 | 4.57e-04 |
| Zr | | 3.50e-05 | | | | 1.60e-04 | 7.50e-05 | 8.90e-05 |
| Mo | | 1.70e-04 | 5.00e-04 | 5.00e-04 | | 1.30e-05 | 1.30e-03 | 0 |
| Pd | | 4.00e-05 | | | | 1.60e-05 | | 1.04e-04 |
| Ag | | 5.00e-05 | | | | 2.00e-05 | 2.40e-04 | 6.80e-05 |
| Cd | 4.20e-04 | 1.30e-04 | 5.00e-04 | 5.00e-04 | 7.00e-03 | 1.80e-05 | 6.30e-03 | 0 |
| In | | 5.00e-05 | | | | 1.00e-05 | 3.65e-04 | 0 |
| Sn | | 5.00e-05 | | | 2.00e-02 | 2.25e-05 | 4.20e-03 | 1.07e-04 |
| Sb | | 3.00e-04 | | | 2.00e-03 | 1.10e-05 | 1.51e-02 | 7.70e-05 |
| Ba | 1.00e-05 | 1.20e-04 | 0 | | | 4.95e-04 | | 0 |
| La | | 0 | | | | 4.35e-05 | | 0 |
| Ce | | | | | | 3.00e-05 | | |
| Hg | | 7.00e-05 | | | | 1.00e-06 | | 0 |
| Pb | 1.36e-03 | 1.70e-04 | 5.00e-04 | | | 7.30e-05 | | 6.70e-05 |
| PMO | 3.44e-01 | 6.27e-01 | 3.29e-01 | 3.65e-01 | 6.21e-01 | 1.53e-01 | 8.55e-01 | 3.71e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

| Species | Calcium Carbide Furnace | Coke Calciner | Industrial Soil | Potato Deep-Frying | Sea Salt | Sludge Combustion | Lead Production | Steel Desulfurization |
|---|---|---|---|---|---|---|---|---|
| OC | 7.30e-02 | | 1.01e-02 | 6.27e-01 | | 8.41e-02 | | |
| EC | 1.20e-02 | | 1.13e-02 | 4.00e-02 | | 1.52e-02 | | |
| $NH_4^+$ | | | 1.43e-03 | | | | | |
| $NO_3^-$ | 5.70e-03 | | 3.25e-04 | | | 1.42e-02 | | |
| $SO_4^{2-}$ | 3.20e-02 | 4.86e-01 | 2.51e-02 | | 9.98e-02 | 1.90e-01 | 2.26e-01 | 8.22e-02 |
| NCOM | 2.92e-02 | | 4.04e-03 | 2.51e-01 | | 3.36e-02 | | |
| MO | 1.72e-01 | 2.38e-02 | 2.26e-01 | 0 | 0 | 1.44e-01 | 1.18e-01 | 2.00e-01 |
| $H_2O$ | 0 | 1.17e-01 | 6.36e-03 | | 2.40e-02 | 0 | 5.43e-02 | 1.97e-02 |
| Na | 9.20e-03 | | 2.94e-03 | | 3.99e-01 | 1.41e-02 | | |
| Mg | 2.40e-02 | | 5.95e-03 | | 4.79e-02 | 6.50e-03 | | |
| Al | 5.80e-03 | 5.40e-04 | 2.80e-02 | | | 1.80e-02 | 4.43e-03 | 8.78e-03 |
| Si | 2.50e-02 | 0 | 1.12e-01 | | | 4.77e-02 | 1.62e-02 | 1.63e-02 |
| P | | | 1.50e-03 | | | 3.26e-02 | 3.89e-03 | 2.26e-03 |
| S | 1.60e-02 | 1.62e-01 | 8.35e-03 | | 3.29e-02 | 6.33e-02 | 7.54e-02 | 2.74e-02 |
| Cl | 1.05e-02 | | 3.60e-03 | | 3.99e-01 | 1.48e-02 | 8.26e-03 | 4.58e-03 |
| K | 1.25e-02 | 3.70e-04 | 7.30e-03 | | 1.40e-02 | 9.60e-03 | 7.31e-03 | 2.54e-02 |
| Ca | 3.00e-01 | 2.58e-01 | 1.37e-01 | | 1.40e-02 | 5.07e-02 | 1.01e-02 | 2.54e-01 |
| Ti | | 9.00e-05 | 2.91e-03 | | | 4.40e-03 | 7.25e-04 | 1.65e-03 |
| V | 0 | 1.05e-03 | 2.00e-04 | | | 1.80e-03 | | 4.10e-04 |
| Cr | | 0 | 6.00e-04 | | | 5.80e-03 | 2.20e-04 | 6.40e-04 |
| Mn | 4.20e-04 | 0 | 2.50e-03 | | | 2.10e-03 | 1.17e-03 | 2.77e-03 |
| Fe | 5.40e-03 | 5.90e-04 | 2.56e-02 | | | 5.38e-02 | 1.88e-02 | 2.05e-01 |
| Co | | | 1.00e-05 | | | | | |
| Ni | 2.20e-04 | 2.80e-04 | 4.50e-05 | | | | 8.45e-04 | 1.60e-04 |
| Cu | 2.00e-04 | 0 | 1.95e-04 | | | 8.80e-03 | 6.01e-03 | 1.60e-04 |
| Zn | 1.50e-04 | 5.00e-05 | 1.25e-03 | | | 3.61e-03 | 3.30e-01 | 9.10e-04 |
| Ga | | | 0 | | | | | 2.00e-05 |
| As | | | 1.00e-05 | | | | 2.65e-02 | |
| Se | | | 0 | | | 1.50e-03 | 4.59e-04 | 4.00e-05 |
| Br | | 2.00e-05 | 0 | | 2.00e-03 | 3.80e-03 | 3.40e-04 | 6.00e-05 |
| Rb | | 0 | 4.00e-05 | | | | | 1.90e-04 |
| Sr | | 1.80e-04 | 3.17e-04 | | | | 6.06e-04 | 1.16e-03 |
| Zr | | | 1.59e-04 | | | | | 0 |
| Mo | | | 0 | | | | | 1.30e-04 |
| Pd | | | 0 | | | | 4.96e-04 | 0 |
| Ag | | | 0 | | | | 1.00e-03 | 1.50e-04 |
| Cd | | | 0 | | | 8.30e-03 | 4.65e-03 | 3.30e-04 |
| In | | | 7.50e-05 | | | | 2.66e-04 | 7.00e-05 |
| Sn | | | 0 | | | 1.39e-02 | 6.13e-03 | 8.00e-05 |
| Sb | | | 0 | | | 5.00e-04 | 9.69e-03 | 6.30e-04 |
| Ba | | 0 | 6.26e-04 | | | 2.70e-03 | 3.49e-04 | |
| La | | | 0 | | | | | 0 |
| Ce | | | | | | | | |
| Hg | | | 1.70e-05 | | | | 8.72e-04 | 1.00e-04 |
| Pb | 8.00e-05 | 5.00e-05 | 1.84e-04 | | | 2.94e-02 | 1.42e-01 | 5.90e-04 |
| PMO | 2.83e-01 | 1.13e-01 | 3.82e-01 | 8.22e-02 | 0 | 1.84e-01 | 0 | 1.71e-01 |

**Table S6.** Mass fractions of all species for each source category determined in this work, continued.

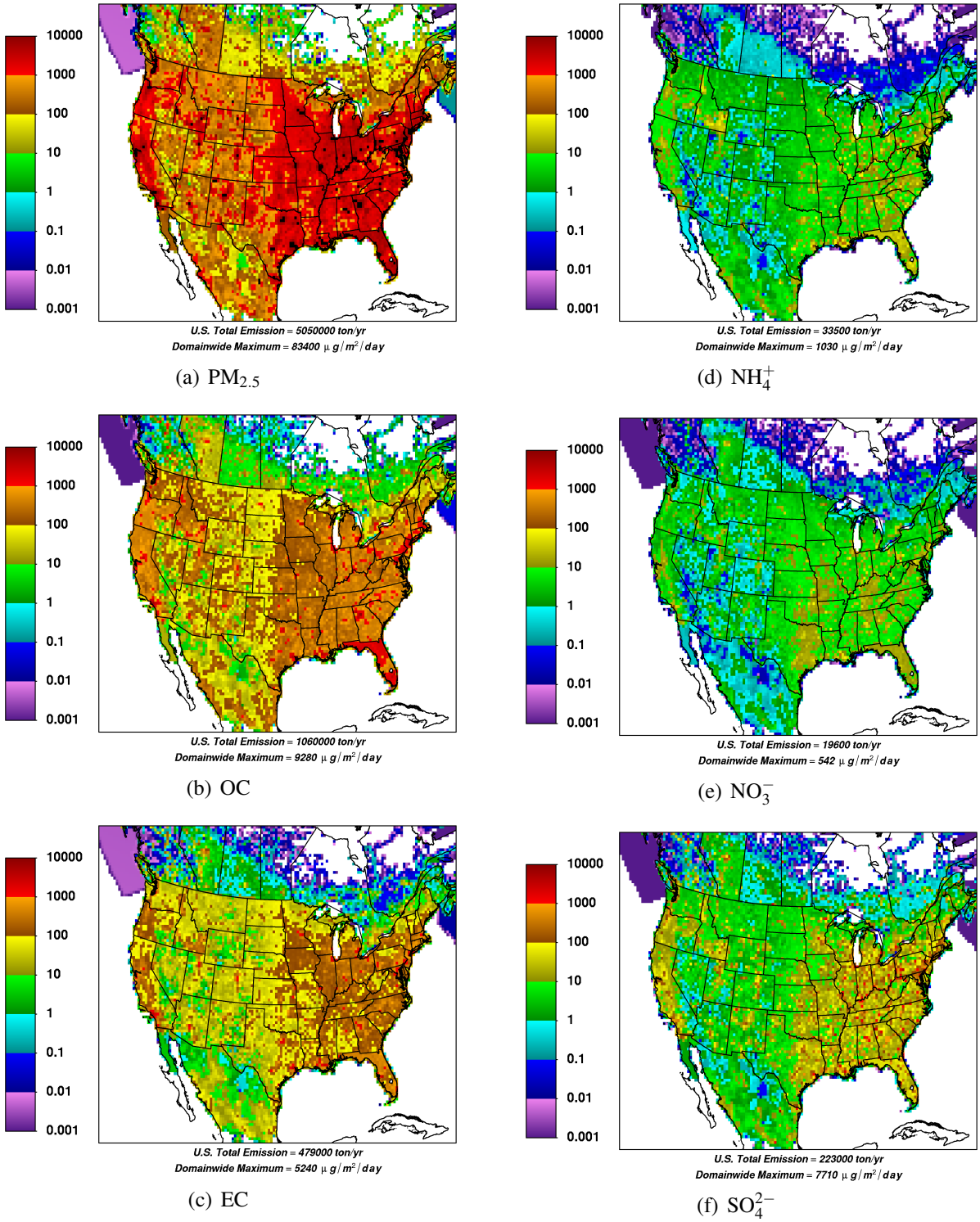
| Species | Auto Body Shredding | Ammonium Sulfate Production | Inorganic Fertilizer | Boric Acid Manufacturing |
|---|---|---|---|---|
| OC | 7.70e-02 | | 2.99e-02 | |
| EC | 6.00e-03 | | 1.11e-02 | |
| $NH_4^+$ | | 2.20e-01 | 3.00e-04 | |
| $NO_3^-$ | 1.50e-03 | | 3.50e-04 | 5.50e-03 |
| $SO_4^{2-}$ | 2.90e-03 | 5.86e-01 | 2.67e-02 | 2.00e-02 |
| NCOM | 3.08e-02 | | 1.20e-02 | |
| MO | 4.50e-02 | 0 | 2.96e-01 | 0 |
| $H_2O$ | 6.96e-04 | 1.94e-01 | 6.47e-03 | 4.80e-03 |
| Na | 1.80e-03 | | 1.22e-03 | |
| Mg | 1.00e-03 | | 8.70e-04 | |
| Al | 4.70e-03 | | 2.11e-02 | |
| Si | 8.70e-03 | | 3.51e-02 | |
| P | | | 1.12e-01 | |
| S | 4.70e-03 | 2.42e-01 | 1.13e-02 | 6.60e-03 |
| Cl | 6.80e-03 | | 3.09e-03 | 5.50e-03 |
| K | 2.70e-03 | | 5.85e-03 | |
| Ca | 4.80e-03 | | 2.38e-01 | 5.00e-04 |
| Ti | 2.50e-04 | | 5.78e-03 | |
| V | 1.00e-05 | | 2.35e-03 | |
| Cr | 4.00e-04 | | 1.59e-03 | |
| Mn | 8.80e-04 | | 1.77e-03 | |
| Fe | 5.76e-02 | | 4.66e-02 | 5.50e-03 |
| Co | | | 1.00e-05 | |
| Ni | 3.10e-04 | | 3.95e-04 | 5.00e-04 |
| Cu | 1.00e-03 | | 2.05e-04 | 5.00e-04 |
| Zn | 2.10e-02 | | 2.21e-03 | 5.00e-04 |
| Ga | | | 0 | |
| As | | | 4.00e-05 | |
| Se | | | 3.00e-05 | |
| Br | 4.90e-03 | | 1.40e-04 | |
| Rb | | | 3.50e-05 | |
| Sr | | | 3.17e-03 | |
| Zr | | | 6.95e-04 | |
| Mo | | | 7.00e-05 | |
| Pd | | | 2.50e-05 | |
| Ag | | | | 5.00e-04 |
| Cd | | | 9.00e-05 | 5.00e-04 |
| In | | | 4.90e-04 | |
| Sn | | | 3.80e-04 | |
| Sb | | | 1.31e-03 | |
| Ba | | | 1.53e-02 | |
| La | | | 4.91e-03 | |
| Ce | | | | |
| Hg | | | 3.50e-05 | |
| Pb | 4.90e-03 | | 7.30e-04 | |
| PMO | 7.14e-01 | 0 | 1.12e-01 | 9.56e-01 |

# S4 Spatial plots of $PM_{2.5}$ Emissions

Section S4.1 shows the spatial distributions of each $PM_{2.5}$ species in our inventory. Section S4.2 shows the spatial distributions of $PM_{2.5}$ emissions from each of the 84 source categories. Maps are arranged in the order indicated by Figure 2 of the main paper. Note that grid cells where the emissions flux exceeds the scale maximum are indicated by black tiles; grid cells lower than the scale minimum are indicated by the dark purple at the bottom of the legend; cells where the flux is zero are indicated by white space.

Spatial allocation was performed with the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (`http://www.smoke-model.org`), and maps were created with the R statistical software (*30*). A number of the maps such as Solid Waste Combustion (Figure S14(c)) and Asphalt Roofing (Figure S18(a)) suggest that emissions from Canada and/or Mexico are substantially greater than those from the U.S.; however this is likely an indicator of different reporting practices on the part of industries and/or local governments rather than actual differences in emissions. A close examination of the spatial plots also reveals data gaps and reporting inconsistencies within the $PM_{2.5}$ NEI in certain source categories from specific states. One of the most prominent inconsistencies is the lack of emissions in Colorado from Agricultural Burning, Residual Oil Combustion, Dairy Soil, and Solid Waste Combustion. In contrast, Illinois appears to be the only state to report emissions from the Food & Ag - Handling, Food & Ag Drying, and Urea Fertilizer source categories. Similarly, Idaho reported strkingly large area source emissions from Wood-Fired Boilers and Asphalt Roofing. Inconsistencies in emission reporting across state boundaries pose a difficulty when interpreting and analyzing numerical model results for specific $PM_{2.5}$ source categories.

## S4.1 Emissions of $PM_{2.5}$ Species

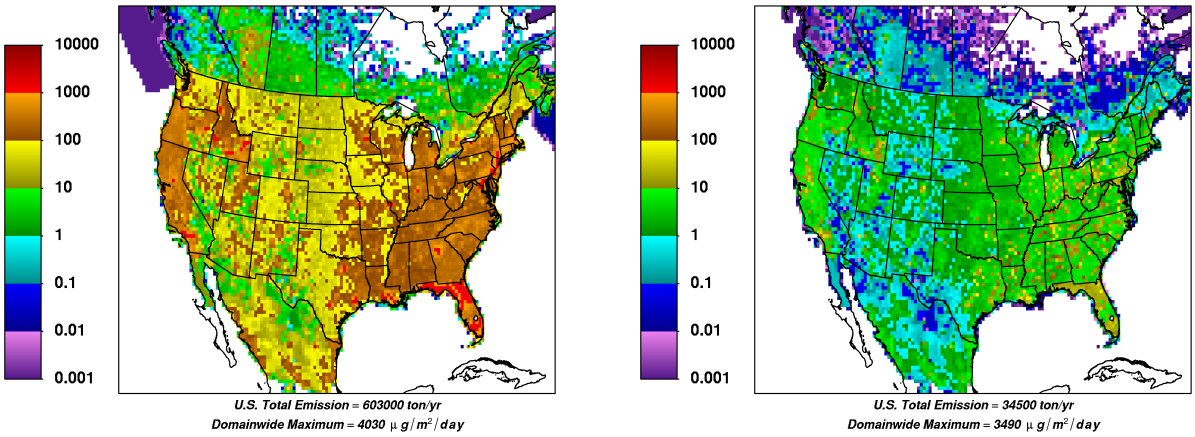



(a) $PM_{2.5}$ (d) $NH_4^+$

(b) OC (e) $NO_3^-$

(c) EC (f) $SO_4^{2-}$

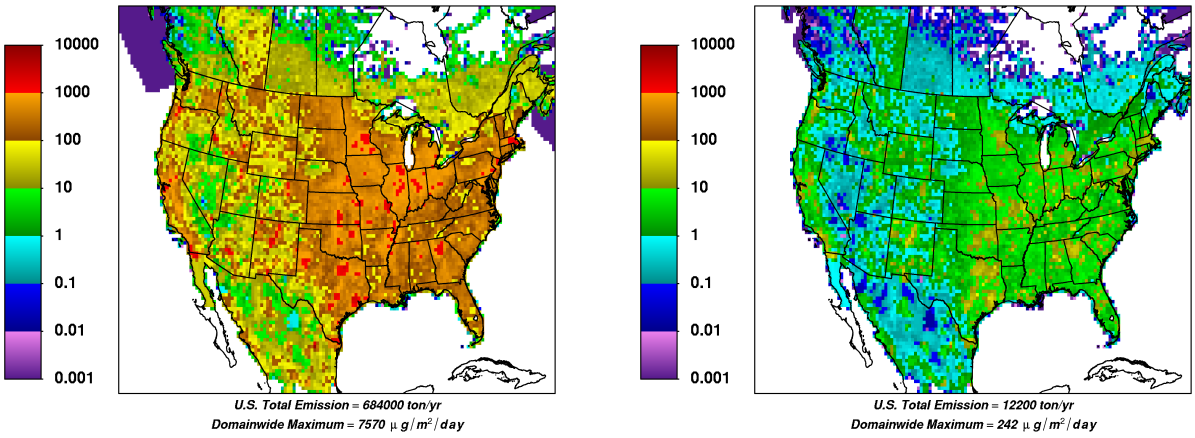
**Figure S2.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu$g/m$^2$/year).




(a) NCOM



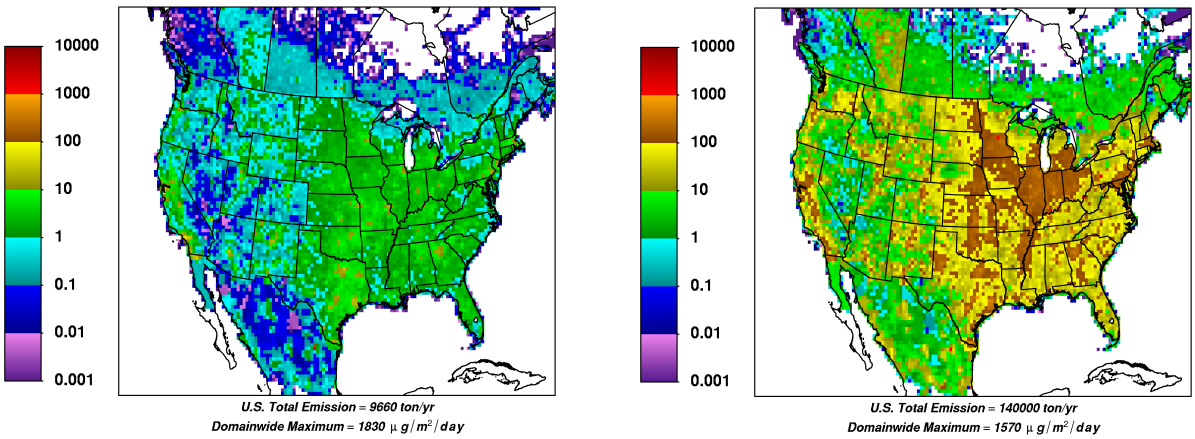

(d) Na




(b) MO




(e) Mg

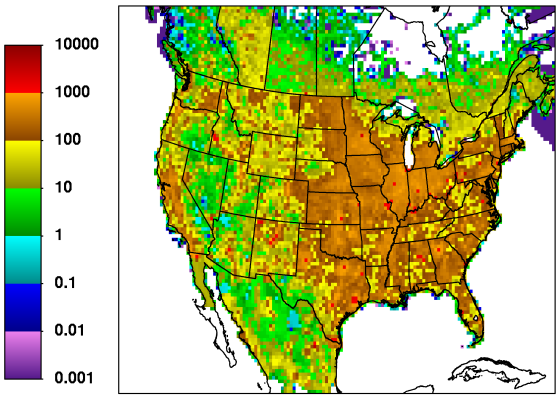



(c) $H_2O$

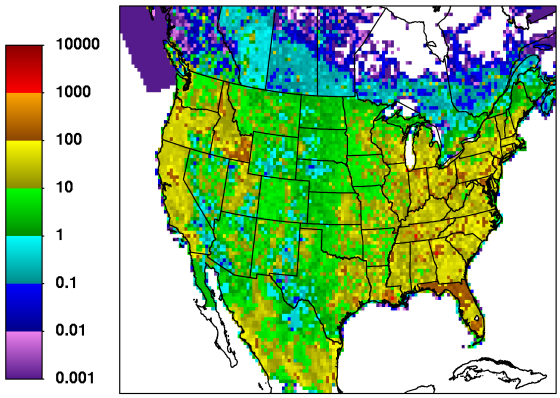



(f) Al

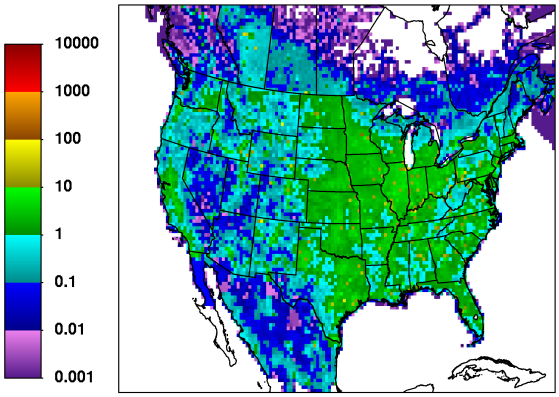
**Figure S3.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu g/m^2$/year), continued.



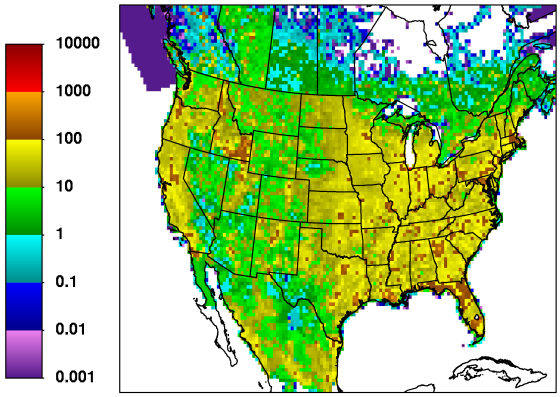

(a) Si



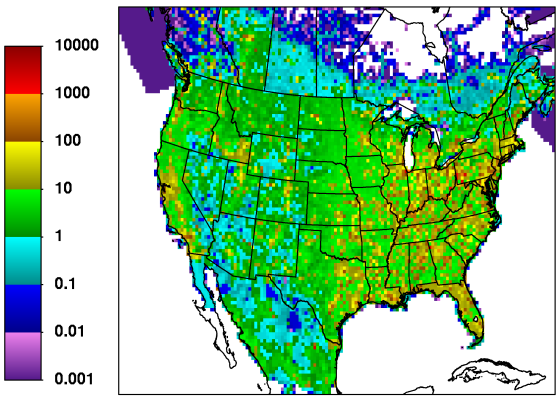

(d) Cl



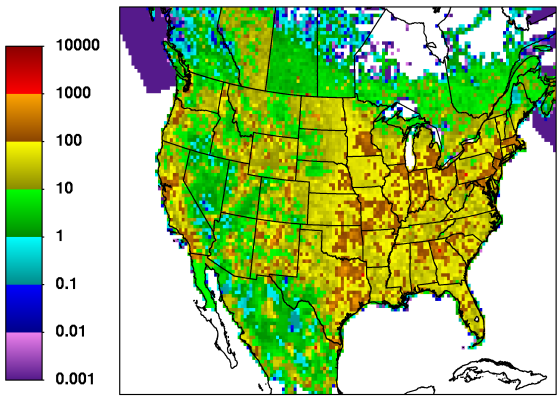

(b) P




(e) K

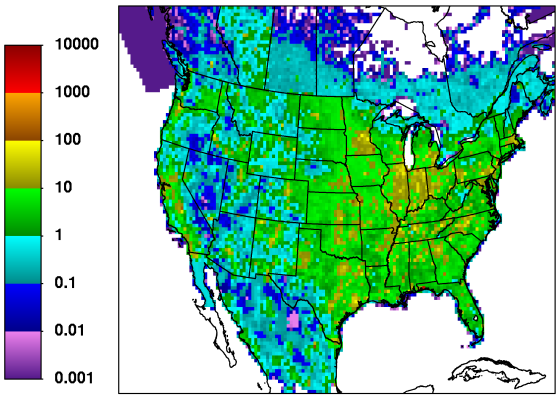



(c) S

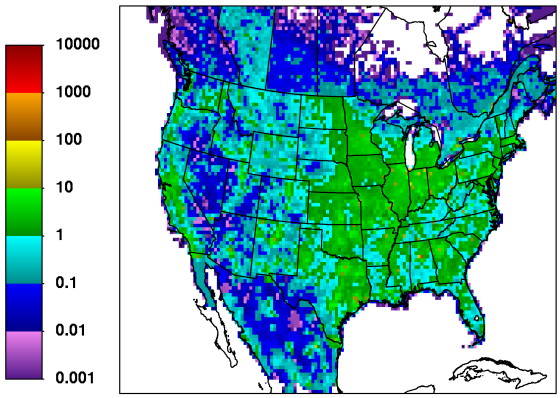



(f) Ca

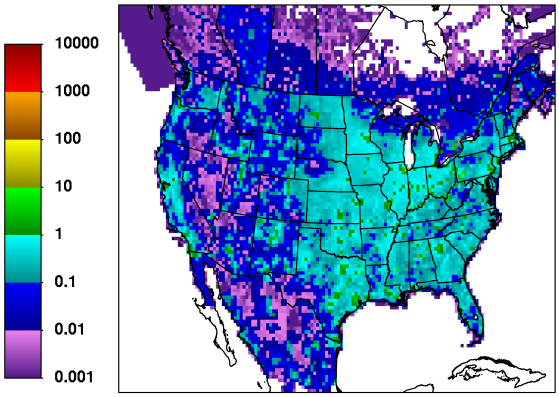
**Figure S4.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu g/m^2$/year), continued.



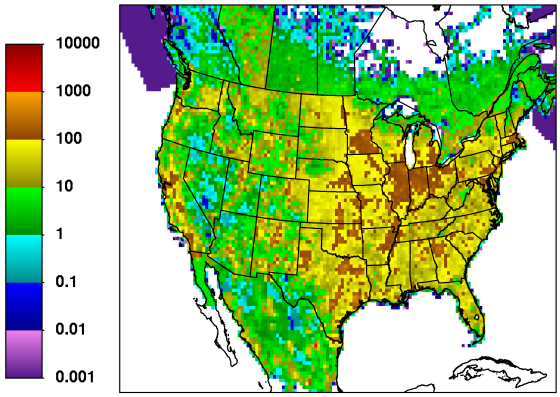

(a) Ti



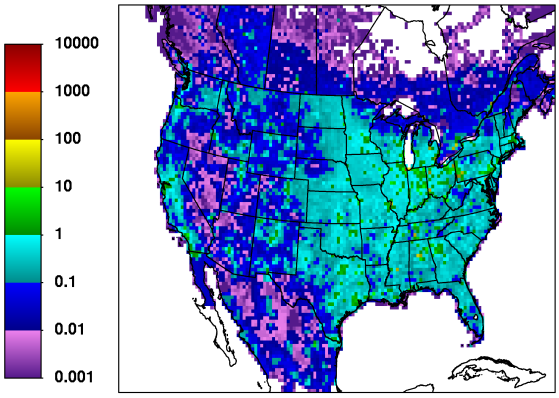

(d) Mn



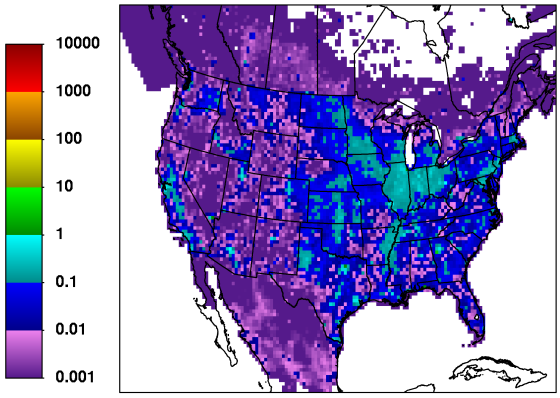

(b) V




(e) Fe

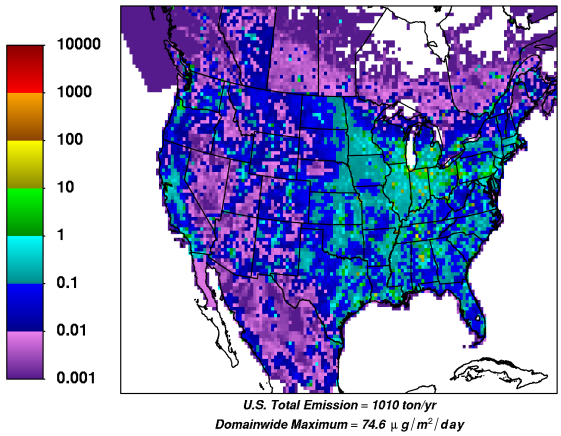



(c) Cr

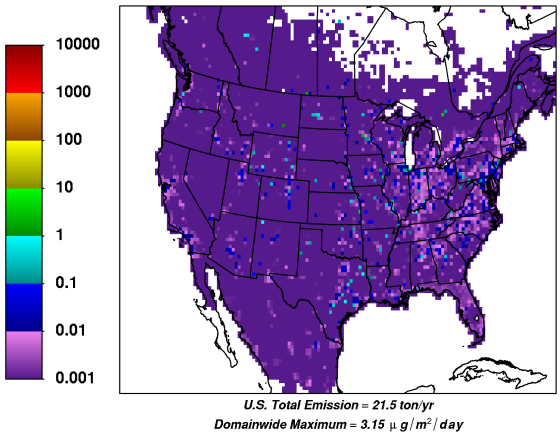



(f) Co

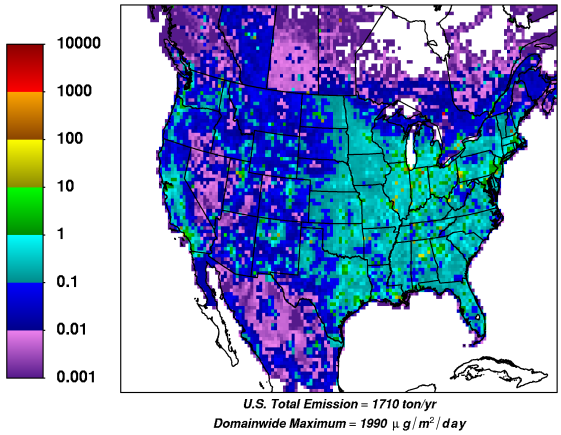
**Figure S5.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu g/m^2$/year), continued.



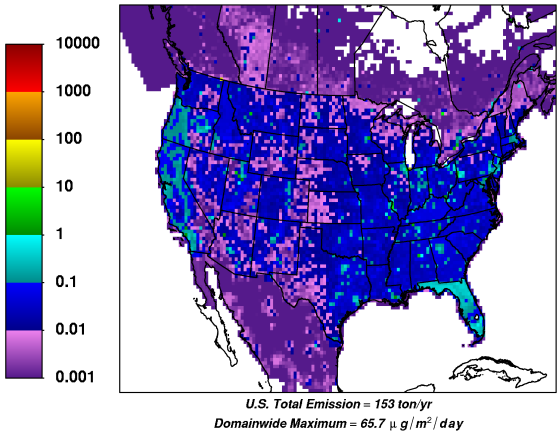

(a) Ni



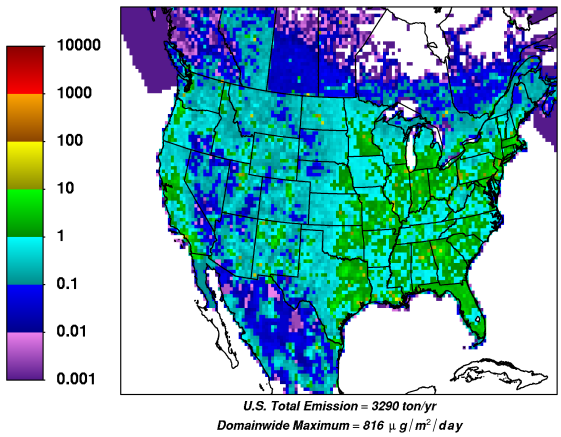

(d) Ga



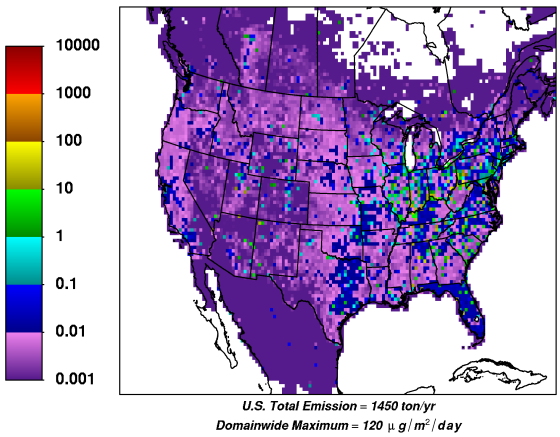

(b) Cu




(e) As

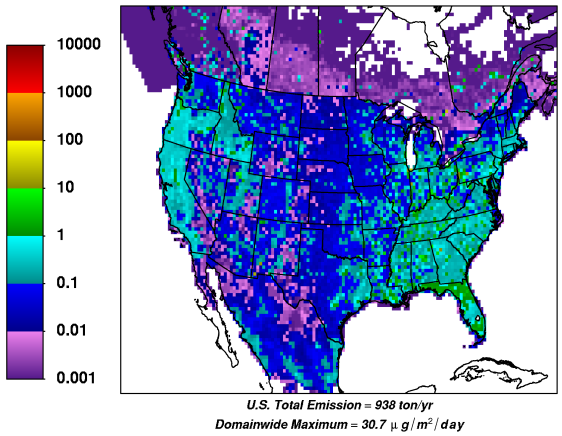



(c) Zn

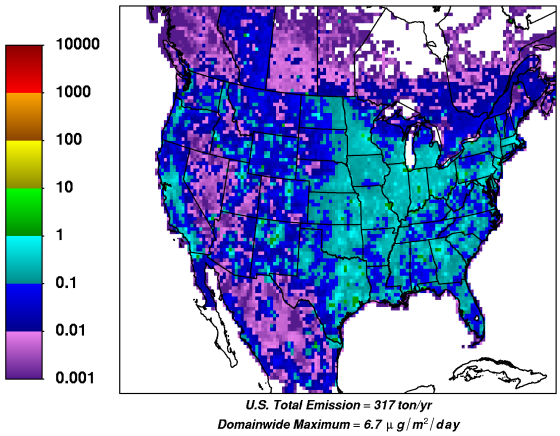



(f) Se

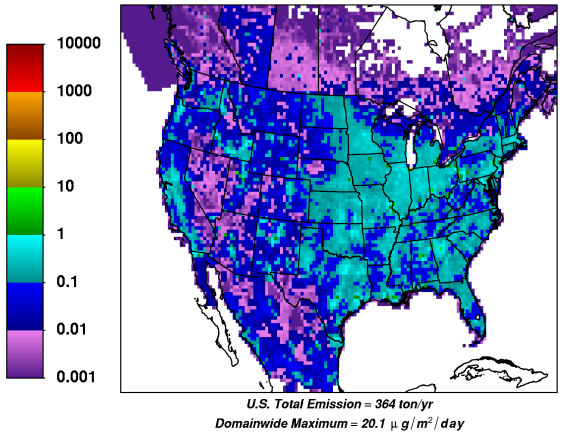
**Figure S6.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu g/m^2$/year), continued.



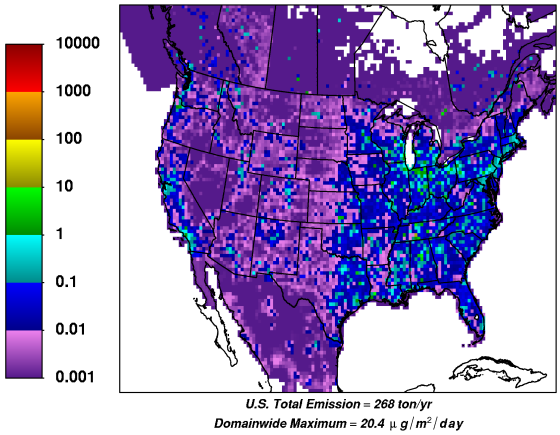

(a) Br



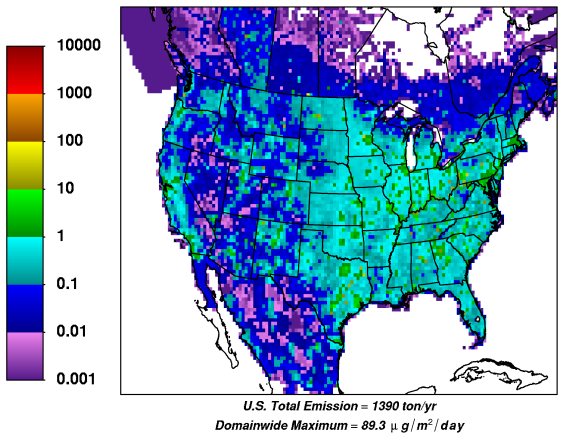

(d) Zr



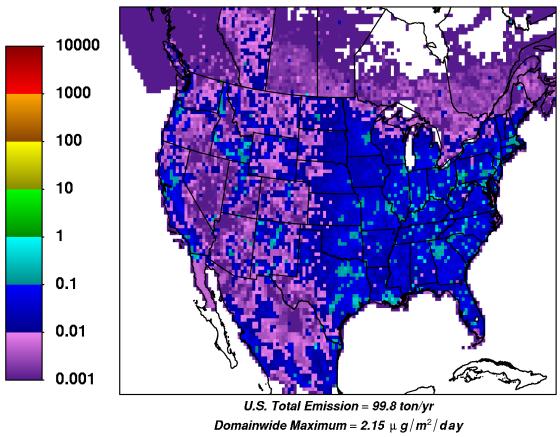

(b) Rb




(e) Mo

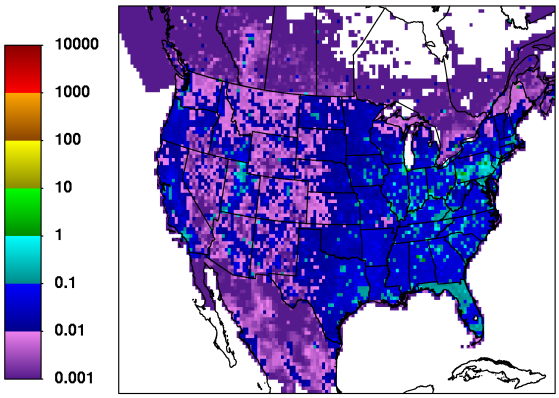



(c) Sr

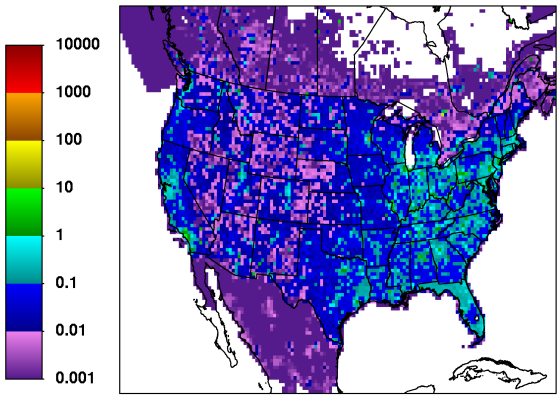



(f) Pd

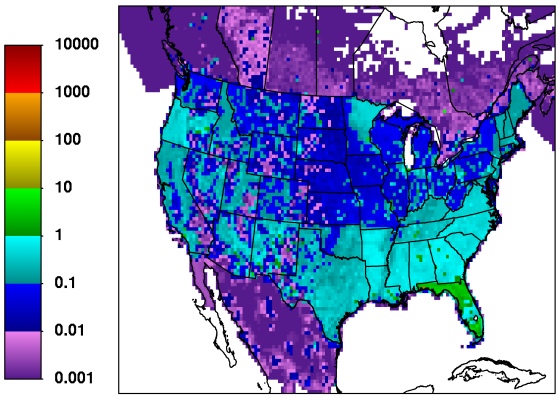
**Figure S7.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu$g/m$^2$/year), continued.



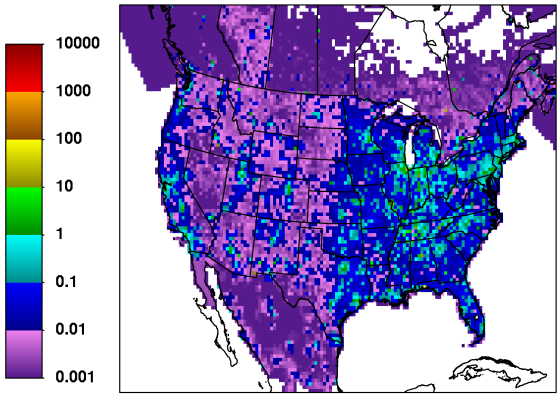

(a) Ag



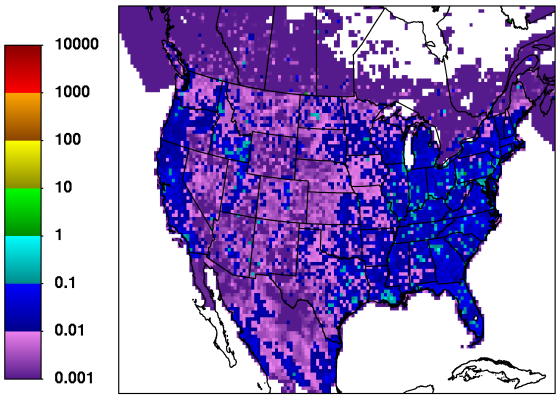

(d) Sn



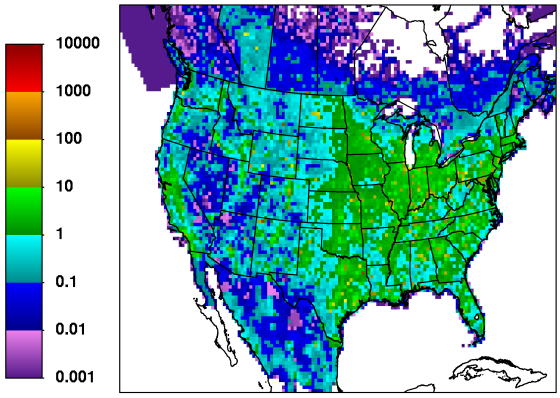

(b) Cd




(e) Sb

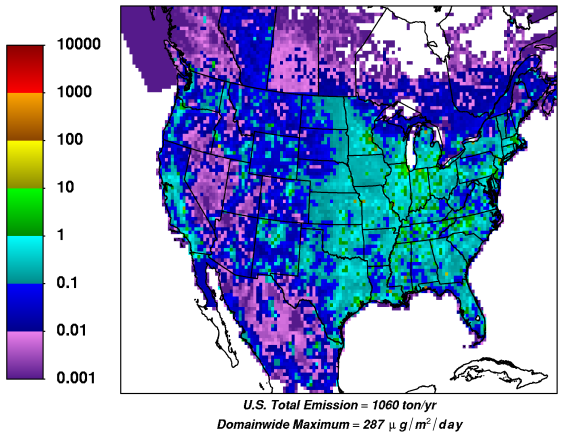



(c) In

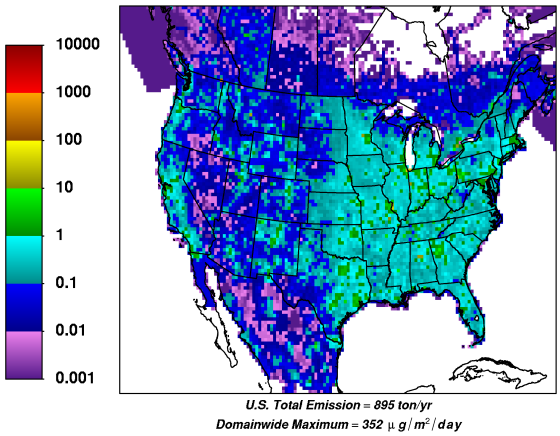



(f) Ba

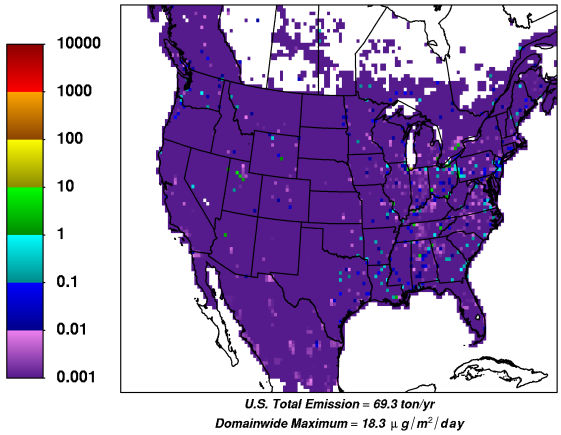
**Figure S8.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu g/m^2$/year), continued.



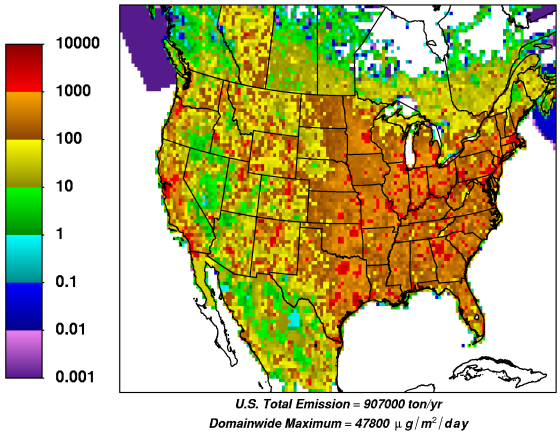

(a) La



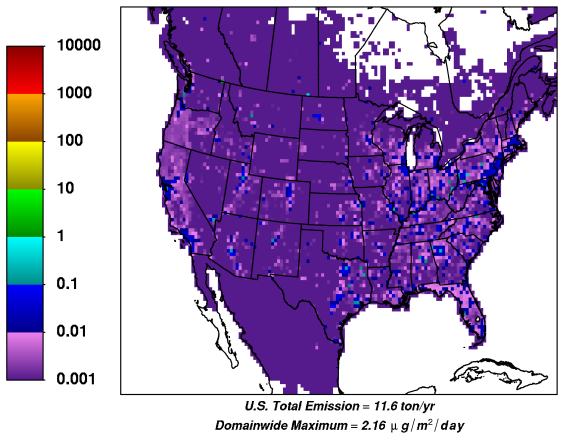

(d) Pb




(b) Ce



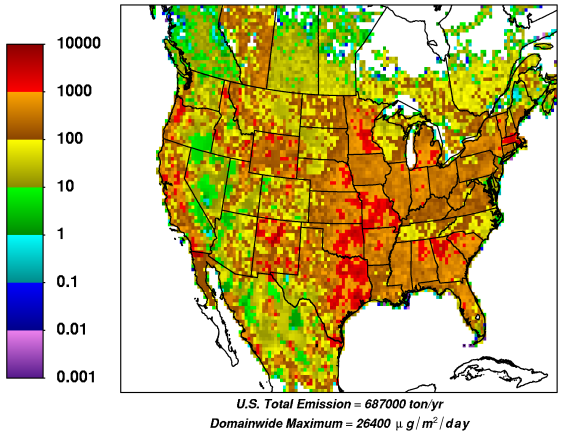

(e) PMO



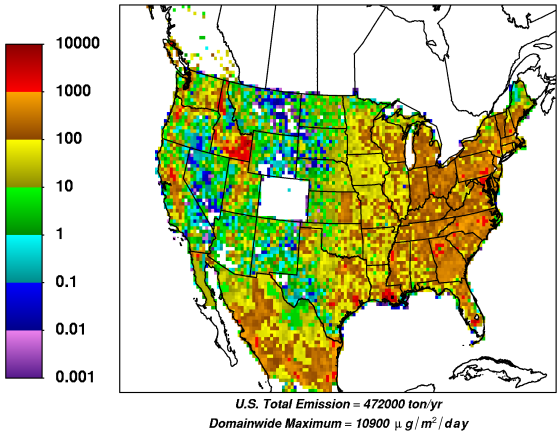

(c) Hg

**Figure S9.** Spatial Plots of $PM_{2.5}$ Species Emissions ($\mu$g/m$^2$/year), continued.

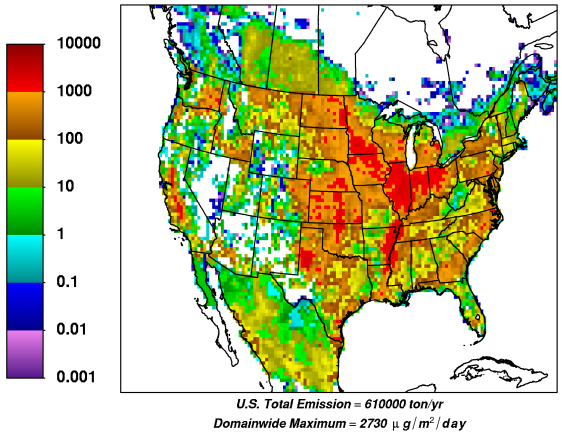
## S4.2 Emissions of $PM_{2.5}$ from Source Categories



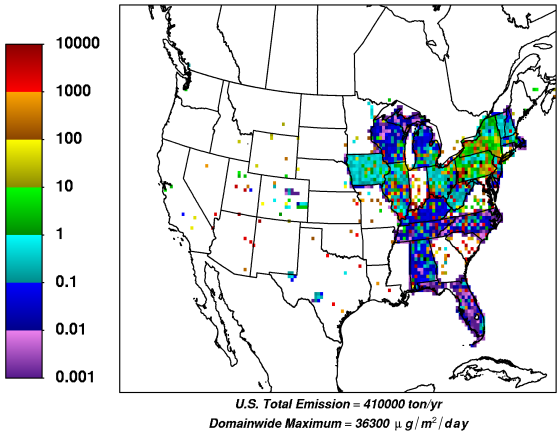

(a) Unpaved Road Dust



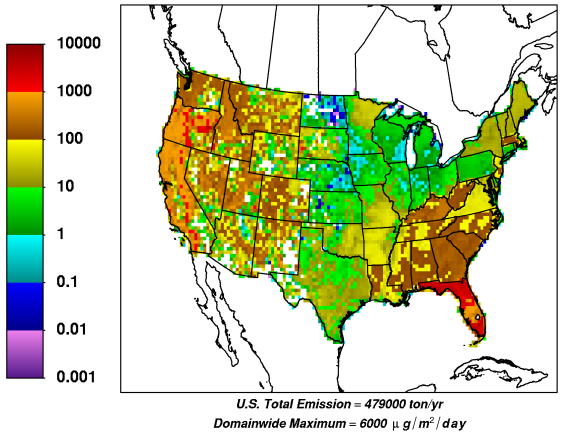

(d) Agricultural Burning



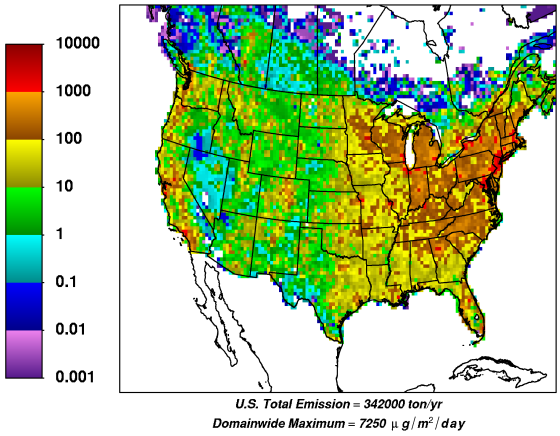

(b) Agricultural Soil




(e) Bituminous Coal Combustion

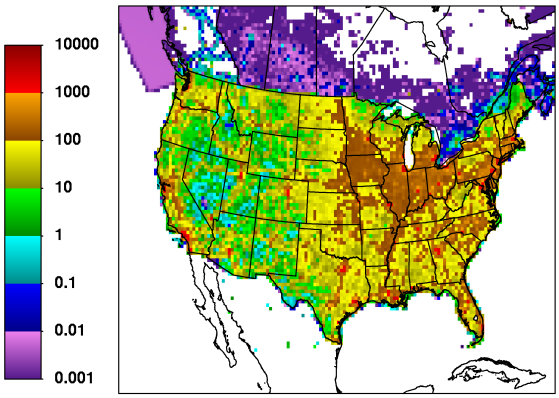



(c) Wildfires

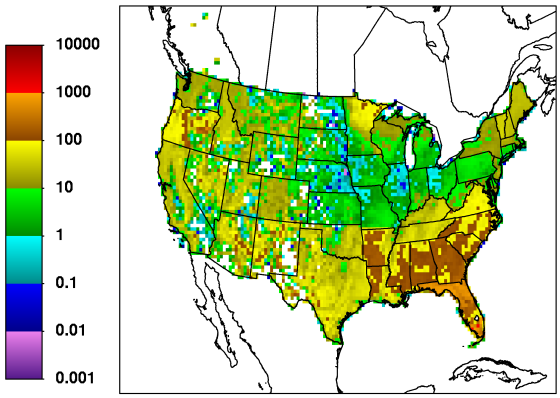



(f) Residential Wood Combustion

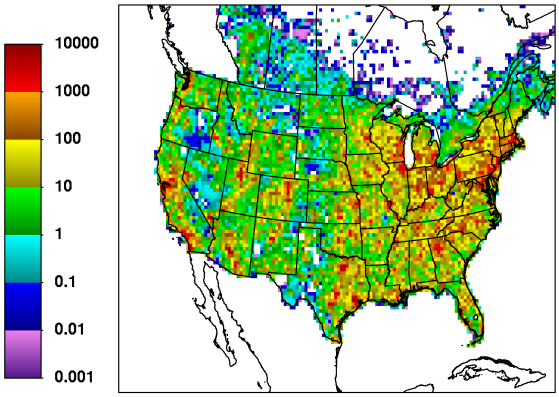
**Figure S10.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category.



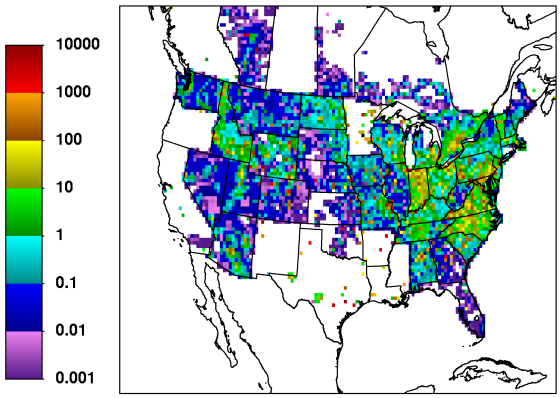

(a) HDDV Exhaust



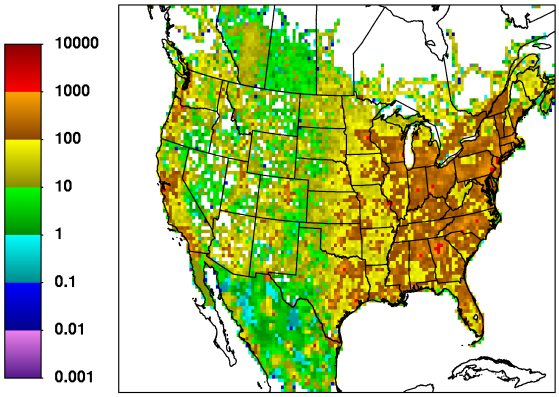

(d) Prescribed Burning



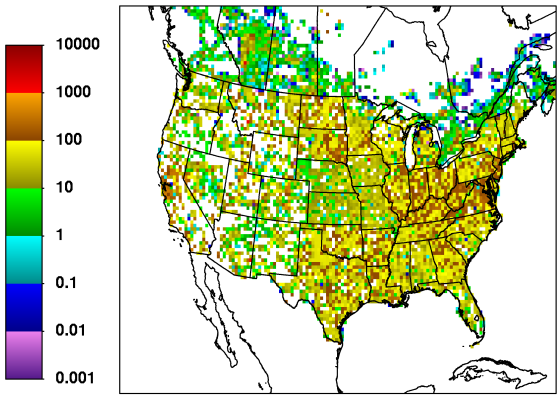

(b) Construction Dust




(e) Sub-bituminous Coal Combustion

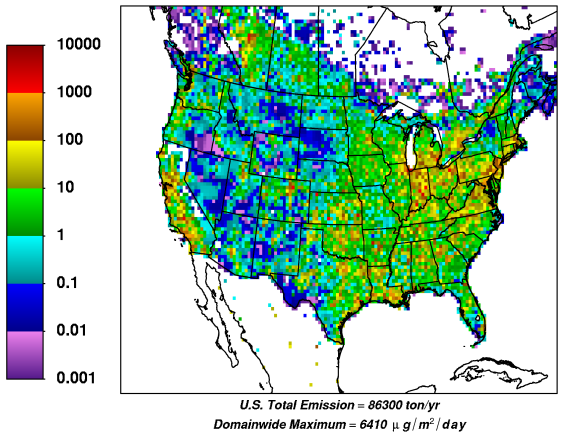



(c) Paved Road Dust

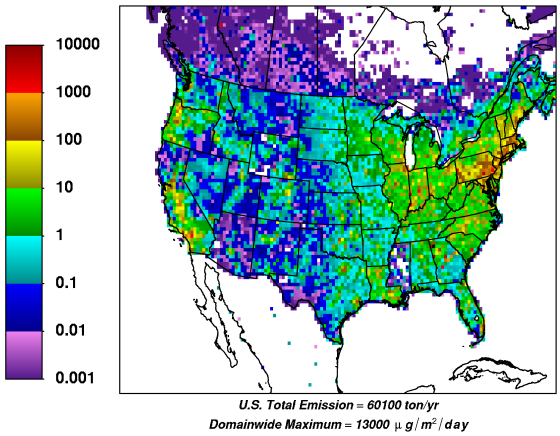



(f) Sand & Gravel

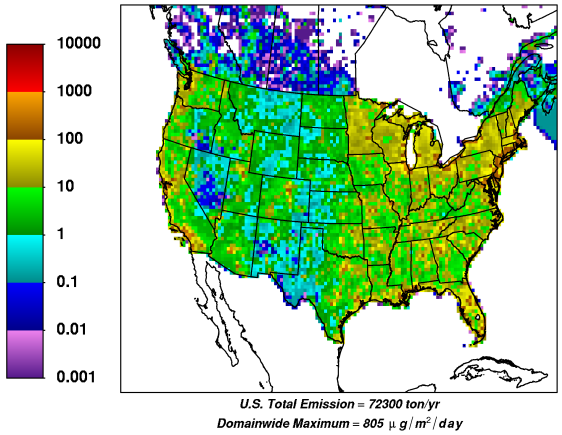
**Figure S11.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



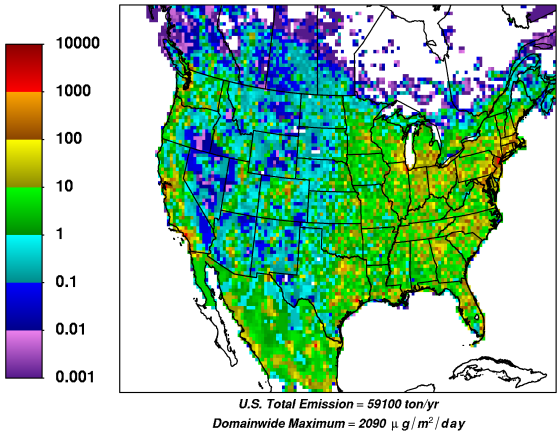

(a) Natural Gas Combustion



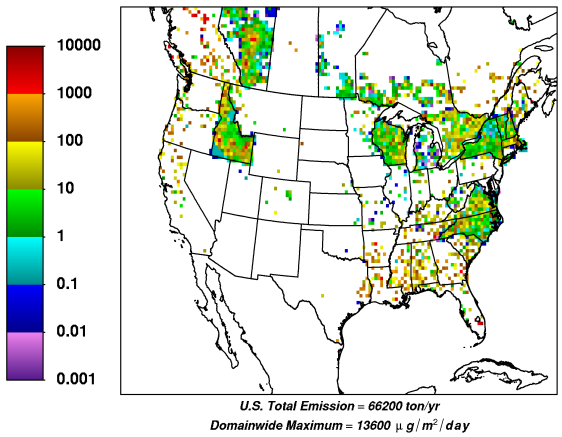

(d) Distillate Oil Combustion



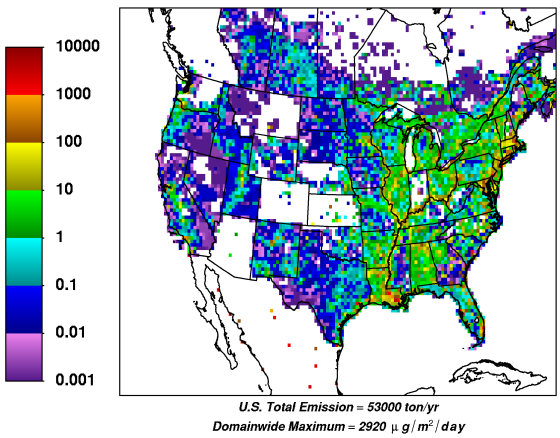

(b) Nonroad Gasoline Exhaust




(e) Charbroiling

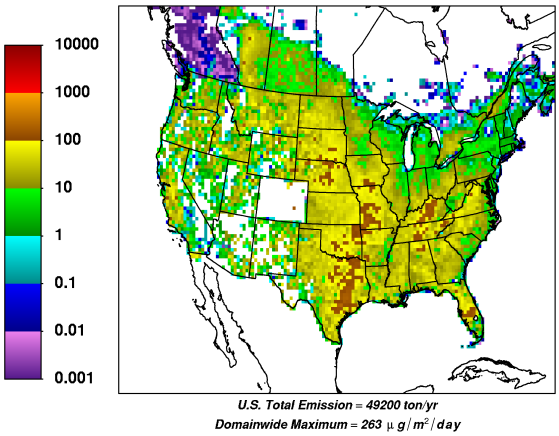



(c) Wood-Fired Boiler

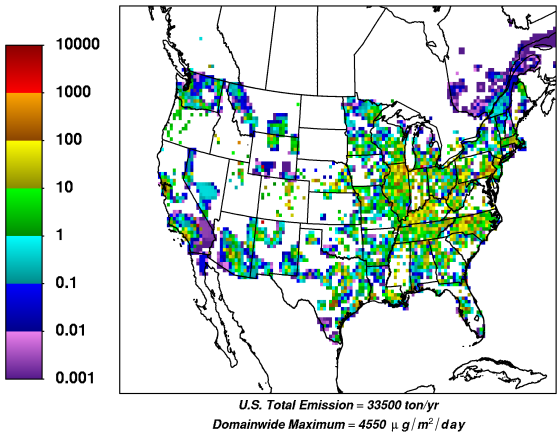



(f) Residual Oil Combustion

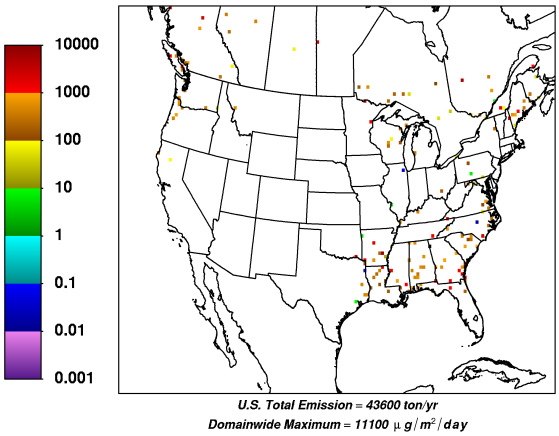
**Figure S12.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu$g/m$^2$/year) by Source Category, continued.



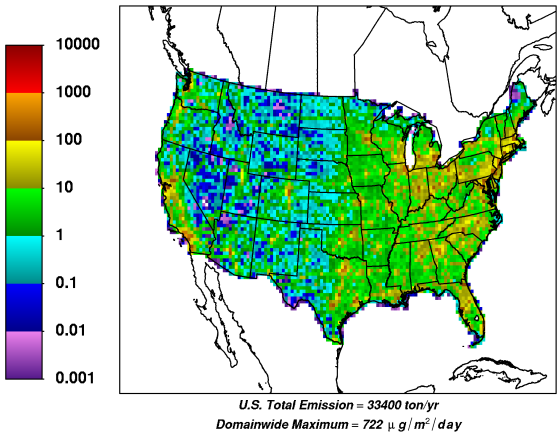

(a) Dairy Soil



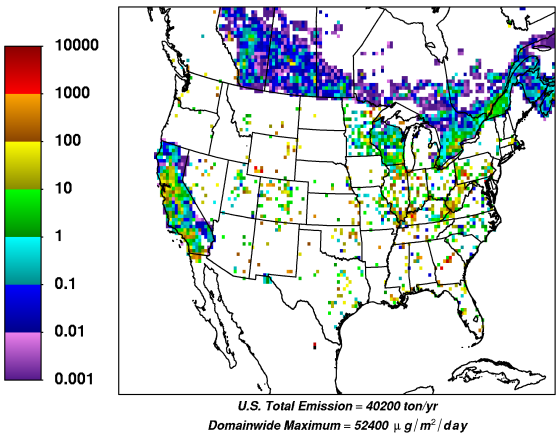

(d) Industrial Manufacturing - Avg



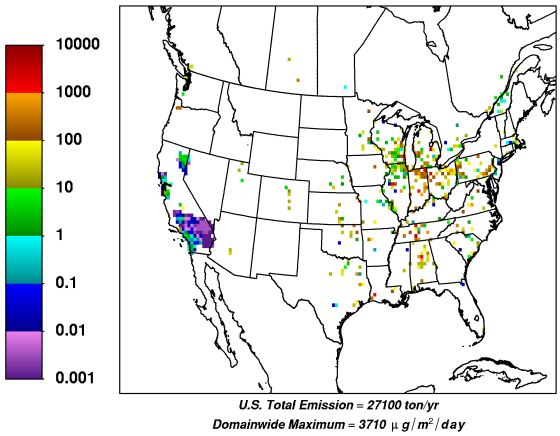

(b) Kraft Recovery Furnace




(e) Onroad Gasoline Exhaust

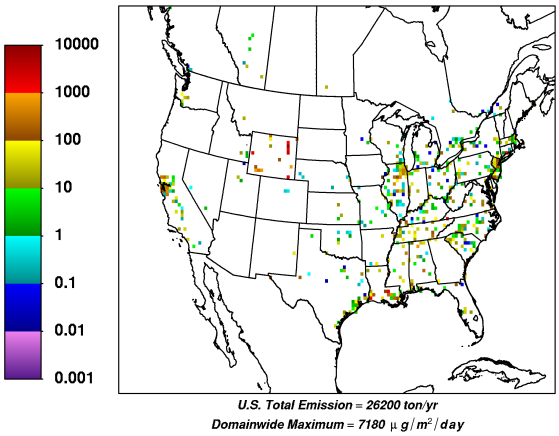



(c) Mineral Products - Avg

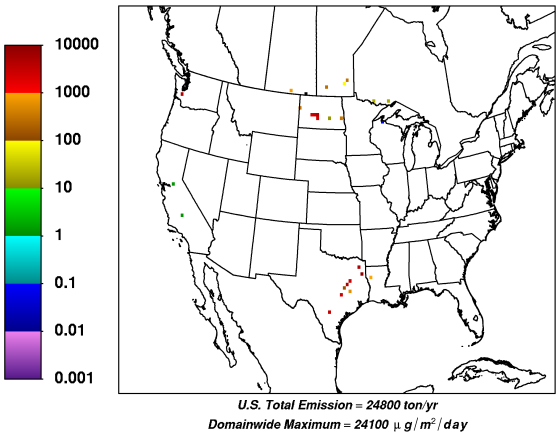



(f) Heat Treating

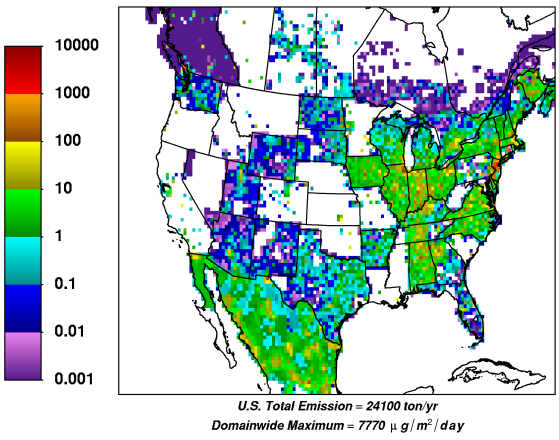
**Figure S13.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



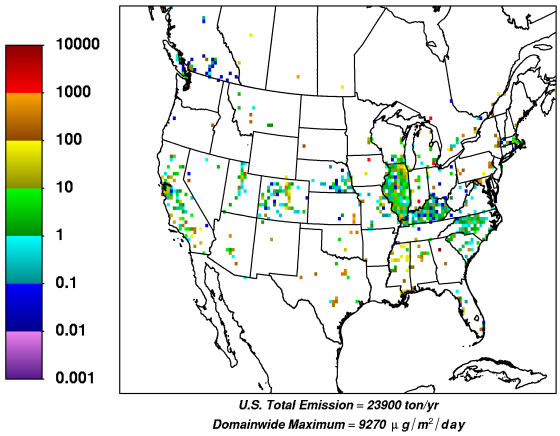

(a) Chemical Manufacturing - Avg



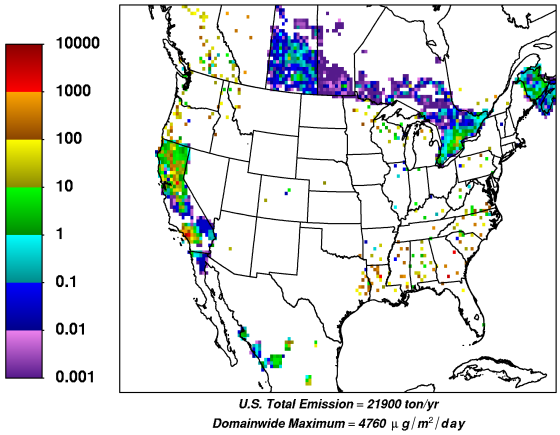

(b) PM & $SO_2$ Controlled Lignite Combustion



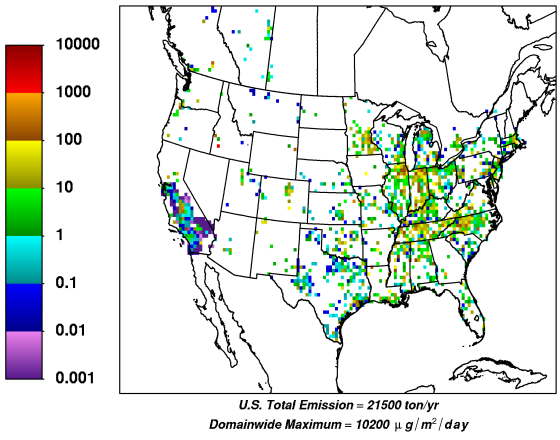

(c) Solid Waste Combustion




(d) Cement Production

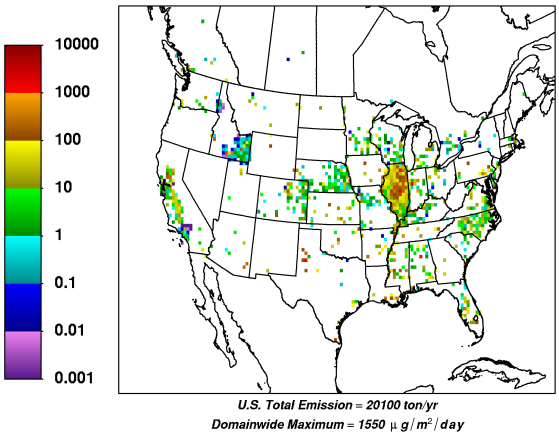



(e) Wood Products - Drying

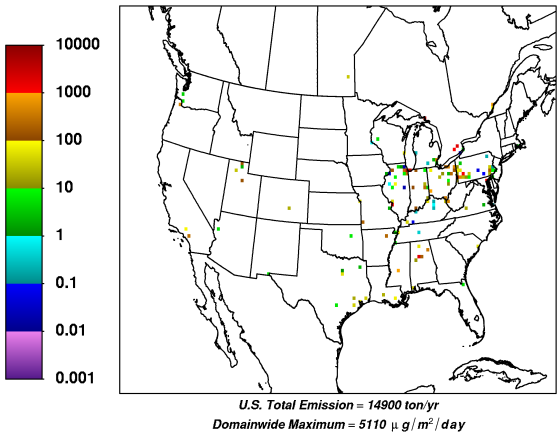



(f) Surface Coating

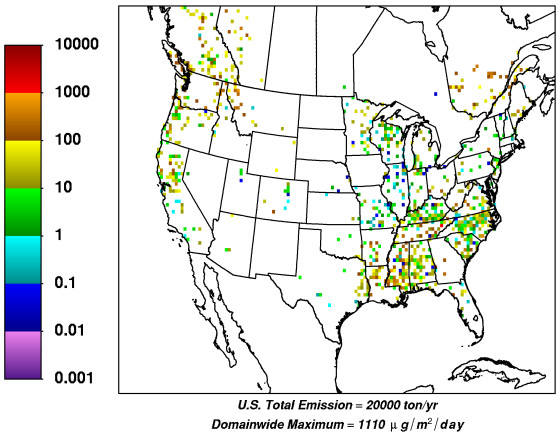
**Figure S14.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu$g/m$^2$/year) by Source Category, continued.



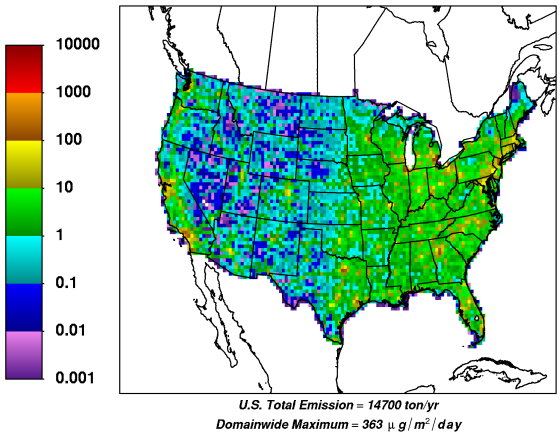

(a) Food & Ag Handling



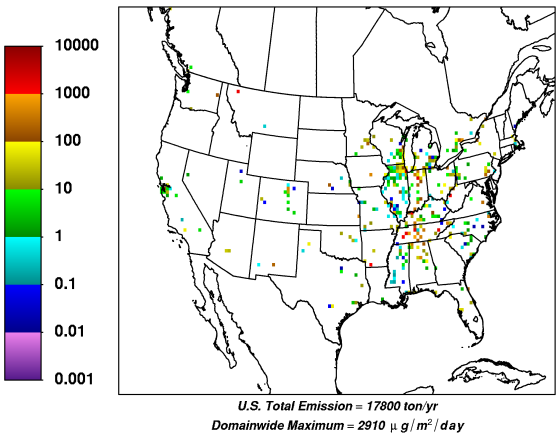

(d) Open Hearth Furnace



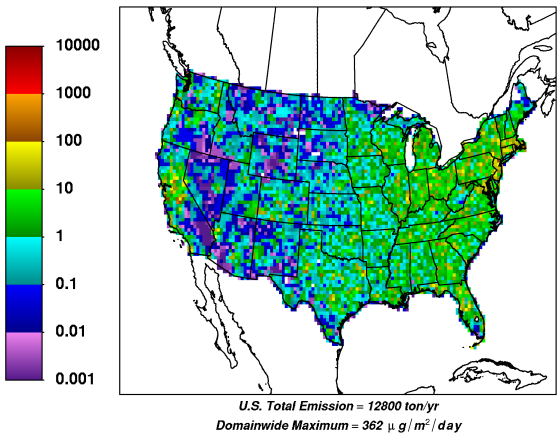

(b) Wood Products - Sawing




(e) Brake Lining Dust

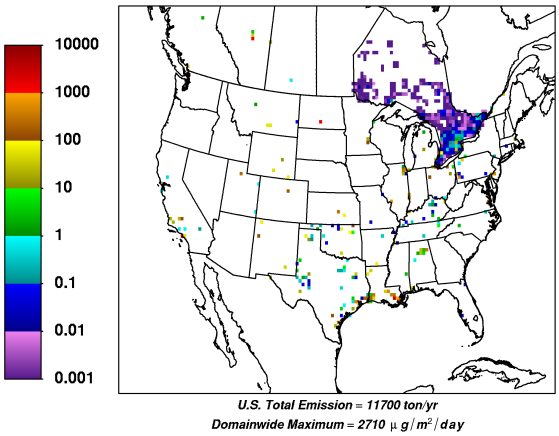



(c) Aluminum Processing

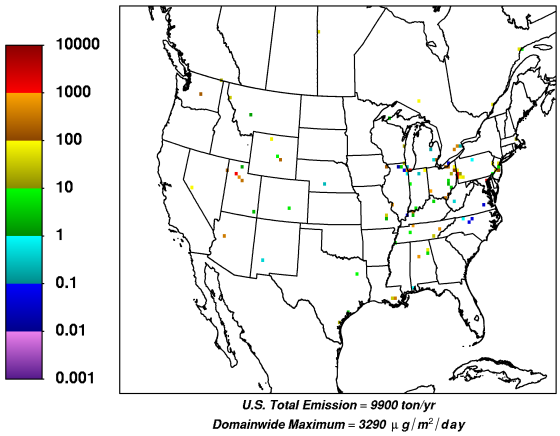



(f) Meat Frying

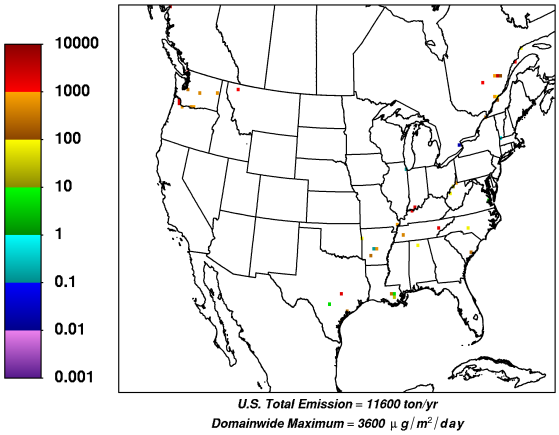
**Figure S15.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



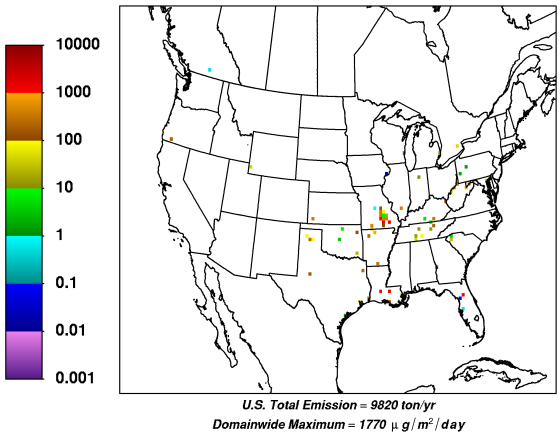

(a) Process Gas Combustion



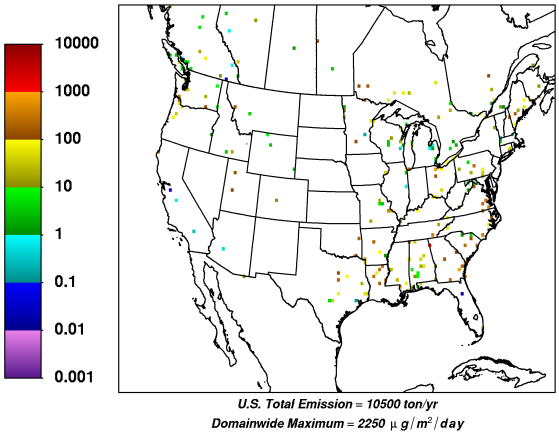

(d) Sintering Furnace



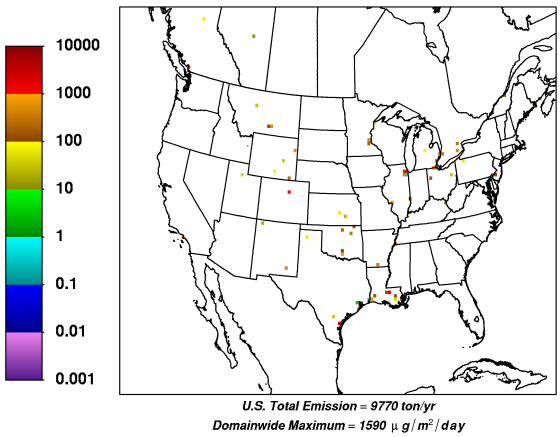

(b) Aluminum Production




(e) Charcoal Manufacturing

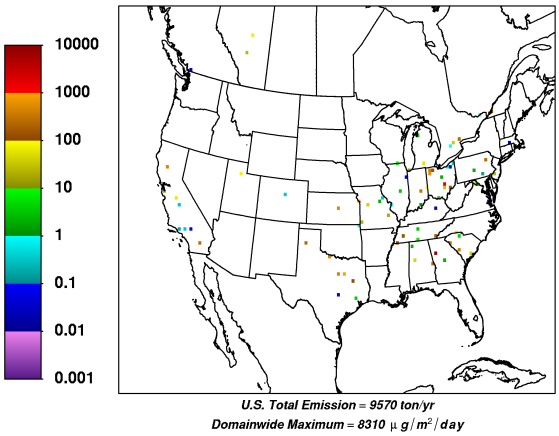



(c) Lime Kiln

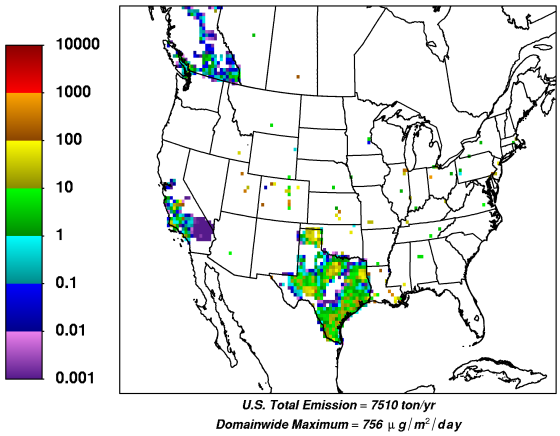



(f) Catalytic Cracking

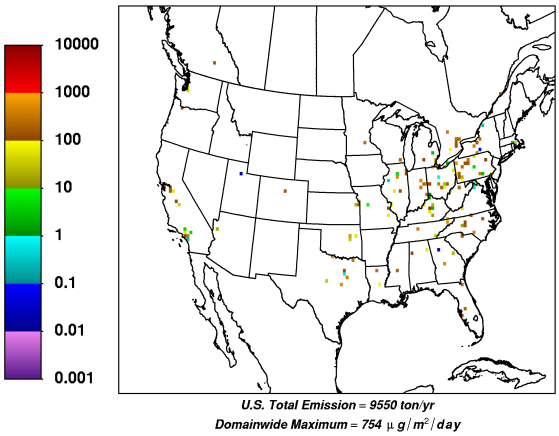
**Figure S16.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



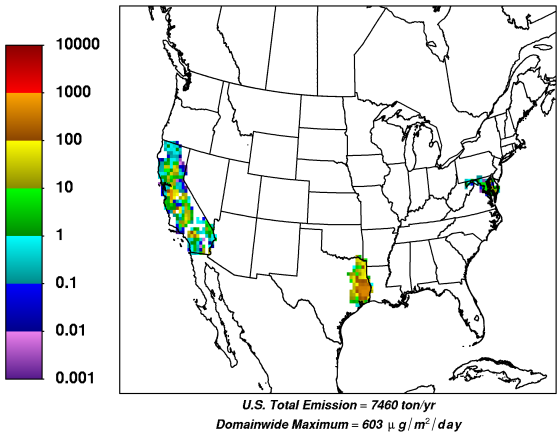

(a) Fiberglass Manufacturing



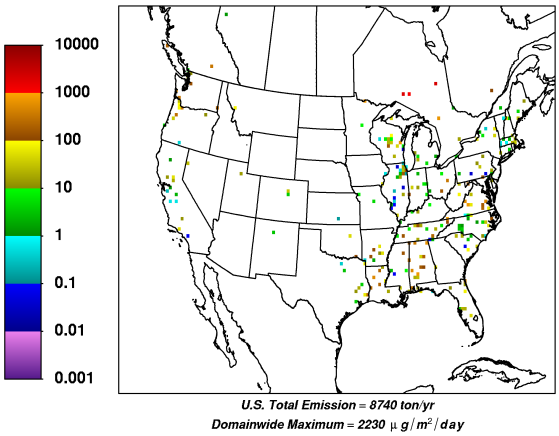

(d) Petroleum Industry - Avg



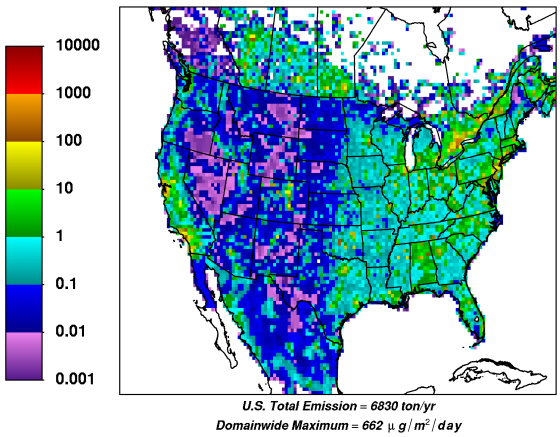

(b) Glass Furnace




(e) Slash Burning

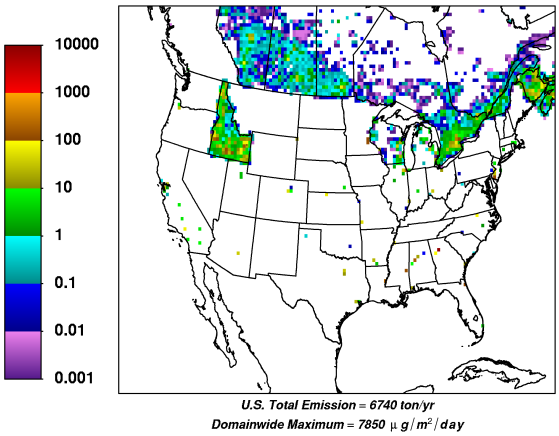



(c) Pulp & Paper Mills

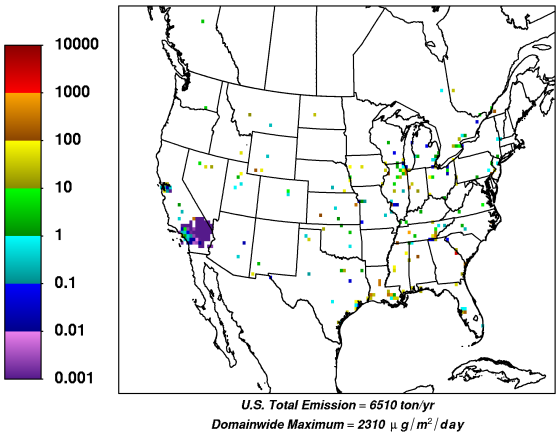



(f) Misc. Sources

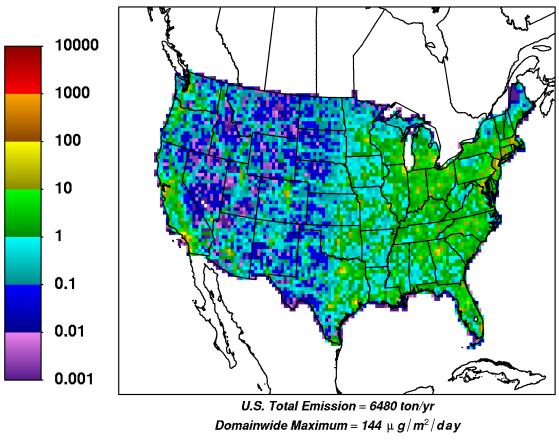
**Figure S17.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



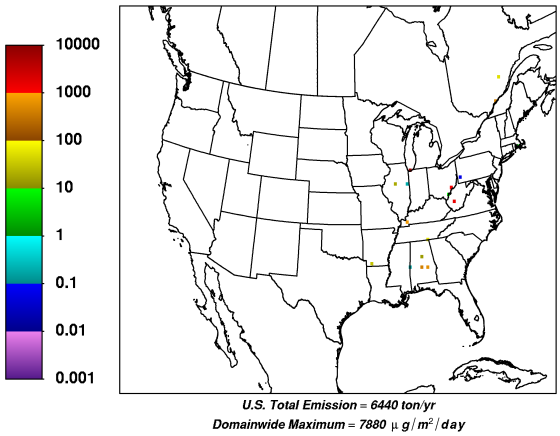

(a) Asphalt Roofing



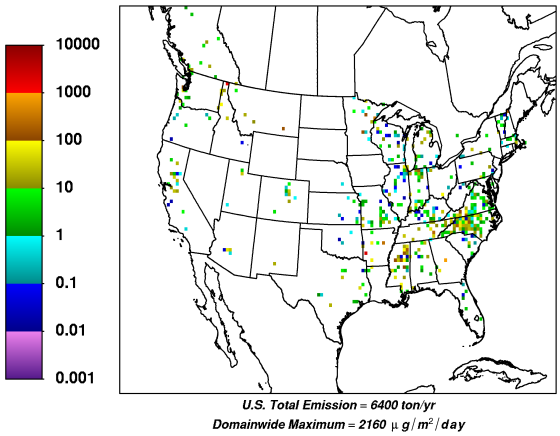

(b) Inorganic Chemical Manufacturing



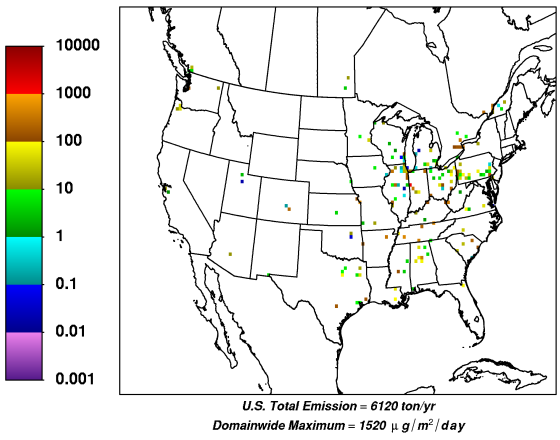

(c) Tire Dust




(d) Ferromanganese Furnace

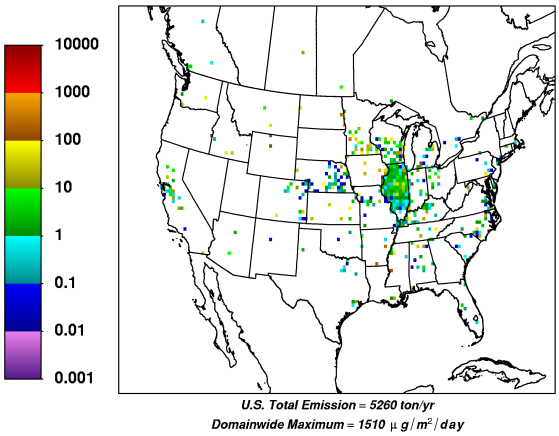



(e) Wood Products - Sanding

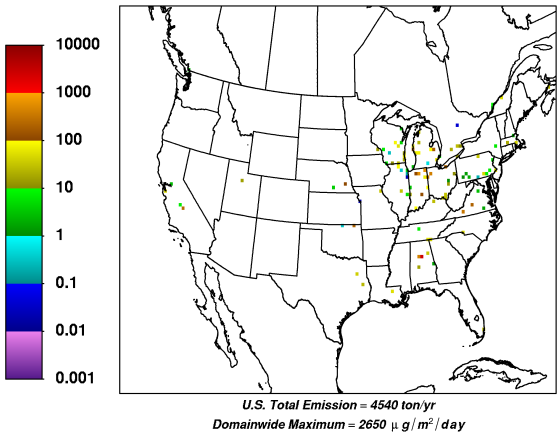



(f) Electric Arc Furnace

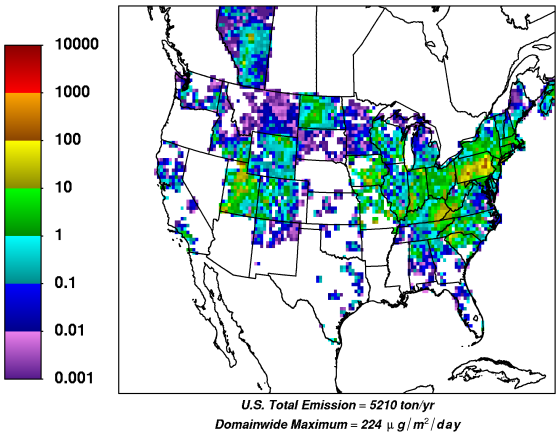
**Figure S18.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



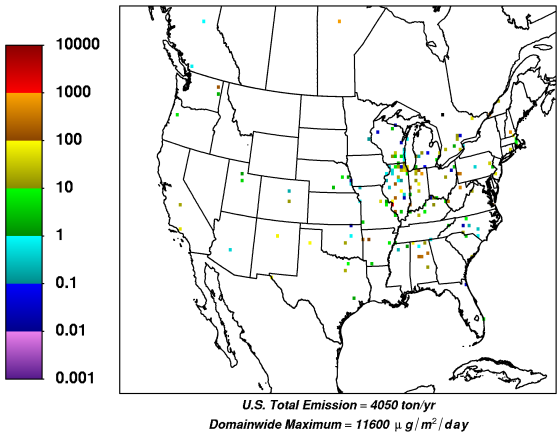

(a) Food & Ag Drying



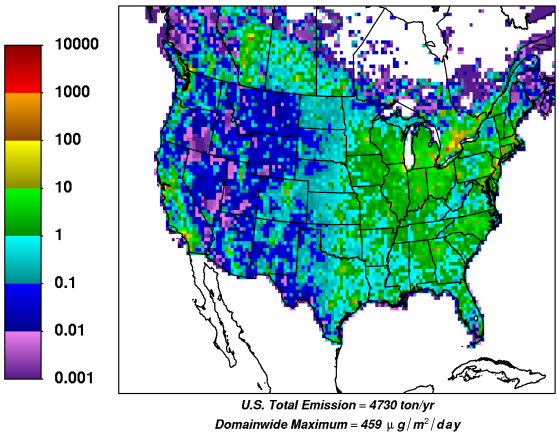

(d) Cast Iron Cupola



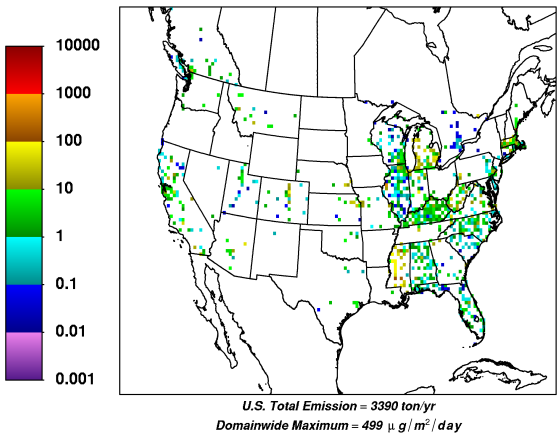

(b) Residential Coal Combustion




(e) Copper Processing

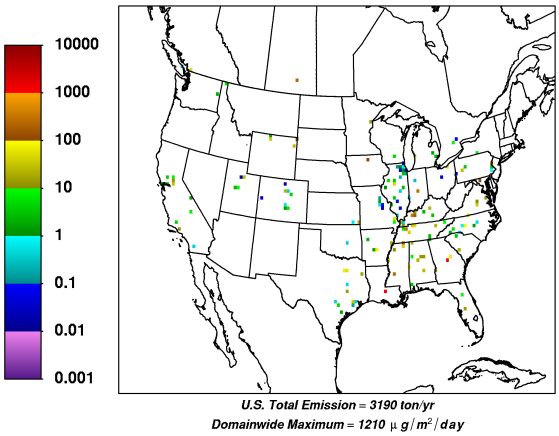



(c) Residential Natural Gas Combustion

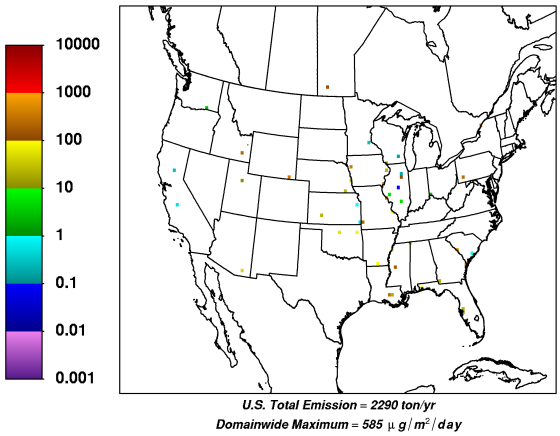



(f) Asphalt Manufacturing

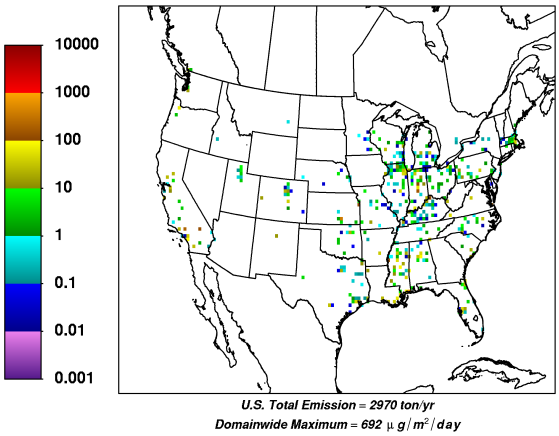
**Figure S19.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



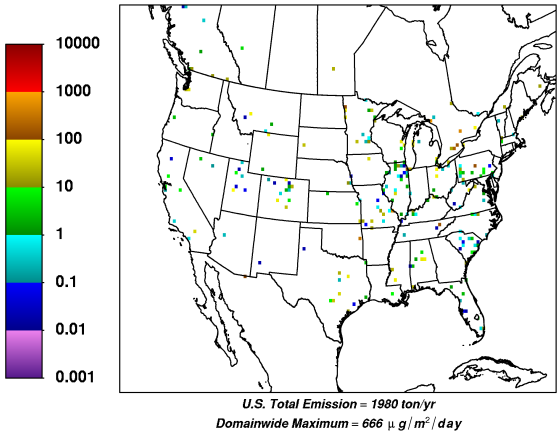

(a) Fly Ash



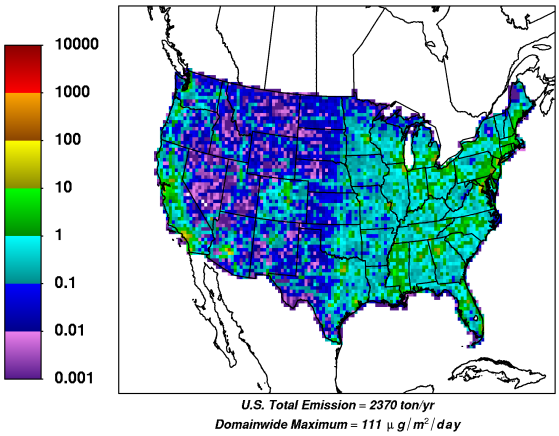

(d) Ammonium Nitrate Production



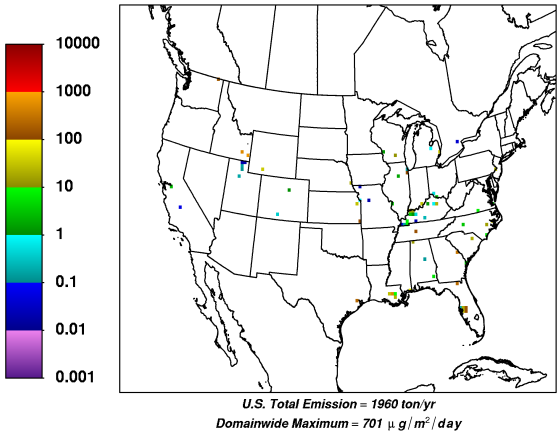

(b) Sandblast




(e) Limestone Dust

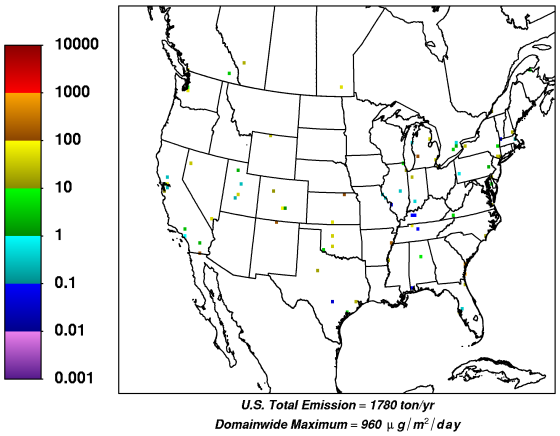



(c) LDDV Exhaust

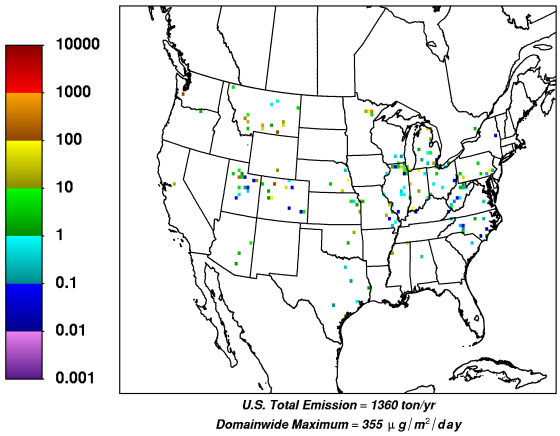



(f) Phosphate Manuf.

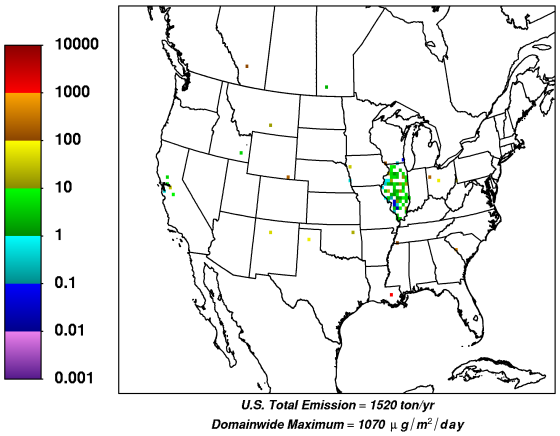
**Figure S20.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



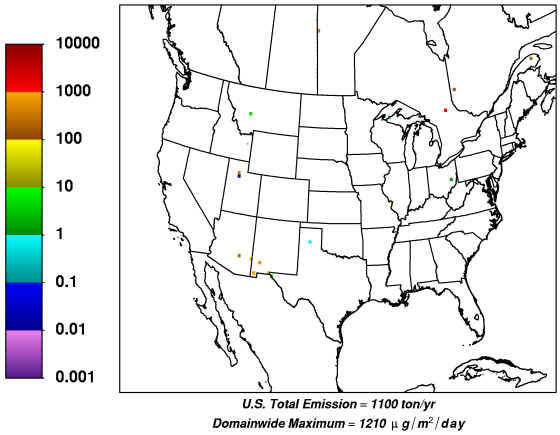

(a) Gypsum Manufacturing



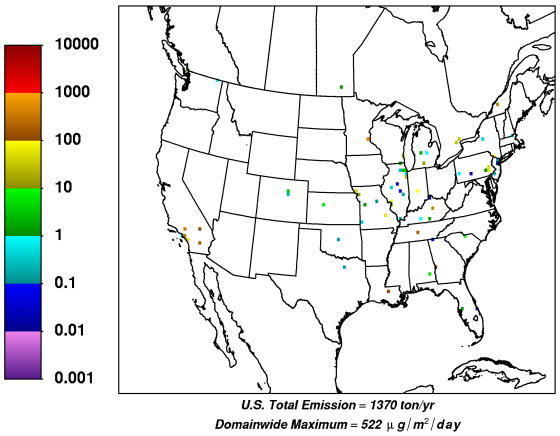

(d) Crustal Material



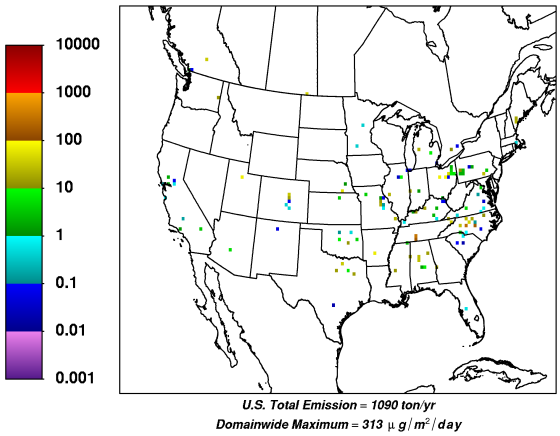

(b) Urea Fertilizer




(e) Copper Production

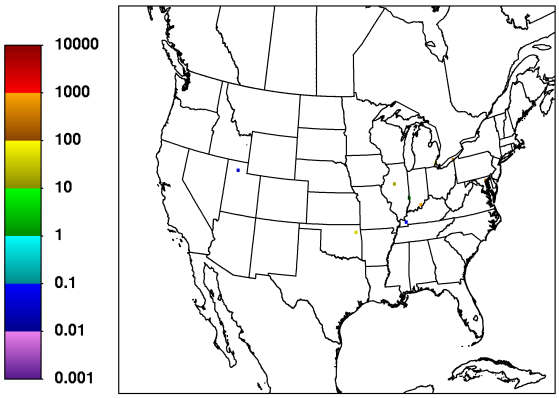



(c) Lead Processing

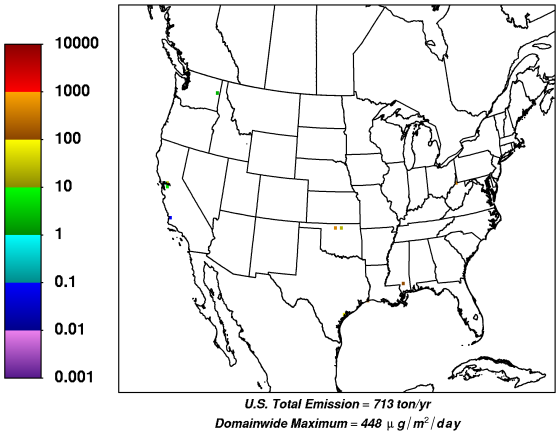



(f) Brick Grinding & Screening

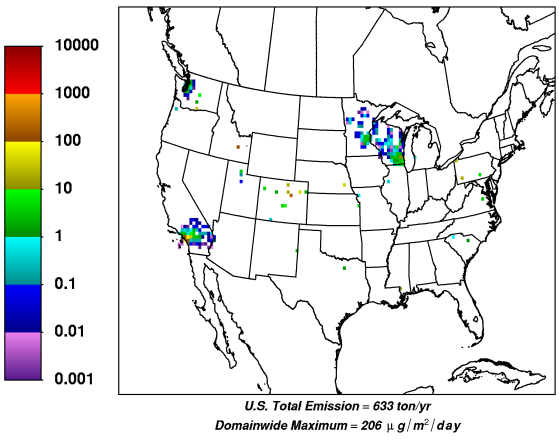
**Figure S21.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu g/m^2$/year) by Source Category, continued.



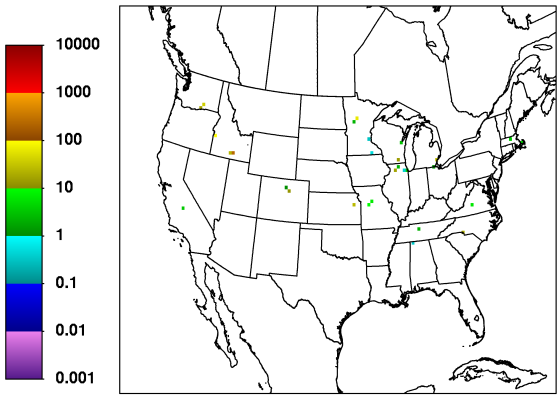

(a) Calcium Carbide Furnace



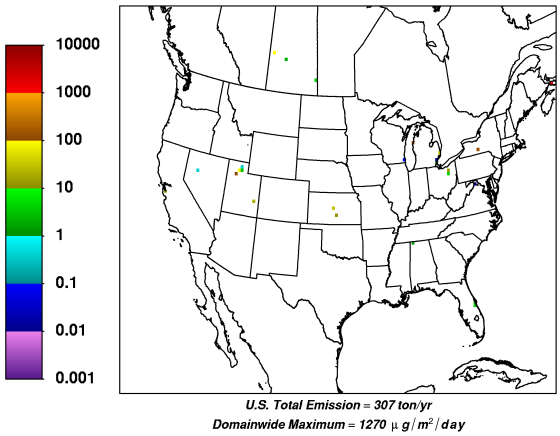

(b) Coke Calciner



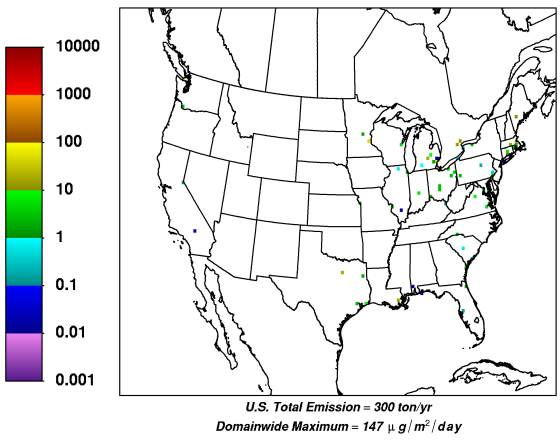

(c) Industrial Soil




(d) Potato Deep Frying

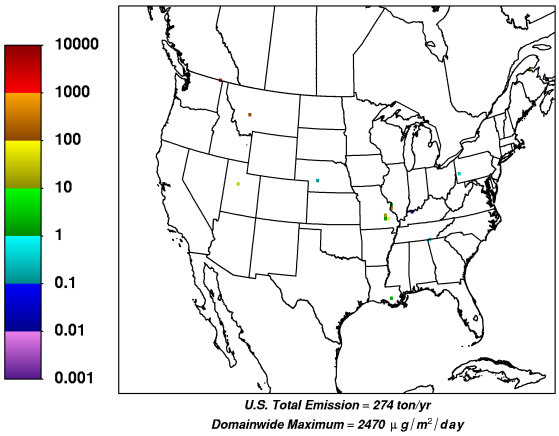



(e) Sea Salt

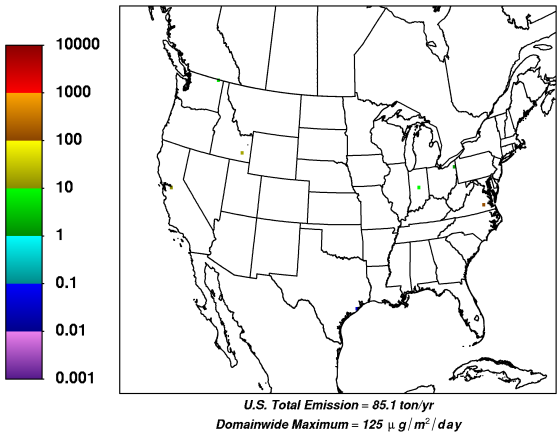



(f) Sludge Combustion

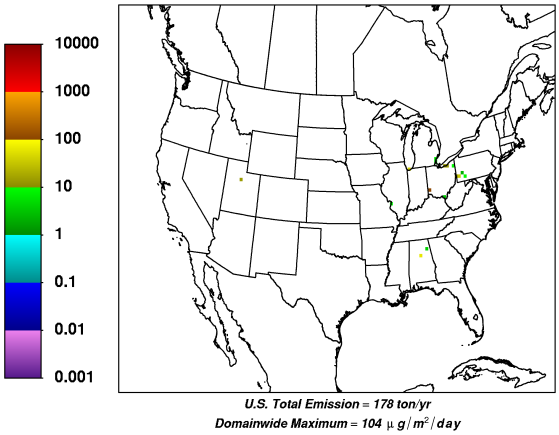
**Figure S22.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu$g/m$^2$/year) by Source Category, continued.



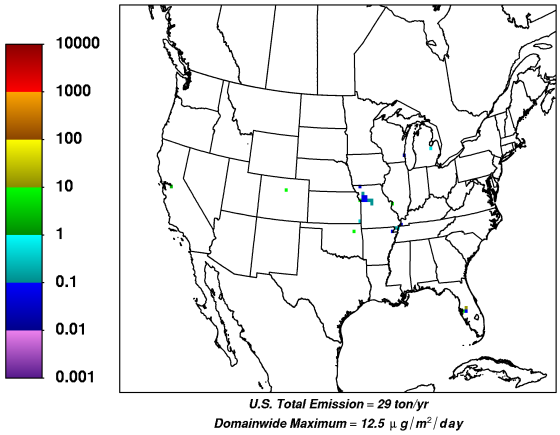

(a) Lead Production



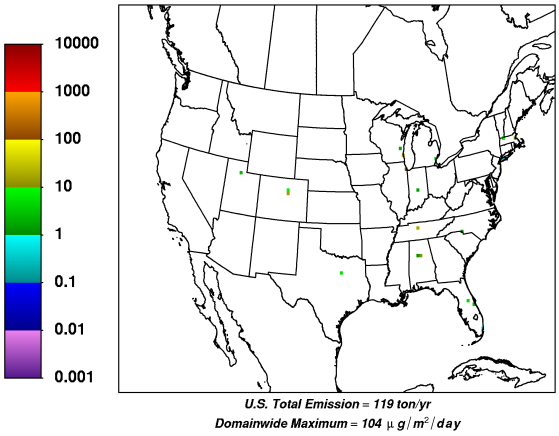

(d) Ammonium Sulfate Production



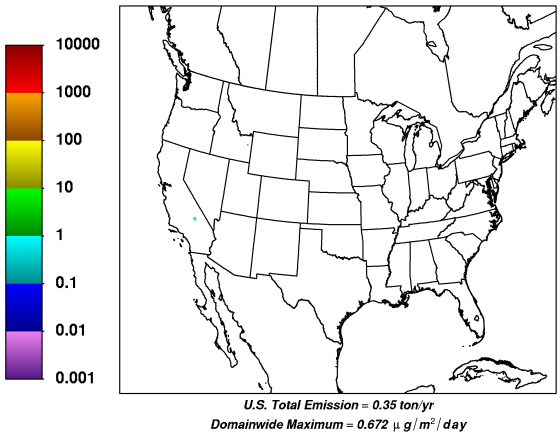

(b) Steel Desulfurization




(e) Inorganic Fertilizer




(c) Auto Body Shredding




(f) Boric Acid Manufacturing

**Figure S23.** Spatial Plots of $PM_{2.5}$ Emissions ($\mu$g/m$^2$/year) by Source Category, continued.

## Literature Cited


(1) Bhave, P. V.; Pouliot, G. A.; Zheng, M. Diagnostic model evaluation for carbonaceous $PM_{2.5}$ using organic markers measured in the southeastern U.S. *Environ. Sci. Technol.* **2007**, *41*, 1577–1583.

(2) EPA, *Source Sampling Fine Particulate Matter: Institutional Oil-Fired Boiler*; Technical Report EPA-600/R-07-005, 2007.

(3) Hays, M.; Beck, L.; Barfield, P.; Willis, R.; Landis, M.; Stevens, R.; Preston, W.; Dong, Y. Physical and chemical characterization of residual oil-fired power plant emissions. *Aerosol Sci. Technol.* **2009**, In Review.

(4) Chow, J.; Watson, J.; Crow, D.; Lowenthal, D.; Merrifield, T. Comparison of IMPROVE and NIOSH carbon measurements. *Aerosol Sci. Technol.* **2001**, *34*, 23–34.

(5) Chow, J.; Watson, J.; Pritchett, L.; Pierson, W.; Frazier, C.; Purcell, R. The DRI thermal/optical reflectance carbon analysis system: description, evaluation and application in US air quality studies. *Atmos. Environ.* **1993**, *27A*, 1185–1201.

(6) EPA, *Mercury Study Report to Congress*; Technical Report, 1997.

(7) Seigneur, C.; Abeck, H.; Chia, G.; Reinhard, M.; Bloom, N. S.; Prestbo, E.; Saxena, P. Mercury adsorption to elemental carbon (soot) particles and atmospheric particulate matter. *Atmos. Environ.* **1998**, *32*, 2649–2657.

(8) *Code of Federal Regulations, Title 40, chapter I, Part 51*; 1997.

(9) Hildemann, L.; Markowski, G.; Cass, G. Chemical composition of emissions from urban sources of fine organic aerosol. *Environ. Sci. Technol.* **1991**, 744–759.

(10) Hays, M.; Geron, C.; Linna, K.; Smith, N. Speciation of gas-phase and fine particle emissions from burning of foliar fuels. *Environ. Sci. Technol.* **2002**, *36*, 2281–2295.

(11) EPA, *Air Quality Criteria for Lead (Final)*; Technical Report EPA/600/R-05/144aF-bF, 2006.

(12) Frank, N. Retained nitrate, hydrated sulfates, and carbonaceous mass in Federal Reference Method fine particulate matter for six eastern US cities. *J. Air Waste Manage. Assoc.* **2006**, *56*, 500–511.

(13) Greenwood, N. N.; Earnshaw, A. *Chemistry of the Elements*; Oxford:Butterworth-Heinemann: Oxford, 1997.

(14) Malm, W.; Sisler, J. F.; Huffman, D.; Eldred, R. A.; Cahill, T. A. Spatial and seasonal trends in particle concentration and optical extinction in the United States. *J. Geophys. Res.* **1994**, *99*, 1347–1370.

(15) Kleeman, M. J.; Schauer, J. J.; Cass, G. R. Size and composition of fine particulate matter emitted from motor vehicles. *Environ. Sci. Technol.* **2000**, *34*, 1132–1142.

(16) Stelson, A. W.; Seinfeld, J. H. Chemical mass accounting of urban aerosol. *Environ. Sci. Technol.* **1981**, *15*, 671–679.

(17) Millero, F. J. *Chemical Oceanography*; CRC Press: Boca Raton, FL, 1996.

(18) Aiken, A. C.; Decarlo, P. F.; Kroll, J. H.; Worsnop, D. R.; Huffman, J. A.; Docherty, K. S.; Ulbrich, I. M.; Mohr, C.; Kimmel, J. R.; Sueper, D.; Sun, Y.; Zhang, Q.; Trimborn, A.; Northway, M.; Ziemann, P. J.; Canagaratna, M. R.; Onasch, T. B.; Alfarra, M. R.; Prevot, A. S.; Dommen, J.; Duplissy, J.; Metzger, A.; Baltensperger, U.; Jimenez, J. L. O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry. *Environ. Sci. Technol.* **2008**, *42*, 4478–4485.

(19) Lipsky, E. M.; Robinson, A. L. Effects of dilution on fine particle mass and partitioning of semivolatile organics in diesel exhaust and wood smoke. *Environ. Sci. Technol.* **2006**, *40*, 155–162.

(20) Russell, L. M. Aerosol Organic-Mass-to-Organic-Carbon Ratio Measurements. *Environ. Sci. Technol.* **2003**, *37*, 2982–2987.

(21) Japar, S. M.; Szkarlat, A. C.; Gorse Jr., R. A.; Heyerdahl, E. K.; Johnson, R. L.; Rau, J. A.; Huntzicker, J. J. Comparison of solvent extraction and thermal-optical carbon analysis methods: Application to diesel vehicle exhaust aerosol. *Environ. Sci. Technol.* **1984**, *18*, 231–234.

(22) Sheesley, R. J.; Schauer, J. J.; Chowdhury, Z.; Cass, G. R.; Simoneit, B. R. Characterization of organic aerosols emitted from the combustion of biomass indigenous to South Asia. *J. Geophys. Res.* **2003**, *108*, Article number 4285.

(23) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. Measurement of Emissions from Air Pollution Sources 2. C1 through C30 Organic Compounds from Medium Duty Diesel Trucks. *Environ. Sci. Technol.* **1999b**, *33*, 1578–1587.

(24) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. Measurement of Emissions from Air Pollution Sources 5. C1 through C32 Organic Compounds from Gasoline-Powered Motor Vehicles. *Environ. Sci. Technol.* **2002**, *36*, 1169–1180.

(25) Turpin, B.; Lim, H. Species contributions to $PM_{2.5}$ mass concentrations: Revisiting common assumptions for estimating organic mass. *Aerosol Sci. Technol.* **2001**, *35*, 602–610.

(26) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. Measurement of Emissions from Air Pollution Sources 3. C1 through C29 Organic Compounds from Fireplace Combustion of Wood. *Environ. Sci. Technol.* **2001**, *35*, 1716–1728.

(27) Bae, M.; Schauer, J.; Turner, J. Estimation of the monthly average ratios of organic mass to organic carbon for fine particulate matter at an urban site. *Aerosol Sci. Technol.* **2006**, 1123–1139.

(28) Jimenez, J. R.; Claiborn, C. S.; Dhammapala, R. S.; Simpson, C. D. Methoxyphenols and

levoglucosan ratios in $PM_{2.5}$ from wheat and Kentucky bluegrass stubble burning in eastern Washington and northern Idaho. *Environ. Sci. Technol.* **2007**, *41*, 7824–7829.

(29) Herner, J.; Green, P.; Kleeman, M. Measuring the trace elemental composition of size-resolved airborne particles. *Environ. Sci. Technol.* **2006**, *40*, 1925–1933.

(30) R Development Core Team, *R: A Language and Environment for Statistical Computing*; R Foundation for Statistical Computing: Vienna, Austria, 2007, ISBN 3-900051-07-0.