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An assessment of important SPECIATE profiles in the EPA emissions modeling platform and current data gaps

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ABSTRACT

The United States (US) Environmental Protection Agency (EPA)'s SPECIATE database contains speciated particulate matter (PM) and volatile organic compound (VOC) emissions profiles. Emissions profiles from anthropogenic combustion, industry, wildfires, and agricultural sources among others are key inputs for creating chemically-resolved emissions inventories for air quality modeling. While the database and its use for air quality modeling are routinely updated and evaluated, this work sets out to systematically prioritize future improvements and communicate speciation data needs to the research community. We first identify the most prominent profiles (PM and VOC) used in the EPA's 2014 emissions modeling platform based on PM mass and VOC mass and reactivity. It is important to note that the on-road profiles were excluded from this analysis since speciation for these profiles is computed internally in the MOVES model. We then investigate these profiles further for quality and to determine whether they were being appropriately matched to source types while also considering regional variability of speciated pollutants. We then applied a quantitative needs assessment ranking system which rates the profile based on age, appropriateness (i.e. is the profile being used appropriately), prevalence in the EPA modeling platform and the quality of the reference. Our analysis shows that the highest ranked profiles (e.g. profile assignments with the highest priority for updates) include $PM_{2.5}$ profiles for fires (prescribed, agricultural and wild) and VOC profiles for crude oil storage tanks and residential wood combustion of pine wood. Top ranked profiles may indicate either that there are problems with the currently available source testing or that current mappings of profiles to source categories within EPA's modeling platform need improvement. Through this process, we have identified 29 emissions source categories that would benefit from updated mapping. Many of these mapping mismatches are due to lack of emissions testing for appropriate source categories. In addition, we conclude that new source emissions testing would be especially beneficial for residential wood combustion, nonroad gasoline exhaust and nonroad diesel equipment.

1. Introduction

Quantitative emissions data for particulate matter with an aerodynamic diameter less than or equal to 2.5 µm ($PM_{2.5}$) and volatile organic compounds (VOCs) are important for air quality management and for investigations of $PM_{2.5}$/VOC toxicity and ozone (O_3) (Pope et al., 2002, 2009; Crouse et al., 2012; Cooke, 1991; Becher et al., 1996; Kostiainen, 1995). The United States (US) Environmental Protection Agency (EPA)'s SPECIATE database (Simon et al., 2010; https://www.epa.gov/air-emissions-modeling/speciate-version-45-through-40) contains speciation profiles for both VOC and PM emission sources. These speciation profiles provide the data needed to disaggregate total VOC and $PM_{2.5}$ emissions compiled in the National Emissions Inventory (NEI) into their component species. For instance, PM emissions are commonly broken out into organic carbon (OC), elemental carbon (EC), sulfate, nitrate, ammonium and a variety of trace metals. Similarly, speciation profiles apportion VOC emissions into a variety of specific organic compounds such as propane, formaldehyde and methanol or classes of compounds such as C-10 paraffins and mineral spirits. Chemical speciation profiles of organic gasses in SPECIATE include

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chemical constituents that are exempt under 40 CFR 51.100(s). For the purpose of this paper, we refer to these profiles as "VOC profiles" despite the inclusion of exempt species in order to maintain consistency with terminology used in the EPA's modeling platform. Speciation profiles are specific to different source types (e.g. residential wood combustion, coal-fired boilers, construction equipment etc.).

The SPECIATE database, and speciation profiles more generally, have a variety of important applications relevant to air quality research and regulatory decision making. First, speciation profiles are an essential component of studies that use ambient measurements to determine source attribution of air pollution through techniques such as chemical mass balance (CMB) (Watson et al., 1989; Schauer et al., 1996; Schauer and Cass, 2000; Watson et al., 2015). While some ambient source attribution methods such as positive matric factorization (PMF) (Larsen and Baker, 2003; Lanz et al., 2007; Aiken et al., 2009; Ulbrich et al., 2009) do not rely on emissions speciation for the initial identification of unique factors, source-specific speciation profiles can play a key role in attributing factors to emissions source types (Watson et al., 2015; Blanchard et al., 2012; Hackstadt and Peng, 2014; Zheng et al., 2011; Hu et al., 2014). Second, speciation profiles can be used to estimate emissions of specific hazardous and toxic air pollutants (HAPs) when primary emissions factors of $PM_{2.5}$ and VOCs are available but emissions factors for specific HAPs are not. Third, the National Emission Inventory (NEI), starting with the 2014 NEI, contains $PM_{2.5}$ species as pollutants for each emission source category and, thus, the SPECIATE database is essential to NEI development. The focus of this paper is the use of speciation profiles to create inputs necessary to run photochemical air quality models. These photochemical models are an important research tool for understanding regional air pollution sources, fate, and transport and are also applied in regulatory analyses of regional haze, $PM_{2.5}$, and ozone. Photochemical models which calculate the impact of emissions, chemistry, atmospheric dispersion, gas-aerosol partitioning and deposition on air pollution concentrations, require gridded, hourly, speciated emissions inputs for both gas-phase and particulate matter pollutants (Reff et al., 2009; Simon et al., 2010).

The data compiled in SPECIATE are obtained through a combination of peer-reviewed literature (e.g., Fujita et al., 1995; Hays et al., 2002; Schauer et al., 2001; Watson et al., 1989) and emissions testing and surveys conducted by industry and government agencies. Profiles derived solely through engineering judgement are also available in SPECIATE but are considered less reliable than profiles derived from direct measurements. While there are more than 5000 profiles in the SPECIATE database, this work focuses on the subset of approximately 400 profiles that are used in constructing EPA's 2014 emissions modeling platform (Strum et al., 2017). The NEI does not report emissions at a fine enough spatial or temporal resolution for model applications and does not include full speciation of VOCs and $PM_{2.5}$. Many emissions modeling tools and emissions data are needed to translate the NEI into a form that is appropriate for air quality model inputs (available at: https://www.epa.gov/air-emissions-modeling/emissions-modeling-tools). To create speciated emissions appropriate for model inputs, each inventory source classification code (SCC) and its associated emitted mass in the inventory is assigned to a speciation profile. In some cases, these assignments vary by region. The platform also allows mass from a single SCC to be apportioned to multiple profiles. The modeling platforms are used by the EPA to support a wide range of national regulatory analyses such as the review of the ozone (US EPA, 2019) and $PM_{2.5}$ National Ambient Air Quality Standards (US EPA, 2012), mobile source emissions control rules (US EPA, 2014), standards for power plants (US EPA, 2018a), rules governing interstate transport of air pollution (US EPA, 2016), and the National Air Toxics Assessment (NATA; US EPA, 2018b). Due to the large effort required to create these model-ready emissions in a detailed and credible manner, EPA's modeling platform is also widely used throughout the air quality modeling community both by state regulators and by scientific researchers (e.g. Ozone Transport Commission (OTC), 2016; Cohan et al., 2007; Kim et al., 2011; Ahmadov et al., 2015; Goldber et al., 2016; Travis et al., 2016). This work focuses on evaluating emissions profiles as they are applied in the 2014 modeling platform which was the most recently available platform at the time of this analysis. Specifically, we aim to identify existing gaps and limitations in the current application of SPECIATE profiles used for air quality modeling and prioritize desired improvements.

Maintenance of SPECIATE requires continuous improvement of the database. Regular additions of speciation profiles in SPECIATE provides users with access to the most current source of quality speciated emissions inventories. This paper identifies emissions sources most in need of new profiles and data, which is the first step in the process for adding new profiles to SPECIATE. Next, data sources must be identified to address those needs and those new data sources must be critically reviewed before being added to SPECIATE, based on the Q-score process described in the Supporting Information (see section S2, Table S1). This rigorous attention to maintaining quality and relevance has established SPECIATE as a uniquely positioned source of information for air quality analysts, modelers, researchers, specialists, as well as interested public officials and individuals. While the SPECIATE workgroup continually searches the literature for new published data, this process is not exhaustive so communications from the scientific community are welcome.[1] New data sources will help fill identified gaps in SPECIATE and, in turn, help to improve air quality modeling conducted by the U.S. EPA and others in the regulatory and scientific modeling community.

2. Methodology

In this analysis, we undertook a multi-step process to evaluate the quality of emission source profiles and to prioritize improvements to speciation data. The analysis starts with the subset of VOC and $PM_{2.5}$ profiles applied in EPA's 2014 version 7.0 air emissions modeling platform which used version 1 of the 2014 National Emissions Inventory (US EPA, 2018c; US EPA, 2016a,b). However, from this set of profiles we selected for this analysis those profiles that are most "prominent" in the emissions modeling platform. In this context, "prominent" profiles are determined based on the total mass assigned to each profile for VOC and $PM_{2.5}$ and on a reactivity-weighting scheme for VOC. For the prominent profiles, we then conducted an in-depth evaluation of both profile reliability and applicability to sources to which they are being assigned using a point-based system. Based on total points assigned, we identify profiles with the highest priority for updates. Highly ranked profiles may indicate either that there are problems with the currently available source testing or that current mappings of profiles to source categories within EPA's modeling platform need improvement. It is important to note that for this analysis, the on road emission source profiles were excluded for both VOC and $PM_{2.5}$ emission source profiles and the nonroad emission source profiles were excluded for the VOC emission source profiles.

2.1. Determining prominent profiles

2.1.1. Mass basis

In order to determine which profiles are most prominent in the emissions modeling platform based on the mass of emissions assigned, we began with a summary of the amount of VOC and $PM_{2.5}$ assigned to each profile by county and SCC for all counties in the Contiguous United States (CONUS). Onroad mobile sources were excluded from both VOC and $PM_{2.5}$ analyses because chemical speciation for this

[1] In addition to direct communication with the corresponding author, the SPECIATE workgroup can also be contacted through EPA's website: https://www.epa.gov/air-emissions-modeling/forms/contact-us-about-air-emissions-modeling.

source category is performed internally within the EPA's MOtor Vehicle Emission Simulator (MOVES) model and is not based on an external mapping of speciation profiles to SCC. Similarly, VOC speciation (though not $PM_{2.5}$ speciation) for nonroad equipment is performed partially within MOVES, so these sources were also excluded from the VOC mass ranking process. In addition, it is also important to note that the 2014v1 NEI $PM_{2.5}$ fugitive dust emissions (e.g., agricultural dust, road dust and construction dust) do not account for very near source deposition (Veranth et al., 2003; Etyemezian et al., 2004) and soil moisture conditions which impact the amount of dust entrained in the atmosphere. These processes are accounted for in the translation of the NEI into the EPA emissions platform through meteorological adjustments and application of the "capture fraction" of dust (Pace, 2005; Pouliot et al., 2011; Appel et al., 2013). Therefore, we adjusted state-level dust emissions based on a state-level summary of the percent of $PM_{2.5}$ emissions adjustments (reductions) in the emissions modeling platform (Strum et al., 2018). For example, Florida's unadjusted 2014 $PM_{2.5}$ dust emissions of 35,137 tons from the NEI was reduced 62% in the emissions modeling platform, so in this analysis we applied a 62% reduction to all Florida dusts SCCs when identifying prominent profiles.

We determined profiles that were prominent both nationally and in each of the National Oceanic and Atmospheric Administration (NOAA) climate regions shown in Fig. 1. Nationally and for each region, we rank-ordered the profiles for both $PM_{2.5}$ and VOC based on the total mass assigned to each within the 2014 EPA modeling platform. Profiles that accounted for greater than 2% of the total mass for VOCs and greater than 1% of the total mass for $PM_{2.5}$ were included in an in-depth evaluation.

2.1.2. Reactivity weighting

Given that the ozone-production efficiency of VOCs varies, profile ranking was also performed on the basis of reactivity (i.e. ozone forming potential). Emissions were weighted based on the reactivity of each profile's component species using the following equation,

$$RWE = VOCmass_{profile} \times VOCtoTOG_{profile} \times \sum_{species\ i}^{all\ species\ in\ profile} (R_{species\ i} \times TOG\_SplitFactor_{species\ i})$$

Where:

RWE = the reactivity weighted emissions (units of moles ozone (O_3)) associated with a profile based on the sum of CB6 species (Yarwood et al., 2010)

VOCmass*profile* = the grams of VOC associated with the profile

R species i = the Maximum Incremental Reactivity (MIR; http://www.camx.com/files/camxusersguide_v5-40.aspx) of species *i*, in moles of O_3/mole of species *i*

VOCtoTOGprofile = the SPECIATE database's VOC to TOG (Total Organic Gases) conversion factor, which is the mass of TOG per mass of VOC (g/g) and is specific to the profile

TOG_Split Factorspecies i = the mole based split factor (i.e. the moles of species *i* per mass of TOG).

For the purpose of this analysis, we converted explicit species into surrogate chemical mechanism species from the CB6 chemical mechanism (Yarwood et al., 2010) which is the primary chemical mechanism being used for the 2014 EPA modeling platform. Like the VOC mass analysis, MOVES-based mobile sources were excluded. For this analysis, we selected profiles that accounted for greater than 1% of the total reactivity-weighted VOC for the in-depth evaluation.

2.2. Priority ranking system

In order to determine which profiles are most in need of updates, we developed a ranking system based on six major criteria: (1) representativeness of current technology (i.e. does the tested emission source still represent the current technology?), (2) the level of documentation (e.g. peer reviewed, report, etc), (3) the appropriateness of SCCs to which profiles are applied, (4) prevalence in the EPA modeling platform, (5) any additional known problems with the profile or the data provided in the original reference or in the SPECIATE database, and (6) applicability of study region. Higher scoring profiles are those with higher priority need for improvements. The criteria and scoring are described in more detail below and provided in Table 1. Each criterion is evaluated for each prominent profile at a regional scale.

(1) The first criterion examined how well the profile for the tested emission source represents the current technology being used. For this purpose, "technology" may refer to engine design, control methods, product formulation or other aspects of the emissions source which may have changed since the original emissions source testing that went into the profile creation. This criterion is determined on a three-point scale, where 0 points are given if the

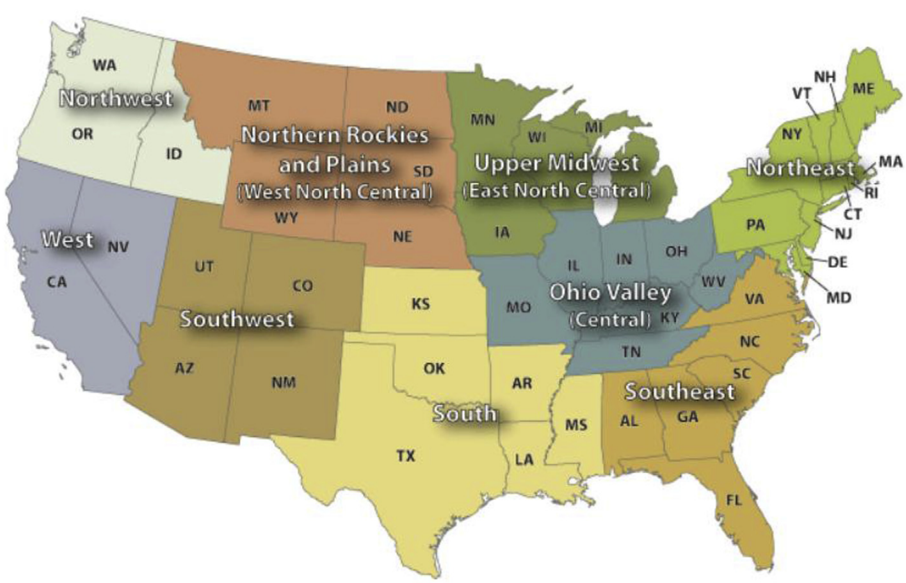

Fig. 1. NOAA climate regions (Source: NOAA - https://www.ncdc.noaa.gov/monitoring-references/maps/us-climate-regions.php).

Table 1
Priority ranking system basis for point assignment to determine speciation profiles most in need of updates in based on EPA 2014 emissions modeling platform.

Criteria		Points Assigned
(1) Tested emissions source representative of current technology	Technology (source) still in use	0
	Mix of outdated technologies and technologies still in use	1
	Teste source no longer represents current sources	2
(2) Level of Documentation	Peer reviewed	0
	Documented in report or other accessible record	1
	No/missing documentation or engineering judgement	2
(3) Inventory source matches tested source	All SCCs match tested source	0
	Less than 10% of SCC emission mass do not match tested source	1
	More than 10% do not match tested source	2
(4) Percent of emitted mass/reactivity assigned to this profile in EPA 2014 emissions modeling platform	< 4% (mass-based); < 3% (reactivity-based)	0
	4–8% (mass-based); 3–7% (reactivity-based)	1
	8–12% (mass-based); 7–14% (reactivity-based)	2
	> 12% (mass-based); > 14% (reactivity-based)	3
(5) Error/Discrepancy in data?	None	0
	Discrepancy	1
	Error	2
(6) Geographic region of source applicable?	Yes	0
	No	1

technology is still in use, 1 point is given if there is a mix of technologies in use (i.e. some that are representative of the emissions source measured to create the profile and some that are newer) and 2 points are given if the source is no longer in use (i.e. the nature of the source has changed (e.g. technologies used) such that the existing speciation profile assigned to that source may no longer be representative of the species emitted by that source).

(2) Each profile is associated with a reference, which describes how the data were developed for the respective profile. The references are graded on a three-point scale based on the level of documentation. If the reference for the profile comes from a peer reviewed journal article, 0 points are given to the profile. If the reference is a report or document, the profile is given 1 point. Finally, if the profile does not have an obtainable reference and/or is based on engineering judgement, 2 points are given.

(3) The next criterion characterizes how well the inventory source matches the tested source. If all of the SCCs being applied to a specific profile match the source associated with the profile, then 0 points are given to the profile. If less than 10% of emissions assigned to a profile come from SCCs that don't match the profile, then the profile is given 1 point. Finally, if more than 10% of emissions assigned to a profile come from SCCs that don't match the profile, then the profile is given 2 points.

(4) Profiles were also evaluated on their prevalence in the modeling platform based on percent of emission mass weighting or reactivity weighting on a regional scale. While prevalence does not indicate a problematic profile, the inclusion of this factor is intended to ensure that the highest priority profiles are not dominated by profiles that have minimal influence on total emitted mass or reactivity. Prevalence is based on a four-point scale with ranges determined using Jenks Natural Breaks, a method of clustering data into different classes based on natural breaks in the dataset (Jenks, 1967). The Jenks Natural Breaks were determined independently for mass (VOC and PM) and reactivity-weighted emissions.

(5) We also considered any notes made either by those who created the profiles or by other researchers who identified any errors or discrepancies with the profile where a noted discrepancy in the data is given 1 point and a noted error in the data is given 2 points. A discrepancy in the data was determined using statements made by the authors in the reference for the profile as well as the J-rating and the Quality rating given in the SPECIATE database. The J rating for a profile in the SPECIATE database is based on the judgment of the SPECIATE workgroup, while the quality rating in the database factors in many different aspects on the quality of the profile, such as the number of samples taken and the age of the data (Hsu et al., 2016). A point was awarded to the profile if one or more of the following occurred: there were notes in the primary reference of the profile about high uncertainty in the data, the profile J rating was 1 (lowest), and/or the Quality rating of the profile was E (lowest). The only one profile with a known error in the data (2 points) is the $PM_{2.5}$ composite[2] wildfire profile (91102). We determined that one of the primary profiles used to construct the composite was for the combustion of tarred fence posts (thus creating an artificial increase in the amount of elemental carbon formed), which is not representative of a wildfire.

(6) Finally, for source categories that vary geographically, the region of applicability for the sources measured in the creation of the profile was compared with the region to which the profile is being applied in the modeling platform. One point is given if the profile is not assigned appropriately to sources that vary geographically. An example of a profile that was given 1 point based on the geographic applicability would be the wildfire composite profile (91102) for $PM_{2.5}$. Measurements for this profile were taken in the South and the Southwest regions, but it is also being applied in the West, Northern Rockies and Plains, Northwest, Northeast and the Southeast. This is inappropriate because vegetation type (fuel) varies regionally.

The profiles were then assigned their respective point values (summarized in Table 1) per each region and the regional sums were averaged to create priority rankings for both $PM_{2.5}$ and VOC source profiles that need updates. The score was calculated for each region in which the profile was prominent (i.e. greater than 2% of PM mass and greater than 1% of VOC mass/reactivity) and then the regional scores were averaged to create a single score per profile. In this ranking system, the higher the number of points awarded to an emission source profile, the more in need is the source profile of updating. Profiles were

[2] It is important to note that some profiles in SPECIATE are composites, or averages, of other profiles that exist within the database. Furthermore, there are also some profiles that are "study composites," in which the author of the study used data from different tests to create a composite profile. The newest version of SPECIATE (v5.0) will distinguish between these two types of composites.

also assigned to different categories that specify the actions needed to update/improve the profile: (1) Need New Data, (2) Specificity, (3) No Action Recommended. Those profiles that are listed under the 'Need New Data' category contain old, poorly documented, and/or poor-quality data and therefore require new measurements. Those profiles that are listed under the 'Specificity' category are applied to inappropriate SCCs. In this case, the profile may be too general or too specific relative to the SCCs to which it is assigned. This does not indicate that data quality is necessarily poor. However, in many cases the reason for the specificity mismatch is due to lack of existing profiles for certain SCCs. In that case, the profile would be placed in both the 'Need New Data' action category and the 'Specificity' action category since new measurement are needed for the source categories without appropriate data. The final action category is 'No Action Recommended' which is composed of profiles that are of either 1) of adequate quality/specificity, but received a high priority ranking due mostly to their prevalence in the modeling platform (criterion #4) or 2) catch-all profiles that may have had poor quality or inappropriate specificity but included numerous sources, each with very small emissions. In the second case, profiles were deemed a low priority for updates because it was expected that there would be a substantial effort required to collect data on many sources, each accounting for very small percentages of total emitted mass. The results of the quality analysis provided the basis for identifying gaps in the current SPECIATE profile database and for identifying profile assignments to SCCs that need to be improved.

3. Results

3.1. Most prominent profiles

3.1.1. Mass basis - top $PM_{2.5}$ profiles

Fig. 2 depicts the relative amount of emitted mass assigned to each of the top (i.e. greater than 1% of the total emission mass in a region) $PM_{2.5}$ profiles on both a regional and national scale in the 2014 US EPA modeling platform. Supplemental Table S2 provides percent contributions to emitted mass from the top three emission source profiles both for the nation and in each region. The most prominent $PM_{2.5}$ emission profiles are composite profiles derived from Reff et al. (2009), who created these category-specific speciation profiles based on the median of multiple high-quality profiles that were already in the SPECIATE database. On a national scale, the three most prominent profiles are Wildfires – Composite, Prescribed Burning - Composite, and Residential Wood Combustion – Composite. These account for approximately 23%, 20% and 9%, respectively, of $PM_{2.5}$ emissions nationally. The Wildfire – Composite profile also accounts for the largest percentage of overall $PM_{2.5}$ emissions in the West (~71%), the Northwest (~57%) and the Southwest (~22%). Similarly, the Prescribed Burning – Composite profile accounts for the largest percentage of overall $PM_{2.5}$ emissions in the Northern Rockies and Plains (~27%), the Southeast (~39%), the South (~31%) and the Ohio Valley (~17%) regions of the United States. In the Upper Midwest and in the Northeast, the Residential Wood Combustion – Composite profile accounts for the largest percentage of emissions of fine particulate matter, ~31% and ~27%, respectively. There are a few additional $PM_{2.5}$ composite profiles that are prominent in one region but not nationally. For instance, the Sub-Bituminous [coal] Combustion – Composite profile is the second most prominent profile in the Ohio Valley (~16%) and the Charbroiling – Composite profile is the second most prominent profile in the Northeast (~18%).

3.1.2. Mass basis - top anthropogenic VOC profiles

Many more VOC profiles (339) are used than $PM_{2.5}$ profiles (79) in the 2014 emissions modeling platform, and there is much more variation on a regional scale in the specific profiles that contribute to over

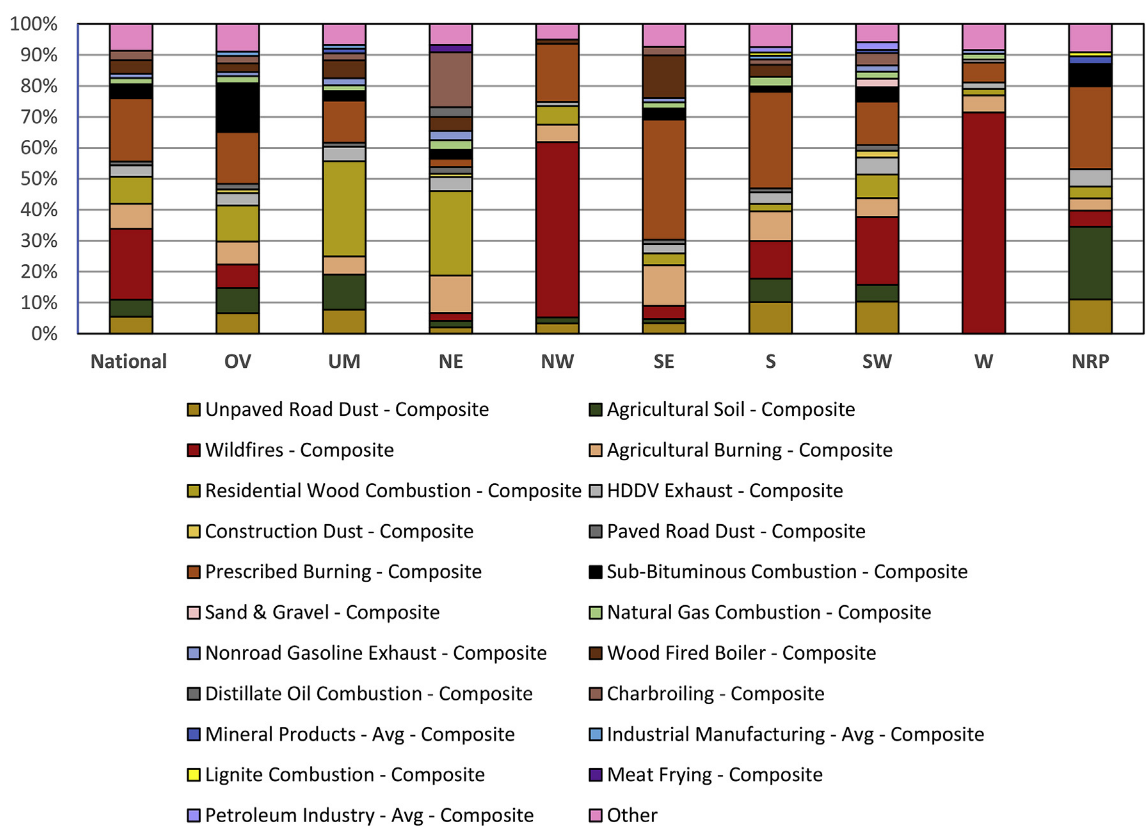

Fig. 2. Percent of emitted mass assigned to each $PM_{2.5}$ profile on a national scale and broken out by region excluding the on-road sector. The "Other" category represents those profiles that account for less than 1% of the total emitted mass. The following abbreviations were used to represent US regions: NE = Northeast, SE = Southeast, OV = Ohio Valley, UM = Upper Midwest, S = South, NRP = Northern Rockies and Plains, SW = Southwest, W = West, NW = Northwest.

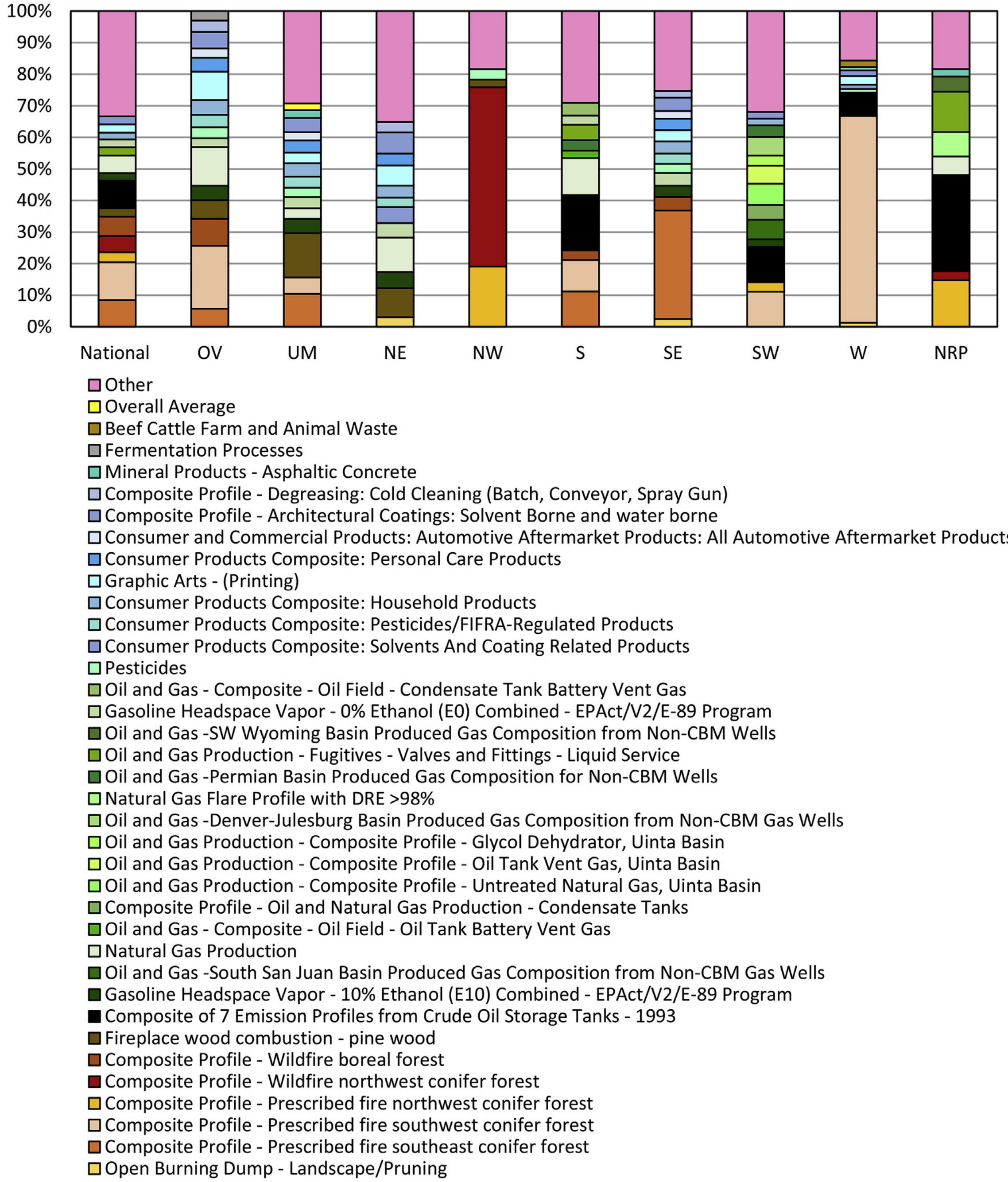

Fig. 3. Percent of emitted mass assigned to each VOC profile on a national scale and broken out by region. The green shades represent major oil and gas profiles, the blue shades represent profiles for solvents, the red and orange shades represent profiles for the combustion of biomass and the pink represents profiles listed as 'Other', which encompasses profiles that account for less than 2% of the total emitted VOC mass. The following abbreviations were used to represent US regions: NE = Northeast, SE = Southeast, OV = Ohio Valley, UM = Upper Midwest, S = South, NRP = Northern Rockies and Plains, SW = Southwest, W = West, NW = Northwest. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2% of the total VOC emission mass compared to $PM_{2.5}$. Like the $PM_{2.5}$ profiles discussed above, several VOC emission profiles are also composites of other profiles already in the SPECIATE database. Fig. 3 shows the most prominent profiles at both the national and regional levels, while Table S3 lists the three most prominent profiles in each region and at the national level. There are three main emission source categories that dominate at the national and regional level: 1) biomass burning (shown in red and orange in Figs. 3), 2) oil and gas (shown in black and green in Fig. 3) and 3) solvents including pesticides, consumer products and architectural coatings (Shown in shades of blue in Fig. 3). Nationally, the most prominent VOC profiles include Composite Profile - Prescribed fire southwest conifer forest (~12%), Composite of 7 Emission Profiles from Crude Oil Storage Tanks – 1993 (~9%) and Composite Profile - Prescribed fire southeast conifer forest (~8%). Biomass combustion profiles account for a substantial portion of the emitted VOC mass in the Ohio Valley (Composite Profile - Prescribed fire southwest conifer forest, ~15%), the Upper Midwest (Fireplace wood combustion - pine wood, ~14%), the Northwest (Composite Profile - Wildfire northwest conifer forest, ~57%), the Southeast (Composite Profile - Prescribed fire southeast, ~34%), and the West (Composite Profile - Prescribed fire southwest conifer forest, ~66%). Oil and gas related profiles are important in the South, Southwest and

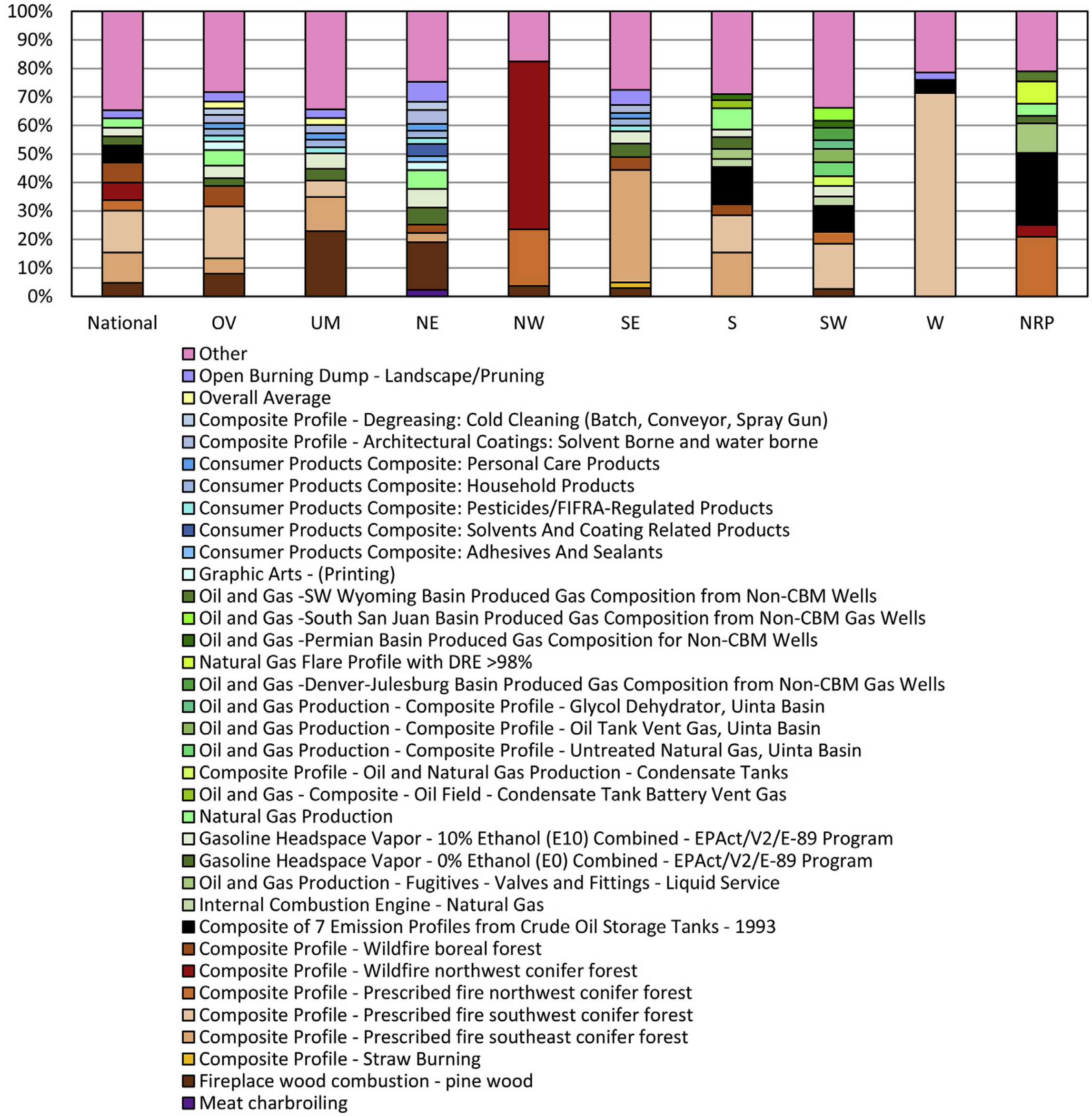

Fig. 4. Percent of VOC reactivity assigned to each VOC profile on a regional and national scale and broken out by region. The green shades represent major oil and gas profiles, the blue shades represent profiles for solvents, the red and orange shades represent profiles for the combustion of biomass and the pink represents profiles listed as 'Other', which are any profiles that accounts for less than 2% of the total emission mass. The following abbreviations were used to represent US regions: NE = Northeast, SE = Southeast, OV = Ohio Valley, UM = Upper Midwest, S = South, NRP = Northern Rockies and Plains, SW = Southwest, W = West, NW = Northwest. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Northeast regions: Composite of 7 Emission Profiles from Crude Oil profile accounts for the largest percentage of VOC emissions by mass in the South (~18%) and in the Southwest (~11%) and the Natural Gas Production profile accounts for the largest percentage of emissions in the Northeast (~11%). In both Figs. 3 and 4, a substantial percentage of the total emitted VOC mass and reactivity is assigned to the 'Other' category. This is due to the large number of source profiles that are mapped to less than 2% of the overall emitted mass individually (e.g. some oil and gas emission source profiles that are basin specific, some consumer product emission source profiles), but together account for a substantial fraction of the total.

3.1.3. Reactivity basis

Fig. 4 presents the relative amount of VOC emissions weighted by ozone-forming potential (i.e. VOC reactivity). Table S4 lists the top three most prominent emissions source profiles at the national level and by region. In general, the importance of biomass burning profiles increases in the reactivity weighting scheme compared to the mass weighting scheme while the importance of oil and gas profiles and solvents marginally decreased. In terms of individual profiles, most of the prominent profiles based on emitted mass are also prominent based on reactivity weighted VOC, although there are several new oil and gas profiles that are identified in the VOC reactivity weighting analysis but not in the analysis based on mass weighting. Nationally, biomass burning profiles dominate (with individual profiles accounting for as much as 15% of the total national VOC reactivity), similar to unweighted VOC emitted mass. Biomass burning also dominates in all regions, except for the Northern Rockies and Plains region. In this region, the composite of 7 emission profiles from crude oil storage tanks profile accounts for the largest percentage of VOC reactivity (~25%). When looking at the broad groupings of individual profiles, it is evident that emissions in the Southwest, the South and the Northern Rockies and Plains are dominated by the sum of emissions using the oil and gas profiles. Because these oil and gas profiles are basin specific, they do not represent a significant fraction of the total emissions, individually. However, when they are combined, oil and gas is dominant based on VOC reactivity in addition to emitted mass in these regions. It is important to note that while the focus of this work is the CONUS, future work with oil and gas profiles for Alaskan basins would be extremely beneficial.

3.2. Prioritization of profiles

Tables S2, S13 and S14 in the supplemental information show the point assignments for each profile evaluated using the criteria in Table 1. Furthermore, the supplemental information also includes a thorough description of each profile. The sections below summarize key findings from this evaluation and prioritization analysis for $PM_{2.5}$ and VOC profiles. The priority ranking system described above (Table 1) was used to prioritize the profiles shown in Figs. 2–4, which are specific to the US EPA, 2014 emissions modeling platform. The profiles considered are those which were applied to > 1% of the emitted $PM_{2.5}$ mass and applied to > 2% of the emitted VOC mass (or reactivity-weighted VOC emissions). Each profile was evaluated at a regional level and then the regional scores were averaged to create the national scoring. In total, rankings were determined for 18 $PM_{2.5}$ profiles and 39 VOC profiles. The results of the priority ranking for the top $PM_{2.5}$ source profiles (mass basis) are presented in Table 2, while Table 3 shows the results of the priority ranking for the most prominent VOC emission source profiles based on both mass and reactivity.

3.2.1. $PM_{2.5}$ profiles

Based on this analysis, the top two profiles in need of an update based on specificity include the Wildfire – Composite profile (profile #91102 in SPECIATE) and the Prescribed Burning- Composite profile (profile #91109 in SPECIATE). As previously mentioned, the wildfire composite profile contains an error because it includes measurements from the combustion of tarred fence posts. In addition, starting with the 2014 NEI, the wildfire SCCs in the emissions inventory split out emissions from smoldering versus flaming conditions, which are two very different fire processes that result in different emissions composition, while the wildfire composite profile itself does not. Furthermore, this profile is extremely prominent across many regions, but does not account for regional variability in the fuels. Similarly, the agricultural burning profile doesn't account for regionality and the SCCs are crop specific, while the profile, itself, is not. Furthermore, this profile was also given a point for a discrepancy in the data based on the Quality rating of E. These are examples of profiles that are not specific enough when compared against the SCCs to which they are being applied.

In some cases, a profile may need new data as well as not matching in specificity as exemplified by the Sub-Bituminous Combustion – Composite profile (profile #91100 in SPECIATE). The sub-bituminous coal combustion source profile is based on measurements from a source with outdated technology (measurements were made in the late 1980s and are not representative of current control technologies that have been implemented since the Mercury and Air Toxics Standards (MATS) rule (40 CFR Part 63, Subpart UUUUU)). In addition, there was substantial variation among samples in measured weight percent of sulfate, elemental carbon, organic carbon and metal measurements. The authors note considerable uncertainty in these measurements due to the high inter-sample variability.

3.2.2. VOC profiles

Table 3 shows the results of the prioritization analysis for those VOC source profiles for both emission mass and reactivity. As a reminder, this analysis excludes both on-road and nonroad mobile emissions for VOC. The highest ranked profile for both mass and reactivity is the Overall Average profile (profile # 0000 in SPECIATE), which is a default profile for those SCCs that are not associated with a profile. Because this profile is associated with a variety of different SCCs that could not be placed elsewhere, this profile is part of the 'Specificity' category. The profile ranked second overall for both reactivity and mass is the Fireplace wood combustion – pine wood (profile #4642 in SPECIATE). This profile is prominent in many regions (Ohio Valley, Upper Midwest, Northwest, West, Northeast, Southeast, Southwest) and is also applied to a wide range of SCCs that are not appropriate to the sources used in the creation of the profile. While this profile is based on two measurements taken from a fireplace in southern California, the SCCs using this profile include a variety of different types of combustors (e.g. wood stoves). Furthermore, the profile is specific to pine wood, while the SCCs to which the profile is being applied are not specific to pine wood. Finally, this profile contains a Quality rating of E. Therefore, emissions testing for this profile needs to be updated and additionally tests should be conducted to better match specific SCC categories. The emissions source profile for crude oil storage tanks (profile #2487 in SPECIATE) was ranked third highest overall for both emission mass and reactivity. The crude oil storage tanks emission source profile is based on measurements taken in 1989 in Oklahoma. The primary concern with this profile is that it is not an appropriate match for all the SCCs to which the profile is being applied (e.g. several SCCs for on shore gas production). Furthermore, this profile is also prominent in several different regions (South, Southwest, West, Northern Rockies and Plains). While basin-specific profiles will be created and are being applied in some locations, other areas are still using the generic crude oil storage tank speciation profile.

3.3. In-progress SPECIATE updates

There are some profiles that are currently in the process of being updated for SPECIATE 5.0. These profiles include the $PM_{2.5}$ emission source profile number 91112 in SPECIATE (natural gas production), VOC profile number 8744 (composite profile for architectural coatings for both solvent borne and water borne coatings), and VOC profile numbers 3142–3147 (consumer product composites). These profiles will all be updated in SPECIATE 5.0 based on new data from reports and the literature. For example, the VOC emission source profile for architectural coatings for both solvent borne and water borne coatings (profile # 8744 in SPECIATE), which was derived based on data obtained from the 1997 California Air Resources Board (CARB) Aerosol Coatings Survey, has been updated based on data from the 2005 CARB Aerosol Coatings Survey (profile # 95513 in SPECIATE). Furthermore, the VOC source emission profiles for consumer product composites (profile #s 3142–3147 in SPECIATE) have been updated based on the 2010 CARB Consumer Product Survey (profile #s 95507–95512 in SPECIATE) (Yang et al., 2014).

It is important to note that while the addition of these data may improve the quality of emission source profiles currently being applied in the SPECIATE database, this does not eliminate the need for further updates to the emission source profiles. In addition, we note that an extensive literature search for new profile data was not a part of this analysis.

4. Conclusions

The results of this analysis showed that the most prevalent $PM_{2.5}$ profiles and VOC profiles used in the 2014 US EPA modeling platform based on emitted mass and reactivity vary by region, and that the top profiles nationally also include the most prevalent profiles across regions. For $PM_{2.5}$, the profiles assigned to the largest amount of emitted mass include those for wildfires and prescribed burning, which are prominent across the majority of the NOAA Climate Regions. The most prevalent VOC profiles based on emitted mass included those for prescribed/wildfires across much of the CONUS, however, emissions from fireplace wood combustion are dominant in the Upper Midwest, natural gas combustion emissions are most prominent in the Northeast and emissions from crude oil storage tanks are most prominent in the

Table 2
Priority ranking for $PM_{2.5}$ source profiles (mass basis). The following abbreviations were used to represent US regions: NE = Northeast, SE = Southeast, OV = Ohio Valley, UM = Upper Midwest, S = South, NRP = Northern Rockies and Plains, SW = Southwest, W = West, NW = Northwest.

Profile # in SPECIATE	Profile Description	Overall Ranking	% of National Emission Mass	Action Category	Needs
91102	Wildfires - Composite	1	22.8	Specificity	New measurements (fuels from OV, UM, NE, NW, SE, W); Separate profiles for smoldering v flaming; Take out fence post measurements from composite
91109	Prescribed Burning - Composite	2	20.5	Specificity	New measurements (need measurements using fuel specific to OV, UM, NE, S, SW, W, NRP); Separate profiles for smoldering v flaming
91110	Sub-Bituminous Combustion - Composite	3	4.5	Need New Data, Specificity	New measurements; SCC issue (control type: e.g. wet scrubber, ESP; coal type: e.g. Anthracite, bituminous, sub-bituminous, petroleum coke, coke, metallurgical coke)
91103	Agricultural Burning - Composite	4	8.1	Specificity	SCC issue (crop specific profiles needed; separate profile needed for house hold waste and yard waste SCCs)
91145[a]	Petroleum Industry - Avg - Composite	5	0.6	No Action Recommended	Extremely general profile with SCCs for industrial petroleum industry processes and oil and gas exploration and production processes
91121[a]	Industrial Manufacturing - Avg - Composite	5	0.8	No Action Recommended	Extremely general profile with SCCs ranging from metal production, cooling towers, electrical equipment manufacturing, transportation equipment, textile products
91120[a]	Mineral Products - Avg - Composite	5	0.6	No Action Recommended	Extremely general profile with SCCs ranging from metal production to mineral production (ceramic clay, brick, asphalt concrete, lime, talc)
91113	Nonroad Gasoline Exhaust - Composite	5	1.3	Need New Data; Specificity	New measurements; SCC issue (extremely wide range, including 4-stroke/2 stroke recreational/ commercial)
91101	Agricultural Soil - Composite	5	5.5	Specificity	New measurements (need information on soils specific to different regions: OV, UM, NE, SE, NRP), crop management specific and livestock type/management specific); SCC issue (crops v livestock)
91106	HDDV Exhaust - Composite	6	3.6	Need New Data, Specificity	New measurements; SCC Issue (wide range, including Agricultural equipment (i.e. tractors), Construction and mining equipment, locomotives, military aircraft, stationary internal combustion engines, commercial marine vessels, Rocket engine testing need profiles specific to source type)
91114	Wood Fired Boiler - Composite	7	4.4	Need New Data; Specificity	New measurements; SCC issue (industry specific)
91105	Residential Wood Combustion - Composite	8	8.8	Need New Data; Specificity	New measurements; SCC issue (device type- e.g. fireplace, wood stove [fireplace inserts, freestanding, pellet fired], furnace, hydronic heater, fire pit)
91112	Natural Gas Combustion -Composite	9	2.0	Specificity	SCC issue (different fuel types e.g. natural gas, process gas, landfill waste gas)
91108	Paved Road Dust - Composite	10	1.2	Specificity	Measurements from new geographic locations (UM, NE, SE) and road types (i.e. interstate versus arterial roads)
91100	Unpaved Road Dust - Composite	10	5.5	Specificity	Measurements from new geographic locations (UM, NE, NW, SE, NRP)
91125	Lignite Combustion - Composite	11	0.2	Need New Data	New measurements representative of current technology for lignite combustion
91107	Construction Dust - Composite	11	0.7	Specificity	Measurements from new geographic locations (Ohio Valley)
91116	Charbroiling - Composite	12	3.1	Specificity	SCCs industry specific but not meat specific

[a] Profiles are extremely general, however, breaking into specific profiles is not a top priority at this time due to the small fraction of weight this profile is applied to.

Table 3
Priority ranking for VOC source profiles (mass and reactivity basis). The following abbreviations were used to represent US regions: NE = Northeast, SE = Southeast, OV = Ohio Valley, UM = Upper Midwest, S = South, NRP = Northern Rockies and Plains, SW = Southwest, W = West, NW = Northwest.

Profile # in SPECIATE	Profile Description	Overall Ranking Mass (reactivity)	% of National Emission Mass (reactivity)	Action Category	Needs
0000	Overall Average	1(1)	0.8(1)	Specificity	Wide range of SCCs including: external combustion boilers, internal combustion engines, chemical manufacturing, food and agricultural industrial processes, metal production, mineral products, petroleum industry, pulp and paper, transportation, photographic film manufacturing, textiles, leather products, chemical evaporation, waste disposal
4642	Fireplace wood combustion - pine wood	2(2)	2.6(4.8)	Need New Data; Specificity	New measurements based on combustor type; SCC issue (specific to combustor, not specific to pine wood)
2487	Composite of 7 Emission Profiles from Crude Oil Storage Tanks - 1993	3(3)	8.7(6)	Specificity	New measurements (preferably NE, SE, SW, W, NRP); SCC issue (condensate)
1011	Oil and Gas Production - Fugitives - Valves and Fittings - Liquid Service	4(7)	2.7(1.8)	Specificity	New measurements (preferably in the S and NRP)
95422	Composite Profile - Prescribed fire southwest conifer forest	5(4)	12(14.7)	Specificity	Separate profiles for smoldering v flaming
0121	Open Burning Dump - Landscape/Pruning	6(3)	1.2(2.8)	Need New Data; Specificity	New measurements; SCC issue (household waste, open burning dump general refuse)
8745	Composite Profile - Degreasing: Cold Cleaning (Batch, Conveyor, Spray Gun)	6(6)	1.3(1.1)	Need New Data	New measurements based on current products
95421	Composite Profile - Prescribed fire southeast conifer forest	6(6)	8.5(10.7)	Specificity	Separate profiles smoldering v flaming
95423	Composite Profile - Prescribed fire northwest conifer forest	6(5)	3.2(3.8)	Specificity	Separate profiles smoldering v flaming
3001	Pesticides	7	1.7	Need New Data	New measurements based on current products
95425	Composite Profile - Wildfire boreal forest	8(6)	6.1(7)	Specificity	New measurements (in OV, S, NE, SW); Separate profiles smoldering v flaming
95424	Composite Profile - Wildfire northwest conifer forest	9(6)	5.2(6.1)	Specificity	Separate profiles smoldering v flaming
8744	Composite Profile - Architectural Coatings: Solvent Borne and water borne	10(7)	2.6(1.9)	Need New Data	New measurements based on current products
3146	Consumer Products Composite: Household Products	11(8)	2.2(1.5)	Need New Data	New measurements representative of current conditions
3145	Consumer Products Composite: Pesticides/FIFRA-Regulated Products	12(8)	1.8(1.3)	Need New Data	New measurements representative of current conditions
3144	Consumer Products Composite: Solvents and Coating Related Products	12(6)	1.4(1.1)	Need New Data	New measurements based on current products
3147	Consumer Products Composite: Personal Care Products	12(8)	2(1.2)	Need New Data	New measurements based on current products
1191	Graphic Arts - (Printing)	13(11)	2.7(1.2)	Need New Data	New measurements based on current techniques
8870	Gasoline Headspace Vapor - 10% Ethanol (E10) Combined - EPAct/V2/E−89 Program	13(9)	2.3(3)	No Action Recommended	N/A
8949	Natural Gas Production	13(12)	5.6(3.4)	Specificity	SCC issue (e.g. "Gas Exploration, well completion", "Natural gas processing")
8869	Gasoline Headspace Vapor - 0% Ethanol (E0) Combined - EPAct/V2/E−89 Program	14(10)	2.4(5)	No Action Recommended	N/A
1188	Fermentation Processes	15	0.4	Specificity	Wide range SCCs (alcohol type, process)
8520	Consumer and Commercial Products: Automotive Aftermarket Products: All Automotive Aftermarket Products	15	1.4	Need New Data	New measurements based on current products
95109a	Oil and Gas - Composite - Oil Field - Condensate Tank Battery Vent Gas	15(14)	1(0.6)	No Action Recommended	N/A
95417	Oil and Gas Production - Composite Profile - Untreated Natural Gas, Uinta Basin	15(13)	0.5(0.3)	No Action Recommended	N/A
95419	Oil and Gas Production - Composite Profile - Oil Tank Vent Gas, Uinta Basin	15(13)	0.5(0.3)	No Action Recommended	N/A
DJVNT_R	Oil and Gas -Denver-Julesburg Basin Produced Gas Composition from Non-CBM Gas Wells	15(13)	0.4(0.3)	No Action Recommended	N/A
PRM01_R	Oil and Gas -Permian Basin Produced Gas Composition for Non-CBM Wells	15(13)	1.1(0.6)	Specificity	SCC Issue (CBM wells)

(*continued on next page*)

Table 3 (*continued*)

SSJCO_R	Oil and Gas -South San Juan Basin Produced Gas Composition from Non-CBM Gas Wells	15(13)	0.5(0.3)	No Action Recommended	N/A
SWVNT_R	Oil and Gas –SW Wyoming Basin Produced Gas Composition from Non-CBM Wells	15(13)	0.4(0.2)	No Action Recommended	N/A
1007	Mineral Products - Asphaltic Concrete	16	1.3	No Action Recommended	N/A
FLR99	Natural Gas Flare Profile with DRE > 98%	16(14)	1.1(0.8)	No Action Recommended	N/A
95087a	Oil and Gas - Composite - Oil Field - Oil Tank Battery Vent Gas	16	0.6	No Action Recommended	N/A
95240	Beef Cattle Farm and Animal Waste	16	0.8	Need New Data	New measurements representative of current conditions
95398	Composite Profile - Oil and Natural Gas Production - Condensate Tanks	16(14)	0.3(0.2)	No Action Recommended	N/A
95420	Oil and Gas Production - Composite Profile - Glycol Dehydrator, Uinta Basin	16(13)	0.2(0.2)	No Action Recommended	N/A
3142	Consumer Products Composite: Adhesives and Sealants	N/A(8)	N/A(0.8)	Need New Data	New measurements based on current products
1001	Internal Combustion Engine – Natural Gas	N/A(11)	N/A(1.4)	Specificity	Specificity (internal combustion engines for natural gas and liquified petroleum gas, compressor engines used in onshore gas production, an SCC for a surface coating oven heater for liquified petroleum gas
8746	Composite Profile - Straw Burning	N/A(13)	N/A(0.7)	Specificity	SCC issue (wide range of agricultural burning SCCs)
4553	Meat charbroiling	N/A(14)	N/A(0.4)	No Action Recommended	N/A

Southwest and in the Northern Rockies and Plains. The most prevalent VOC profiles based on reactivity include emissions from prescribed fires and wildfires across much of the country, however, emissions from crude oil storage tanks dominate in the Northern Rockies and Plains while emissions from residential wood combustion are most prominent in the Northeast and in the Upper Midwest.

The SPECIATE database is routinely updated and air quality modeling systems using current speciation profiles generally produce reasonable estimates of ozone and $PM_{2.5}$ concentrations (Simon et al., 2012). However, opportunities for refinement and improvement remain and this analysis serves as the basis for future updates to the EPA emissions modeling platform and the SPECIATE database. Potential updates include: 1) reassigning SCCs to other, more appropriate existing profiles, 2) creating new profiles from existing data, and 3) searching the literature for additional data. This analysis is also intended to communicate the highest-priority needs to the research community and to promote further studies that provide measurements to create profiles for SPECIATE. Based on this analysis, new studies incorporated into the SPECIATE database should: 1) provide a complete set of speciation data for $PM_{2.5}$ or VOC (i.e. data should account for all VOC or $PM_{2.5}$ mass even if that involves reporting an 'unspeciated' or 'unresolved' fraction."), 2) be peer-reviewed, 3) be representative of current technology/conditions, 4) have a sound data collection quality methodology and sampling design (e.g. logical design, limitations and assumptions clearly stated, capture natural variability of source, use EPA testing methods (if applicable), document standard deviations), and 5) provide data expressed as an emission factor.

The most common concern for profiles in need of updates is their appropriateness for the sources to which they are applied. Many of these mapping mismatches are due to lack of emissions testing for appropriate source categories. The needs assessment ranking system identified $PM_{2.5}$ profiles for wildfires, agricultural burning, heavy-duty diesel vehicle exhaust (which is being applied to nonroad diesel equipment), sub-bituminous coal combustion and nonroad gasoline exhaust as the highest priority $PM_{2.5}$ profiles in need of new data. The assessment also found VOC emission source profiles for crude oil storage tank emissions, fireplace wood combustion, prescribed fires, and open burning in the most need of updates.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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