

Emissions Inventory of PM_{2.5} Trace Elements across the United States

Adam Reff,* Prakash V. Bhave, George A. Pouliot, Thompson G. Pace, J. David
Mobley, and Marc Houyoux

U. S. Environmental Protection Agency, Research Triangle Park, NC 27711

E-mail: reff.adam@epa.gov

Abstract

This paper presents the methods and results of speciating the EPA 2001 PM_{2.5} National Emissions Inventory (NEI) using source profiles from the SPECIATE v4.0 database to create a national inventory of the full suite of PM_{2.5} trace elements (atomic number > 10) that are measured at national ambient monitoring sites. PM_{2.5} emissions in the NEI were organized and aggregated into a set of 85 source categories for which speciation profiles were available. Nationally, the largest PM_{2.5} emissions were found to originate from the Unpaved Road Dust and Agricultural Soil source categories. SPECIATE data for metals classified as Hazardous Air Pollutants (HAP) were updated using data from a recent HAP NEI. All emissions were spatially gridded and U.S. emissions maps for a number of species (e.g. Si, Cu, Sr) are presented for what we believe is the first time. Nationally, the trace elements emitted in the highest quantities are silicon (3.8×10^5 ton/year), aluminum (1.4×10^5 ton/year), and calcium (1.3×10^5 ton/year). The new chemical characterization of the PM_{2.5} inventory shows that most of the

previously un-speciated emissions are comprised of crustal elements and metal-bound oxygen. This work also reveals that there are insufficient data available in SPECIATE for speciating the emissions from some major PM_{2.5} source categories such as chemical manufacturing processes and cooling towers.

Introduction

Trace elements constitute only a small fraction of the mass of ambient fine particulate matter (PM_{2.5}), but their concentrations are routinely measured at hundreds of air monitoring sites across the United States (1). These data have proven useful in calculations of visibility degradation (2), receptor-based source-apportionment calculations (3), and are likely to be used as a basis for studying the health effects of exposure to metals (4, 5). While the ambient concentrations have been analyzed extensively, emission inventories of PM_{2.5} trace elements have received relatively little attention.

Inventories of particulate emissions contain information spanning as many as five dimensions: spatial, temporal, source-categorical, particle-size, and chemical-composition. A perusal of the literature shows that the resolutions and ranges over which each of these dimensions are spanned varies widely depending on the intended purpose of the inventory. For example, soil-dust and elemental emissions have been inventoried for specific regions of China (6, 7), and metals from the industrial sector have been inventoried in Mumbai, India (8). In southern California, comprehensive inventories of trace elements in PM₁₀ and ultrafine PM have been developed for select time periods (9, 10). In the U.S., nationwide inventories of lead from all major anthropogenic sources have been assembled since 1970 (11). Nationwide emissions data have been compiled for PM_{2.5} and numerous hazardous air pollutants (HAP), which includes ten toxic metals, since 1990

(12, 13).

The U.S. National Emission Inventory (NEI) for PM_{2.5} is extremely detailed in the source-categorical and spatial dimensions. Anthropogenic emissions are segregated into 3780 distinct source classes. In the spatial dimension, point-source locations are specified by latitude and longitude, mobile- and area-source emissions are tabulated for each county ($N = 3044$). In the chemical-composition dimension, however, the PM_{2.5} NEI is speciated for modeling using §simplified profiles that include only sulfate (SO₄), nitrate (NO₃), elemental carbon (EC), organic carbon (OC), and other mass (14). National-scale inventories of PM_{2.5} metals have been assembled such as the HAP NEI (<http://www.epa.gov/ttn/chief/net/2002inventory.html>) and the 1985 National Acid Precipitation Assessment Program (NAPAP) Emissions Inventory (15), but a national inventory that contains the full suite of PM_{2.5} species measured routinely at U.S. monitoring sites has yet to be assembled. Without such an inventory, air quality models cannot be used to predict ambient concentrations and deposition patterns of that suite of species across the country.

In this work, we identify a set of source categories for which PM_{2.5} speciation profiles are available and into which all major emission sources in the NEI can be categorized. We survey speciation profiles from the literature that contain the bulk species listed above plus up to 38 trace elements to develop a comprehensive speciation profile that is representative of emissions from each source category. We demonstrate the utility of these profiles by speciating the NEI PM_{2.5}, thus constructing the first national-scale emissions inventory to contain the full suite of PM_{2.5} species measured across the major air monitoring networks such as the Interagency Monitoring of Protected Visual Environments (IMPROVE) network (16) and the Chemical Speciation Network (17). The inventory is spatially gridded, and maps of selected species and source categories are displayed and analyzed. Similar approaches to emissions inventory development have recently

been performed for volatile organic compounds (18) and polycyclic aromatic hydrocarbons (19); this is the first known demonstration of speciating the trace elements in a national PM_{2.5} inventory.

Methods

The general strategy for calculating trace elemental emissions is to multiply each source of PM_{2.5} emissions in the 2001 NEI by a speciation profile appropriate to the emission source type. A flow diagram of the methods is provided in Figure Figure 1. The procedures followed to generate and process the emissions and speciation data are summarized below, with additional details in the Supporting Information.

U.S. EPA National Emissions Inventory

PM_{2.5} mass emissions were obtained from the 2001 Clean Air Interstate Rule (CAIR) version of the NEI (<http://www.epa.gov/air/interstateairquality/technical.html>). That inventory consists of PM_{2.5} emissions from eight sectors: 1) onroad mobile sources, 2) non-road mobile sources, 3) fires, 4) point and 5) area sources of fugitive dust, 6) electric generating units (EGUs), 7) non-EGU point sources, and 8) other area sources. Both point and area sources of fugitive dust emissions are reduced by a county-specific factor – ranging from 3% in barren areas to 95% in forested areas – to account for removal processes within several hundred meters of dust sources (20). PM_{2.5} emissions within the inventory are labeled using a set of 3780 source classification codes (SCCs) that are defined based on the emission generating processes and source characteristics.

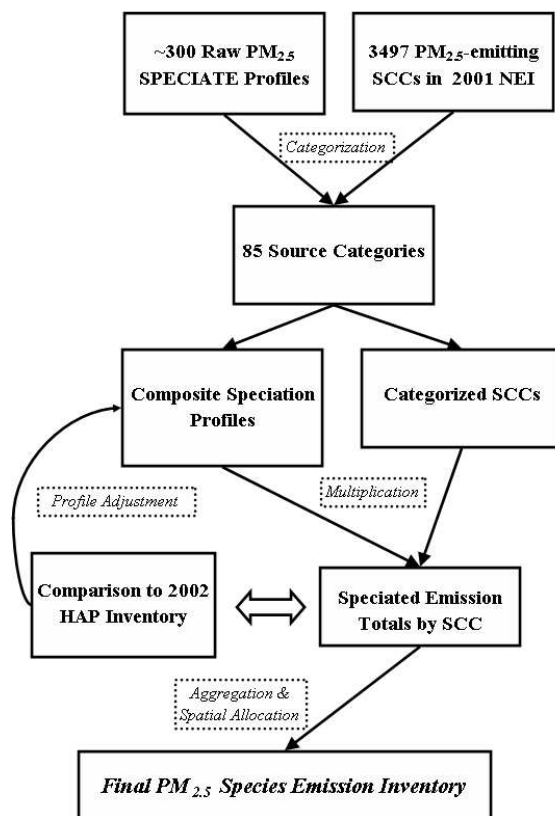


Figure 1: Schematic diagram illustrating the procedure followed to create the PM_{2.5} trace elements emissions inventory.

Source Categories

To compute emissions of each trace element, PM_{2.5} mass emissions from the NEI were multiplied by source-specific speciation profiles. The SPECIATE database (<http://www.epa.gov/ttn/chief/software/speciate/>) is currently the most comprehensive collection available of such profiles known, containing over 1000 PM_{2.5} speciation profiles from the literature (Hsu et al., 2006). Most of these profiles consist of trace elements measured by X-ray Fluorescence (XRF), OC and EC measured by thermal optical methods, and inorganic ions measured by ion chromatography, each normalized by PM_{2.5} mass. A detailed analysis of the database revealed

that about 300 of the profiles are suitable for speciating PM_{2.5} emission sources in the NEI. The remaining profiles could not be used for a number of reasons as described in the Supporting Information. A 1-to-1 mapping between these profiles and the 3497 SCCs in the NEI with non-zero PM_{2.5} emissions was judged to be implausible due to the varying degrees of source specificity, the absence of appropriate PM_{2.5} profiles for certain SCCs, and the presence of multiple speciation profiles in SPECIATE v4.0 for certain sources. A list of 85 *source categories* was thus developed for classifying SCCs and SPECIATE source profiles on a common basis. Detailed steps that were followed to create the source categories can be found in the Supporting Information.

Composite Speciation Profiles

After classifying the emissions into the source categories, the next step was to create a speciation profile for each category by either identifying an existing profile or constructing one using profiles in SPECIATE v4.0 that contain PM_{2.5} mass fractions of trace elements, inorganic ions, and carbon species. In cases where multiple high-quality profiles were available for a single source category, profiles were averaged together to create a composite speciation profile. The raw profiles used for each composite are listed in a Notes field within the SPECIATE database. The general procedure for creating composite profiles was to compute the median of SPECIATE profiles that were classified into each 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. However, a number of profile- and source category-specific adjustments were found to be necessary both before and after the averaging step that were based on the data quality and profile information in either SPECIATE or the references from which the profile originated. For example, sulfate and elemental sulfur were forced to be stoichiometrically equal (assuming all sulfur existed in the form of SO₄²⁻) in profiles

where measurements of only one of the two species were available. Other case-specific issues include the treatment of OC and EC data to account for different analytical methods that generated the data in SPECIATE, and treatment of motor vehicle emissions that needed to be disaggregated into brake dust, tire dust, and exhaust fractions. The resulting composite profiles were reviewed by a large number of emissions and source-testing specialists from various Offices within the EPA, after which they were included in the public release of SPECIATE v4.0. However, further analyses of the composite profiles after the public release led to some minor differences between the composite profiles used in this paper and those in SPECIATE v4.0. Detailed descriptions of these differences and the case-specific calculations mentioned above are provided in the Supporting Information. The full set of composite profiles used in the study will be included in a future public version of the SPECIATE database.

Emission Speciation

After classifying both SCCs and composite profiles into the 85 source categories, the NEI was speciated by multiplying the each source category's PM_{2.5} emission against composite profile's set of species-specific mass fractions. This provided a set of PM_{2.5} species emissions for each SCC. As indicated in Figure Figure 1, a subset of these results were compared against emissions from another inventory to evaluate the validity of some species' mass fractions in the composite profiles. Profiles were adjusted as described in the next section, and emissions were re-calculated. This cycle iterated a number of times to ensure quality in the resulting inventory.

Profile Updates Using HAP NEI

Many of the profiles in SPECIATE were measured during the 1980s and 1990s, and so are expected to be outdated with regards to the composition of hazardous air pollutants (HAPs)

since control technologies and strategies have been developed more recently to specifically meet federally-mandated limits on HAP emissions. Data from version 2 of the 2002 NEI for HAPs were therefore used to identify potentially outdated mass fractions of Cr, Mn, Co, Ni, Se, Cd, Sb, Hg, and Pb in the composite speciation profile of each source category. Although the year of the HAP inventory (2002) differs from the year of the speciated PM_{2.5} inventory (2001, which was selected due to having been processed into the SMOKE modeling system described below at the time of this analysis), we felt that the years were sufficiently close for the qualitative evaluation that we performed. To perform the evaluation, 2002 emissions of the 9 hazardous elements were aggregated into the same 85 source categories that were used to develop our trace-elemental inventory. Differences between the two inventories were then calculated for each combination of 9 elements and 85 source categories. Differences greater than 20% of the E_{EI} totals were used to identify composite speciation profiles needing adjustment. Detailed discussion of these calculations, results and the subsequent adjustments made to the speciation profiles are provided in the Supporting Information.

Spatial Allocation

The Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system (21) was used in this work to process the inventory, which included distributing both the pre- and post-speciated emissions to a 36-km grid and using the smkreport diagnostic utility to generate text reports of 2001 annual total PM_{2.5} emissions by SCC for the continental U.S..

Results

Speciated PM_{2.5} Emissions

Figure Figure 2 depicts the emissions of all PM_{2.5} species from each of the 85 source categories, summed across the U.S. during 2001. Source categories are sorted based on their total emissions of PM_{2.5} mass, with the highest-emitting categories shown at the top. Those categories are Unpaved Road Dust (6.9×10^5 ton/year), Agricultural Soil (6.1×10^5 ton/year) and Wildfires (4.8×10^5 ton/year). The ranking can differ greatly at individual locations within the U.S. due to the different mixture of sources that dominate urban airsheds as compared to the regional background. The columns of Figure Figure 2 show emissions of PM_{2.5} mass, OC, EC, major ions, and 37 trace elements sorted by atomic number. In general, the magnitude of trace-elemental emissions tends to decrease with increasing atomic mass.

Unlike source apportionment analyses, in which the PM_{2.5} contributions from five to ten different source categories are typically resolved, the source specificity of the NEI and the composite speciation profiles allows one to compute emission contributions from a much larger number of sources. When similar source categories are aggregated together, a number of features in Figure Figure 2 are found to be consistent with results commonly reported in the PM_{2.5} source apportionment literature.

1. The dominant source of crustal elements such as Mg, Al, Si, Ca, and Fe is fugitive dust. As shown in Figure Figure 2, the major sources of fugitive dust in the NEI are unpaved and paved roads, agricultural soil, construction dust, and sand/gravel from mining and quarrying operations. Natural windblown dust is likely to be a major source of crustal elements as well, but it is omitted from the NEI.

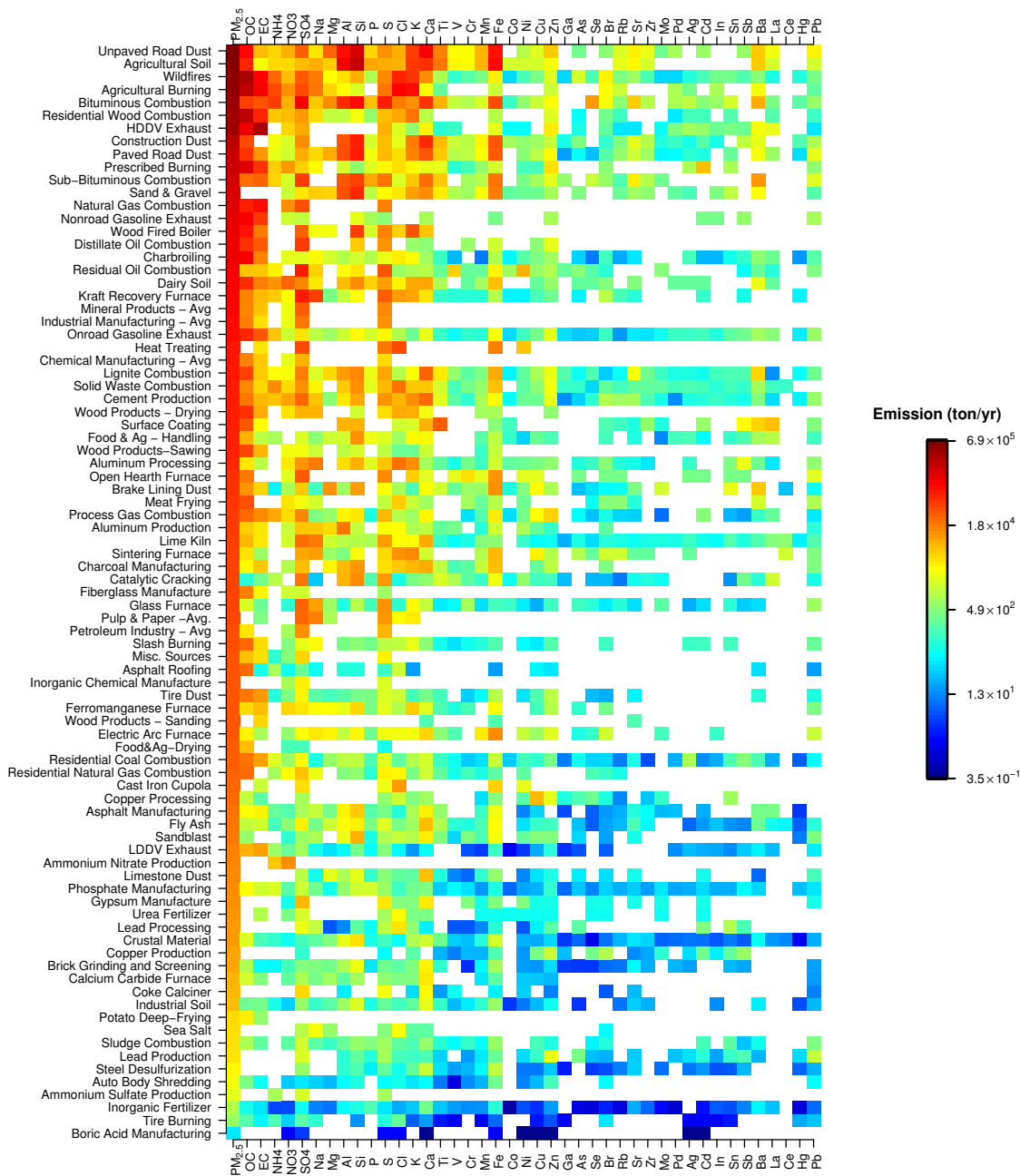


Figure 2: Continental U.S. national total emissions of PM_{2.5}, bulk species, and trace elements (x-axis) in each source category (y-axis), calculated by applying SPECIATE profiles to the 2001 NEI. White space means that the species mass fraction was either missing or 0 in the speciation profile.

2. Biomass burning makes a dominant contribution of particulate Cl, K, and primary OC. The major categories of biomass burning shown in Figure Figure 2 are wildfires, agricultural burning, residential wood combustion, prescribed burning for land management, and wood-fired boilers. During select time periods, the relative contributions from each of these categories may differ from the annual totals shown here (e.g., wildfire activity peaks in Summer, prescribed burning in Spring, and residential wood combustion in Winter), but a more detailed discussion of temporal trends is beyond the scope of this work.
3. Coal combustion is the largest source of S, Se, Sr, and primary SO₄. In our speciated inventory, these emissions are further segregated based on coal type, with bituminous coal producing the largest emissions (4.0×10^5), followed by subbituminous (1.1×10^5) and lignite coal (25,000 ton/yr). Residential coal combustion makes a relatively small contribution to the PM_{2.5} inventory (5200 ton/yr).
4. It is well known that diesel engines are the largest source of EC. Figure Figure 2 shows that they also emit large quantities of primary OC and Zn. Due to the dearth of speciation data from different types of diesel engines, the 'HDDV Exhaust' source category used in this work includes contributions from nonroad vehicles such as construction equipment, aircraft, marine vessels, and railroad locomotives, as well as onroad diesel vehicles.
5. Residual oil combustion is the largest source of V and Ni in the inventory. These elements have commonly been used as oil-combustion tracers in the source apportionment literature (22).
6. Barium has often been used as a tracer for dust from brake linings (23). While this is clearly a major source of Ba, our analysis reveals that coal combustion, agricultural soil, and unpaved road dust, emit much larger quantities of Ba on a national basis.

7. Industrial processing of copper is the largest source of Cu.

In addition to the features listed above, Figure Figure 2 displays some aspects of trace-elemental emissions that are not commonly reported in the literature. For example, surface coating operations emit the largest quantities of Ti, Sb, and La. While coal combustion is shown to be the largest source of Hg nationwide, wildfires also make a significant contribution (24) although a substantial portion of these emissions is expected to be re-emission of accumulated Hg depositions originating from anthropogenic sources. Unpaved road dust is the largest source of Pb on a national basis. This may be due to the resuspension of motor vehicle exhaust that deposited on road surfaces prior to banning of leaded fuel, though this has not been corroborated with ambient measurements (25). Fugitive dust emissions also make the greatest contributions to Mn and Zr.

The colorless portions of Figure Figure 2 provide a visual indication of data completeness in the final speciation profiles. Most of the source categories contain nonzero values for at least 21 out of the 37 elements considered here. Elements found most commonly in the profiles are S, K, Ca, and Fe, whereas the least common are Ga, Ce, and Hg. For the 12 highest-emitting PM_{2.5} source categories, composition data are available for at least 18 different elements. The most notable data gaps illuminated by Figure Figure 2 are in the source categories which contain “Avg” in their names (e.g., “Mineral Products – Avg” and “Industrial Manufacturing – Avg”). Speciation data used for these categories are from generic, historically based profiles that were mapped to a diversity of SCCs for which specific source testing data are not available in SPECIATE v4.0. Trace elements were removed from these profiles due to our lack of confidence in the applicability of these profiles to all the SCCs to which they are mapped. The major PM_{2.5} sources falling within these source categories are cooling towers, wastewater treatment, mineral production, pulp and paper production, petroleum production, and production processes of various chemicals such as

plastics, chlorine and derivatives, rubber, inks, and olefins. Future source characterization efforts could target emissions from these specific sources to fill in the data gaps.

Of course, the mere availability of trace-elemental data does not guarantee high quality. In particular, emissions of trace elements from industrial sources are a large source of uncertainty in this work because of difficulties that the XRF analysis has in measuring the small quantities of these elements that may be present in the source samples that provided measurements for SPECIATE. Applying source profiles collected using analytical techniques with lower detection limits (e.g., ICP-MS) should improve the quality of the inventory presented here (26).

The speciated breakdown of national total PM_{2.5} emissions is shown in Figure Figure 3. The first four bars on the left of Figure Figure 3 show the speciation of the NEI prior to applying profiles that include trace elements. The original speciation scheme included only sulfate, nitrate, OC, and EC, with all other species collected into the remaining portion labeled “PM_{Other}”, shown in the fifth bar of Figure Figure 3. Application of our composite profiles led to more detailed speciation of the PM_{Other} material, as indicated by the stacked layers inside the right-most bar and the accompanying pie chart in Figure Figure 3. Major species within PM_{Other} include Si, Al, Ca, Fe, K, and Cl, as well as organic material (hydrogen and oxygen bound to the carbon in the OC fraction) and oxygen bound to the elements (calculations of these are provided in the Supporting Information). Particle bound water was also calculated in this work as 24% of sulfate and ammonium emissions (27), and its contribution to national emissions is shown in the PM_{Other} bar in Figure Figure 3. Even after calculating particulate water, metallic oxygen, and organic material, most profiles used to speciate the inventory under-accounted for the total mass of PM_{2.5} emissions. The result of aggregating this “Unspeciated” fraction of PM_{2.5} mass is also indicated in the PM_{Other} bar in Figure Figure 3; on a national basis, this accounts for 20.5% of PM_{Other}, or 13% of PM_{2.5} emissions. We hypothesize

that this lack of mass closure of the PM_{2.5} species emissions is due to both analytical error in the SPECIATE profiles and errors inherent in the assumptions behind the calculations of the particulate water, organic material, and metallic oxygen.

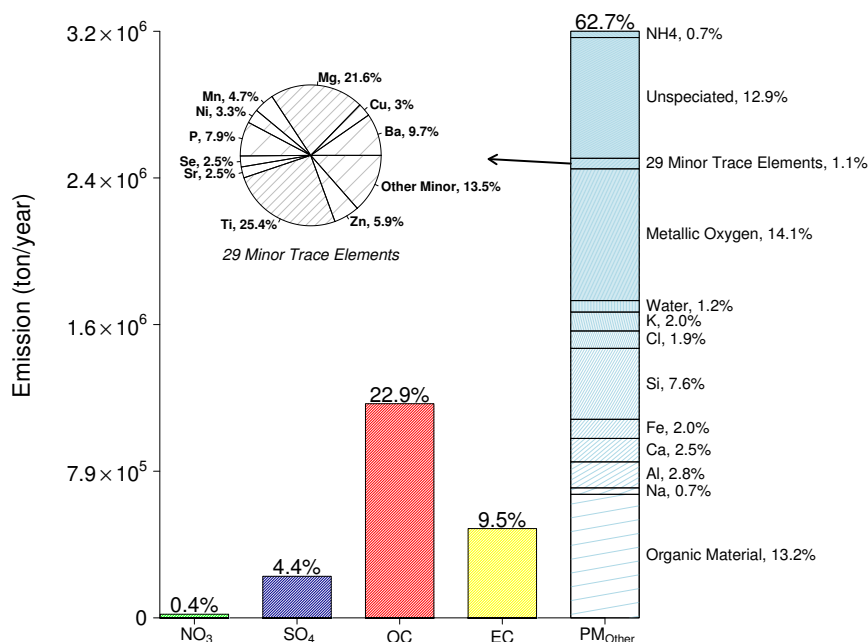


Figure 3: Chemical composition of the speciated PM_{2.5} NEI. The species on the x-axis are the chemical composition of emissions prior to this work; the stacked layers inside of the right-most bar show the contributions of the species previously grouped under “PM_{Other}” to national total PM_{2.5} emissions. The percentages atop each bar show the associated bulk species’ contribution to the U.S. annual total PM_{2.5} emission. The pie chart shows the percent breakdown of the minor trace elements too numerous to fit into the PM_{Other} bar; in the pie, the “Other Minor” slice contains trace elements that contribute to less than 2% of the minor elements’ emissions.

Spatial Patterns of PM_{2.5} Trace Element Emissions

Maps of emissions of PM_{2.5} from each source category and of each PM_{2.5} species were created by gridding emissions across the continental U.S. using the utility called smkmerge in the SMOKE modeling system and plotting the resulting output with the Package for Analysis and Visualization of Environmental data (PAVE) software (http://www.cep.unc.edu/empd/EDSS/pave_doc/). Figure Figure 4 shows maps of six elements’ emissions, which have features that were

observed to correspond to different types of sources. Emissions maps of the remaining 36 species and PM_{2.5} from each of the 85 source categories are contained in the Supporting information. The inclusion of emissions from Canada and Mexico in the 2001 CAIR inventory can be seen from many of these maps; however, these emissions were not included in the U.S. national totals discussed elsewhere in this paper.

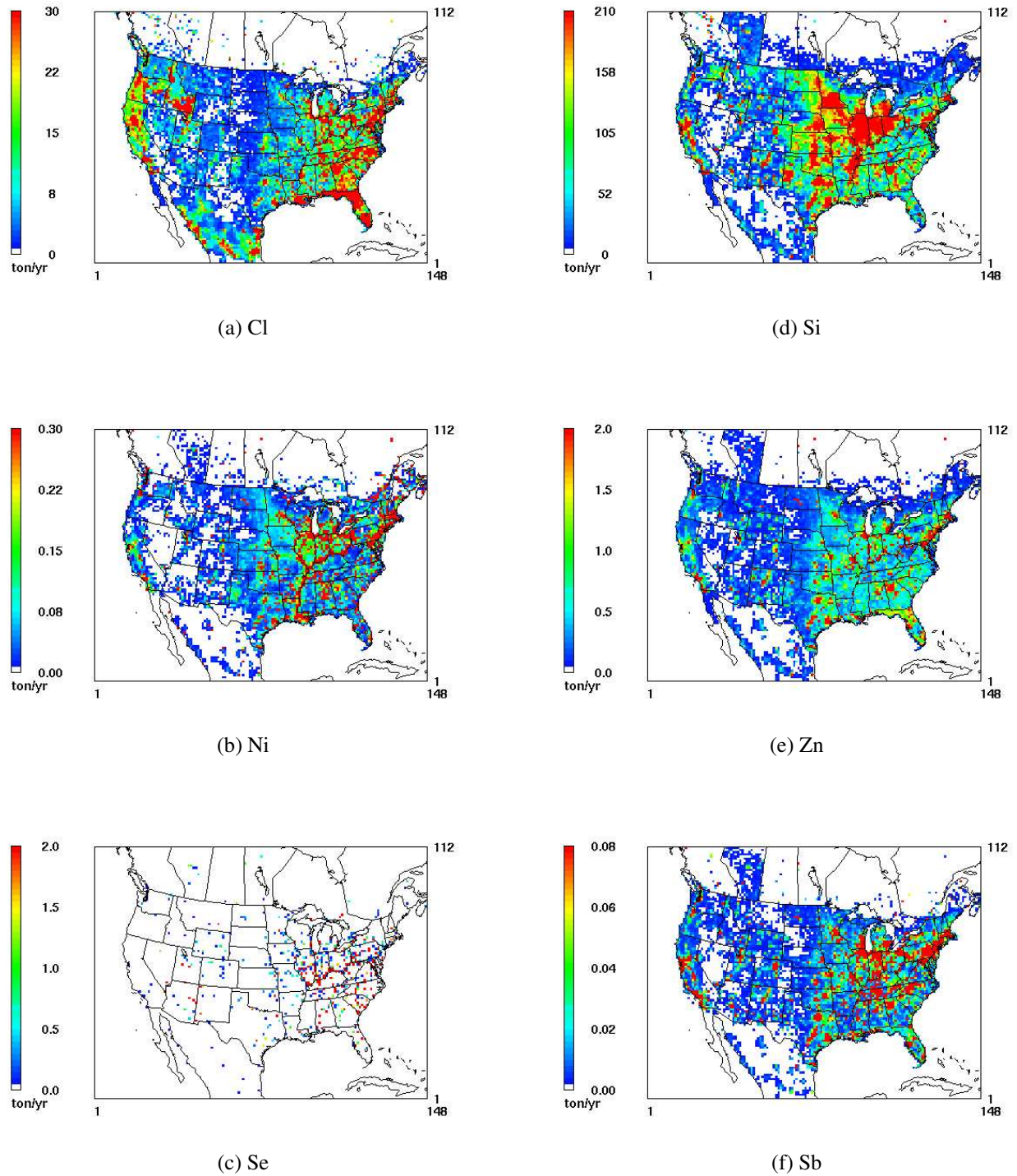
Emissions of species such as Cl (Figure 4(a)) are primarily from biomass burning-type sources. Cl emissions in Figure 4(a) are largest in Florida, and a similar pattern can also be seen in the spatial distributions of species such as Br and K. Biomass burning emissions in Florida are due primarily to Wildfires; Prescribed Burning also contributes to emissions in both Florida and nearby states in the southeast. This signal in Florida is due to a recent reallocation of PM_{2.5} emissions from 2001 fire events (28) and should not be expected to be representative of emission inventories of other years. Biomass Burning on the west coast are also due primarily to Wildfires; in the Northeast and other urban areas, they are from Residential Wood Combustion. Agricultural Burning is responsible for Biomass Burning in the southeast regions of Idaho and Louisiana.

Combustion of oil and coal are the major sources of fossil fuel-type emissions in the inventory. Maps of Ni and Se exhibited spatial patterns representative of these two types of emissions, which are shown in Figures 4(b) and 4(c). The large emissions of Ni along the Mississippi and Ohio rivers and in the Northeast are spatial pattern signatures of Residual Oil Combustion. Major emissions of Se show a distinct point-source pattern concentrated in the eastern U.S., which is strongly characteristic of the PM_{2.5} emissions from the Bituminous Coal Combustion source category.

Emissions of crustal elements are exemplified by the map of Si emissions in Figure 4(d), which exhibits a regional spatial pattern in the central portion of the continental U.S., overlaid by high emissions from most urban centers. Emissions from the Agricultural Soil source category are largely responsible for the regional Midwest emissions pattern near Iowa and Illinois in Figures, whereas Paved Road Dust emissions are responsible for the urban hotspots in those same maps (see Supporting Information). Fugitive dust source categories (Figure 4(c)) exhibit very sparsely distributed emissions in the western half of the U.S. This is due to the intentional absence of windblown dust emissions in the inventory; inclusion of such emissions in the inventory would enhance emissions of this source category in the western U.S. Similar spatial patterns can be seen in the maps of other crustal elements such as Al, Si, Ca, Fe, and Ti in the Supporting Information.

Motor Vehicle emissions unsurprisingly exhibit a primarily urban pattern, which is illustrated by the map of Zn emissions in Figure 4(e). This spatial pattern is also reflected in the distribution of EC emissions as well as in the emissions of species that motor vehicles influence less heavily such as Ag.

Many of the inventory's source categories that did not fit into any of the types discussed above contained emissions from a number of miscellaneous industrial sources such as Aluminum Production, Electric Arc Furnace, Catalytic Cracking and the "Avg" categories. The element Sb is emitted from a number of these types of sources, and the map of its spatial distribution is shown in Figure 4(d). Figure 4(d) shows that emissions from miscellaneous industrial sources are scattered throughout the country, and are somewhat more concentrated in the eastern half of the U.S.

Figure 4: Spatial distributions of 2001 NEI PM_{2.5} emissions of select trace elements.

Discussion

The process of speciating the 2001 PM_{2.5} NEI using SPECIATE profiles revealed a number of critical gaps in the available data. For example, the “Avg” source categories shown in Figure Figure 2 to have no trace element emissions; the speciation data for these categories are generic, historically based profiles that were mapped to a diverse set of SCCs for which data more specific to these emissions were not available in the current version of the SPECIATE database. Sources in these categories are in need of more specific speciation information – major examples include cooling towers; wastewater treatment; production processes of various chemicals such as plastics, chlorine and derivatives, rubber, inks, and olefins; mineral production processes; pulp and paper production processes; and petroleum production processes. Major sources of PM_{2.5} that do not have emissions in the 2001 NEI at all include wind-blown dust, vegetative detritus, pollen fragments, sea salt (the “Sea Salt” source category in the 2001 NEI contains only salt mining SCCs), and cigarette smoke. Most source category profiles were composited using 10 or less source profiles, which may or may not have originated from spatially disparate source tests. The maps in Figure Figure 4 and in the Supporting Information can be used as a guide for prioritizing sources to sample in the future. Compositing a more spatially disparate collection of profiles will result in a more statistically robust set of source category profiles, and will provide data for region-specific emissions speciation.

This paper has presented data and methods for creating the first known emissions inventory of the full suite of PM_{2.5} species and metals measured by national monitoring networks. The new set of 85 speciation profiles accounted for about 87% of the chemical composition of PM_{2.5} emissions in the NEI, and were used to provides U.S. national spatial distributions of emissions of a number of fine particulate trace elements for the first time. In addition, the emissions of HAP

metals from fugitive dust sources have been estimated for the first time. A number of the trace elements are known to induce adverse health effects (13), and their sources need to be known and effectively controlled to limit their effects on air quality and mitigate exposure. Speciating PM_{2.5} inventories is expected to be a cost-effective aid to apportioning the sources of the metals both by providing input to air quality models (29) and by providing confirmatory data for receptor modeling studies (30). Air quality models using such an inventory would also be able to predict the ambient concentrations and deposition patterns that result from implementing controls on the sources of each element, providing a tool to evaluate the effectiveness of source control programs.

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Supporting Information Available

Detailed methods employed for creating the composite speciation profiles and the full set of emissions maps are available.

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