Response Letter to Reviewers for:

Reactive Organic Carbon Emissions from Volatile Chemical Products

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Thank you to the editor and reviewers for taking the time to consider our manuscript and provide helpful comments. These comments significantly improved the rigor and quality of our manuscript, and your time and efforts are much appreciated. All reviewer comments are addressed individually below, point-by-point, with the original comments featured in bold text and our response followed in non-bold text.

All updates to the original submission were tracked in the revised submission.

Anonymous Referee #1:

One of the main conclusions the authors make is that this new framework includes spatial allocation to regional and local scales. Have you compared this to current surrogates provided with the 2017 NEI, CARB surrogates or published work such as "Improving spatial surrogates for area source emissions inventories in California" by Li et al. 2020? How do the regional and local distributions vary with this approach? What is the level of resolution the census data is applied to? County/census block? Possibly adding a difference plot comparing to current estimates would be helpful.

We used the same spatial allocation methods as the 2017 NEI. This largely includes population and employment-based allocation, as outlined in Table S6 of the SI. For employment-based allocation, we match the employment NAICS codes used by the 2017 NEI, as outlined in the Technical Source Documentation of the inventory.

In addition, we are familiar with Li et al. 2020 and cite the paper in our manuscript. However, there is a key difference in their analysis and our applications. Namely, application of their spatial surrogates is at the sub-County level for photochemical modeling purposes, whereas we use spatial proxies only to downscale national-level emissions to the county-level.

Comparing possible inventory differences due to variable spatial surrogates, especially at the sub-County level, was outside the scope of this analysis. Nonetheless, leveraging the results from Li et al. 2020 would certainly be considered for those types of applications.

Figure 5c shows a high amount of emissions per capita in Colusa, CA – what is the driver behind this in a relatively small county?

Colusa County is sparsely populated, but relative to their population, features high agricultural pesticide usage. As a result, while small in magnitude (see Colusa County in Fig. 5b), their per-capita emissions are quite high.

Since observed data is available to do comparison, it would be beneficial to show a range of predicted VCP emissions for LA county of the 30 reported species. It is noted that the observed total is 0.259 g while the inventory total is 0.226g; can you add uncertainty to the inventory value based on the discussions from sector 3.6 and 4?

It would be difficult to confidently show a range of predicted individual compounds for the evaluation since the uncertainty associated with the evaporative organic composition of individual product types is not known or provided by the source data. Our uncertainty analysis using Monte Carlo simulations focused on the total emission magnitude and did not perturb the relative abundance of an individual species in a product composition. For example, we do not know the uncertainty associated with the assignment of x% of toluene within a product type. One sentence regarding this limitation has been added to Section 2.2 and are included below:

Furthermore, the uncertainty associated with the evaporative organic composition of individual product types is not known or provided by the source data.

Additional emissions that could result from the discussion in Section 3.6 would not yield a change to the inventory evaluation. Since the evaluation is exclusive to 30 compounds, and these compounds are not influenced by the volatility (or emission) of the organics that are assumed to be non-evaporative, the predicted 0.226 g (g CO)$^{-1}$ would not change. As for the uncertainties discussed in Section 4, these would be difficult to translate without (1) incorporation of a two-box model into the framework to account for indoor loss, which is planned for future work or (2) localized post-use control status, which was outside the scope of this analysis.

In Section 5. on line 562 "The 95% confidence interval for the national level emissions from the complete sector for 2016 is 2.68 – 3.60 Tg (1.81 – 2.42 TgC). This is consistent with the 2017 National Emission Inventory and half the emissions magnitude reported elsewhere (McDonald et al., 2018)." Can the authors provide the 2017 NEI values that are being compared? It would also be helpful to add a national difference plot showing the variability between this new method and 2017 NEI totals for the three panels on figure 5 (state, county, county/capita).

We have added 2017 NEI values (2.84 Tg) for comparative purposes to both Section 5 and Section 3.2. We also direct the referee to Figure S4c in the SI for a county-level per-capita difference plot between the VCPy inventory and the 2017 NEI.

<u>**Anonymous Referee #2:**</u>

What is the role of disposal in this framework? If VCPs that are used on a short timescale but are disposed of using open methods, is that included in the framework? For example, VCPs could enter wastewater treatment plants and enter the atmosphere.

In this framework, disposal is treated as a permanent sequestration of organics. Research does suggest that organics entering a wastewater treatment plant are largely removed through biodegradation or sorption to sludge (Shin et al. 2015). However, there is significant aeration at such plants, providing opportunity for air release. The NEI does report emissions from Publicly Owned Treatment Works, but given the massive quantity of organics that feature this down-the-drain fate (organics from shampoos, soaps, conditioners, detergents), even marginal changes in the emission of organics from VCPs through this route could have notable impacts.

The Monte Carlo analysis is focused on the uncertainty in the total emissions per capita as the primary outcome. The assumption here is the uncertainty lies primarily in the model inputs, and the outcome is deterministic. I expect this assumption to be valid for total emission per capita as the primary outcome, but may not be so if we examine the composition instead. For example, how does uncertainty in the composition profile affect the emissions?

We do partially include uncertainty in the composition of VCPs in our Monte Carlo assumptions. This is accomplished by assigning uncertainty in the evaporative organic proportions of each sub-PUC, as noted in Section 2.2. However, due to data limitations, we do not assign uncertainty to the composition of evaporative organics. Nonetheless, changes in the organic composition of the evaporative organics would result in changes in the volatility distribution of the organics, which is implicitly accounted for via assigned uncertainty to the characteristic evaporation timescale.

Similarly, for the input variables that were examined (e.g. uncertainty in v_e, depth), what is the uncertainty in the composition of emissions? E.g. what is the uncertainty in median c*? I expect that if v_e increases, it might increase emissions of lower volatility compounds more than it increases those of higher volatility ones.

This is a great question. With available data, the uncertainty associated with the composition of the emissions would be extremely difficult to determine. For example, we do not know the uncertainty associated with the assignment of x% of toluene within a product type. However, we can test the second question. Our median log(C*) for the emissions from the complete sector is 7.6 and the 95% uncertainty associated with that number due to the model inputs we can perturb (including v_e) is 7.5 – 7.8. This is certainly an underestimate in the uncertainty since, again, we do not know how the uncertainty associated with the composition of individual product types.

As for v_e, if this variable were increased, the characteristic evaporation timescale of all components would increase, assuming all other variables are held constant. If this were to happen, all compounds that are already predicted to evaporate (i.e. higher volatility ones) would still evaporate. However, as the reviewer points out, this could have notable impacts on lower volatility compounds that were previously predicted to not evaporate.

For many of the water-based VCPs, I would expect that evaporation will be based more on K_AW (or Henry's Law constant) rather than K_OA. How much would that change the estimates?

When using the QSAR predicted K_{OA} and K_{OW} and equation 3.4 of Weschler and Nazaroff (2008) for two compounds (propylene glycol and PCBTF), we yield a calculated K_{WA} that is larger for one (propylene glycol), indicating less partitioning to air, and one that is smaller (PCBTF), indicating more partitioning to air. However, the change in the subsequent characteristic evaporation timescale from such changes seems to be only relevant for VCPs that would feature small to medium use timescales (e.g. < 1 day). For VCPs with a long use timescale (e.g. architectural coatings), this change would not yield a different conclusion regarding the proportion of emissions.

What is the fraction of VCPs that are based on fossil-carbon vs modern carbon? Is that something that can be estimated?

This is difficult to determine. Some compounds emitted from VCPs (e.g. ethanol, many fragrances) are likely dominated by modern carbon, whereas others (e.g. mineral spirits) are likely dominated by fossil-carbon. However, it is our understanding that the demand for "green" solvents has and continues to grow. According to an industry study (The Freedonia Group, 2016), approximately 15% of functional solvent usage is derived from natural or renewable resources.

Lines 296-297: For 4 PUCs, employment statistics were used for the spatial allocation of commercial VCP emissions. I am wondering that regarding the automation considerations, the number of employees might underestimate the VCP emissions from those sites and sales allocation or GDP of the production sector distribution might be better tools for that purpose.

Since emission more likely occurs at or near the time of use, we prefer employment (for industrial sub-PUCs) as a spatial downscaling proxy over production sector statistics. In addition, larger production facilities are more likely to have add-on controls. While we did not consider add-on controls in this manuscript, emissions from these sources are likely non-linear with production volume.

It is not surprising that regional and localized differences are significant. For many of the compounds, the atmospheric lifetimes could be long enough that these differences probably do not matter too much. This might also depend on the scale of air quality modeling.

We agree.

In Lines 25-27 (abstract) and in Section 3.3, when comparing to 2017 NEI, the terms "increase" and "decrease" are misleading, since they can be confused with year-to year increase/decrease. I suggest using terms like "overestimate" or "underestimate", or just "higher" or "lower".

We adjusted both the text in the abstract and Section 3.3 to help alleviate this confusion. The two relevant sentences in the abstract now reads:

VCPy predicts more VCP emissions than the NEI for approximately half of all counties, with 5% of all counties featuring emissions more than 55% higher. Categorically, VCPy reports higher emissions for personal care products (150%) and paints/coatings (25%) when compared to the NEI, whereas pesticides (-54%) and printing inks (-13%) feature lower emissions.

Line 80: there are two references for Li et al. 2018.

We believe this is a mistake on the part of the reviewer. We refer to the first reference as "Li et al., 2018" in the text and the second reference as "Li and Cocker 2018," since there are only two authors of that manuscript.

Figure 6: what does X stand for?

The "X" represents each compound in the figure. A clarification has been added to the Fig. 6 caption.

<u>Additional edits:</u>

Following submission, we learned of more recent sales proportions for water vs. solvent-based architectural coatings in a California Air Resource Board survey. As a result, we updated the water vs. solvent-based proportions from 88/12% to 94/6% (i.e. the proportion of water-based coatings to total architectural coatings increased). These changes had marginal impacts on the complete inventory (~2%) and modest impacts on the architectural coatings sub-PUC (~25%).

Reactive Organic Carbon Emissions from Volatile Chemical Products

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Abstract. Volatile chemical products (VCPs) are an increasingly important source of anthropogenic reactive organic carbon (ROC) emissions. Among these sources are everyday items, such as personal care products, general cleaners, architectural coatings, pesticides, adhesives, and printing inks. Here, we develop VCPy, a new framework to model organic emissions from VCPs throughout the United States, including spatial allocation to regional and local scales. Evaporation of species in the VCPy framework is a function of the compound specific physiochemical properties that govern volatilization and the timescale relevant for product evaporation. We introduce the terms evaporation timescale and use timescale, respectively, to describe these processes. Using this framework, predicted national, per-capita organic emissions from VCPs are 9.~~7~~ 5 kg person^{-1} year^{-1} (6.~~5~~ 4 kgC person^{-1} year^{-1}) for 2016, which translates to 3.~~12~~ 05 Tg (2.~~10~~ 06 TgC), making VCPs a dominant source of anthropogenic organic emissions in the United States. Uncertainty associated with this framework and sensitivity to select parameters were characterized through Monte Carlo analysis, resulting in a 95% confidence interval of national VCP emissions for 2016 of 2.~~68~~ 61 – 3.~~60~~ 53 Tg (1.~~81~~ 76 – 2.~~42~~ 38 TgC). This nationwide total is broadly consistent with the US EPA's 2017 National Emission Inventory (NEI); however, county-level and categorical estimates can differ substantially from NEI values. VCPy predicts ~~larger~~ more VCP emissions than the NEI for approximately half of all counties, with 5% of all counties featuring ~~increases~~ emissions more than ~~> 60~~55% higher. Categorically, VCPy reports higher emissions for personal care products (150%) and paints/coatings (~~34~~25%) when compared to the NEI, ~~feature the largest increases,~~ whereas pesticides (-54%) and printing inks (-13%) feature ~~the largest decreases~~lower emissions. An observational evaluation indicates emissions of key species from VCPs are reproduced with high fidelity in the methods employed here (normalized mean bias of -13% with r = 0.95). Sector-wide, the effective secondary organic aerosol yield and maximum incremental reactivity of VCPs are 5.3% by mass and 1.~~59~~ 58 g O_3 g^{-1}, respectively, indicating VCPs are an important, and likely underrepresented to-date, source of secondary pollution in urban environments.

1 Introduction

Reactive organic carbon (ROC), which includes both non-methane organic gases and organic aerosol (OA), is central to atmospheric oxidant levels and modulates the concentration of all reactive species (Heald and Kroll, 2020; Safieddine et al., 2017). Gas-phase ROC features both biogenic and anthropogenic sources and, following oxidation, can lead to the formation of tropospheric ozone and secondary organic aerosol (SOA). Organic aerosol is often the dominant component of total fine particulate matter ($PM_{2.5}$) throughout the world (Jimenez et al., 2009; Zhang et al., 2007), and SOA is often the dominant component of OA in both urban and rural settings (Jimenez et al., 2009; Volkamer et al., 2006; Williams et al., 2010; Xu et al., 2015). Since ozone and $PM_{2.5}$ are both associated with impacts on human health and welfare (U.S. Environmental Protection Agency, 2019a; U.S. Environmental Protection Agency, 2020) that are global in nature (Burnett et al., 2018; Mills et al., 2018) and persist at low concentrations (Di et al., 2017; Kazemiparkouhi et al., 2020), accurately understanding the sources, magnitude, and speciation of organic emissions is critical.

Historically, the ~~dominant~~ leading source of anthropogenic organic emissions in the United States has been motor vehicles (Khare and Gentner, 2018; McDonald et al., 2013; Pollack et al., 2013). However, successful emission reduction strategies implemented over several decades have dramatically reduced mobile emissions (Bishop and Stedman, 2008; Khare and Gentner, 2018; McDonald et al., 2013), resulting in substantial declines in both ambient gas-phase non-methane volatile organic compounds (NMVOC) and OA concentrations (Gentner et al., 2017; McDonald et al., 2015; Pollack et al., 2013; Warneke et al., 2012). Due to these changes, volatile chemical products (VCPs) are now viewed as the foremost source of anthropogenic organic emissions (Khare and Gentner, 2018; McDonald et al., 2018). The U.S. EPA has long accounted for VCPs in the National Emissions Inventory (NEI) as the “solvent sector.” In 1990, the mobile and VCP sectors were the two highest emitters of volatile organic compounds (VOC; a regulatory defined collection of organic species that excludes certain compounds, such as acetone) at the national level. Mobile and VCP sources emitted 7.2 Tg and 5.0 Tg of VOCs, respectively (U.S. Environmental Protection Agency, 1995). By 2017, EPA estimates of VOC emissions from both the mobile and VCP sectors each dropped to 2.7 Tg (U.S. Environmental Protection Agency, 2020). For VCPs, factors driving the emissions decrease over this period include, but are not limited to, reformulation of consumer products (Ozone Transport Commission, 2016) and implementation of National Emissions Standards for Hazardous Air Pollutants regulations for industrial processes (Strum and Scheffe, 2016). Potentially complicating the trend and assessment of relative roles of different sectors, new inventory methods have suggested that VCP emissions in the NEI could be biased low by a factor of 2-3 (McDonald et al., 2018).

The decades-long increasing relative contribution of VCPs to total anthropogenic organic emissions could have several important implications for modelling and improving air quality. First, modelling studies of SOA from anthropogenic VOCs have generally focused on combustion sources (Hodzic et al., 2010; Jathar et al., 2017; Murphy et al., 2017), which are typically rich in aromatics and alkanes (Gentner et al., 2012; Lu et al., 2018). In contrast, emissions from VCPs occur through evaporation and contain large fractions of oxygenated species (e.g. glycol ethers, siloxanes), many of which feature uncertain SOA yields (McDonald et al., 2018). Second, adequate chemical mechanism surrogates for species common in VCPs (e.g. siloxanes) are lacking (Qin et al., 2020). As VCPs and their components could have significant SOA potential (Li et al., 2018; Shah et al., 2020), revisiting VCP emissions mapping to chemical mechanisms could help reduce modelled bias, which has historically been difficult to resolve (Baker et al., 2015; Ensberg et al., 2014; Lu et al., 2020; Woody et al., 2016). Third, VCPs feature substantial quantities of intermediate-volatility organic carbon (IVOC) compounds (CARB, 2019) and better representing their source strength could help resolve the high IVOC concentrations observed in urban atmospheres (Lu et al., 2020; Zhao et al., 2014). Fourth, if the VCP sector is systematically low biased in the NEI or select urban areas, there could be implications for ozone pollution (Zhu et al., 2019). Finally, reducing organic emissions from VCPs has traditionally been viewed through the lens of minimizing near-field chemical exposure (Isaacs et al., 2014) or mitigating ozone pollution (Ozone Transport Commission, 2018), both of which can be accomplished through product reformulation. For example, reducing the magnitude of regulatory VOC emissions from VCPs can be accomplished by reformulating a product with lower-volatility ingredients that are less likely to evaporate (Ozone Transport Commission, 2016). However, if these lower-volatility replacement ingredients eventually evaporate on atmospherically relevant timescales, they could be efficient SOA precursors (Li et al., 2018).

Given these ~~concerns~~implications, the need to understand and resolve differences among inventories becomes increasingly important. Here, we develop VCPy, a new framework to model organic emissions from VCPs throughout the United States, including spatial allocation to the county-level. In this framework, fate and transport assumptions regarding evaporation of a species in a product into ambient air are a function of the compound specific physiochemical properties that govern volatilization and the timescale available for a product to evaporate. We introduce the terms evaporation timescale and use timescale, respectively, to describe these processes. Since product ingredients are considered individually, determination of emission composition is explicit. This approach also enables quantification of emission volatility distributions and the abundance of different compound classes. In addition, we test the sensitivity of predicted emission factors to uncertain parameters, such as evaporation and use~~ and evaporation~~ timescales, through Monte Carlo analysis, evaluate the VCPy inventory using published emission ratios, and estimate the effective SOA and ozone formation potential of both the complete sector and individual product use categories.

2 Methods

2.1 VCPy: A Framework for Estimating Reactive Organic Carbon Emissions from Volatile Chemical Products

The VCPy framework is based on the principle that the magnitude and speciation of organic emissions from VCPs are directly related to (1) the mass of chemical products used, (2) the composition of these products, (3) the physiochemical properties of their constituents that govern volatilization, and (4) the timescale available for these constituents to evaporate (Fig. 1). ~~VCPy attempts to address each of these points by utilizing the most relevant datasets available.~~ Since the VCP sector includes residential, commercial, institutional, and industrial sources, a consistent stream of data sources for all product categories is difficult. As such, this work implements a hybridized methodology that utilizes the best features of prior emission inventory methods, while introducing new methods to make improvements where necessary. The result produces national-level, per capita emission factors for all product categories in the VCP sector that can be further tailored for regional or localized analysis. The per capita basis is useful for comparison across frameworks and over time, but emissions can be recast in other units as needed. Briefly, survey data are used to generate a 1st-order product composition profile for a composite of product types, which quantifies the fraction of organic, inorganic, and water components. The organics component is further divided into individual species (e.g. ethanol, isobutane, isopropyl alcohol). A variety of data sources are used to estimate the national-level product usage and each composite is assigned a use timescale, reflecting the elapsed time between use and any explicit removal process. Finally, the characteristic evaporation timescale of each organic component is calculated using quantitative structure-activity relationship (QSAR) modelled physiochemical properties and compared to the assigned use timescale. If the characteristic evaporation timescale of the organic component is less than the assigned use timescale of the composite, it is assumed that the compound is emitted. Else, the compound is retained in the product or other condensed phase (e.g. water) and permanently sequestered.

2.1.1 Product Use Categories (PUCs) and sub-Product Use Categories (sub-PUCs)

VCPy disaggregates the VCP sector into several components called Product Use Categories (PUCs). An individual PUC is not exclusively used in a singular setting (e.g. residential vs. commercial) and examples include Personal Care Products, Cleaning

Products, and Paints & Coatings. PUCs are further divided into sub-PUCs, which are composites of individual product types featuring similar use patterns. In addition to permitting tailored fate-and-transport assumptions, similar hierarchical product schema are also useful for models estimating near-field exposure to chemicals, through routes such as dermal contact and indoor inhalation (Isaacs et al., 2020). As an example, there are two sub-PUCs allocated to the Personal Care Product PUC: Short Use Products and Daily Use Products. These two sub-PUCs are differentiated by the length of use prior to removal (i.e. the use timescale). The mass of chemical products used and subsequent organic emission factors, which are the main output from VCPy, are calculated at the sub-PUC level (Fig. 1). Currently, there are ~~nine~~ ten PUCs and sixteen sub-PUCs implemented in VCPy (Table 1).

2.1.2 National-Level Product Usage

To estimate VCP product use, some prior work has used national economic statistics, such as market sales or shipment values (e.g. U.S. Environmental Protection Agency, 2020; McDonald et al., 2018). Others have incorporated product usage statistics based on consumer habits and practices (e.g. Isaacs et al., 2014; Qin et al., 2020), but these statistics are generally unavailable for commercial and industrial chemical usage, which limits their application. To better ensure the capture of all chemical product usage, including usage in residential, commercial, institutional, and industrial settings, national economic statistics are utilized, where possible (Table S1).

Product usage from twelve sub-PUCs is estimated using national-level shipment statistics, commodity prices, and producer price indices. National-level economic statistics are retrieved from the U.S. Census Bureau's Annual Survey of Manufactures (ASM; U.S. Census Bureau, 2016a), which provides annual statistical estimates for all manufacturing establishments nationally. Values are available for all 6-digit North American Industry Classification System (NAICS) codes, provided as product shipment values (\$ year^{-1}), and are reported with associated relative standard errors (generally < 5%). To translate shipment values (\$ year^{-1}) to usage (kg year^{-1}), we use commodity prices (\$ kg^{-1}) from the U.S. Department of Transportation's 2012 Commodity Flow Survey (U.S. Department of Transportation, 2015). An exception is for all Paint & Coating sub-PUCs. Commodity prices for these sub-PUCs are taken from the U.S. Census Bureau's Paint and Allied Products Survey (U.S. Census Bureau, 2011a) and representative of 2010. To translate these commodity prices, which are from 2010 and 2012, to values reflective of 2016, we use producer price indices reported by the Federal Reserve Bank of St. Louis (U.S. Bureau of Labor Statistics, 2020). Commodity price indices from the Federal Reserve Bank are updated for all NAICS manufacturing codes monthly, which we average to create annual price indices (Table S2). An implicit assumption in this methodology is that manufacturing and product usage are, on average, annually balanced.

We preferentially utilize product usage numbers derived from the above methodology, when possible, as all data sources have the following characteristics: (1) they are nationally derived and therefore less influenced by regional differences in manufacturing and formulation, and (2) all datasets are freely available to the public. However, due to data limitations, product usage for four sub-PUCs are estimated using other sources. The Dry Cleaning and Oil & Gas product usage estimates are derived from the national-level solvent mass usage reported by an industry study (The Freedonia Group, 2016). The Miscellaneous Products and Fuels & Lighter product usage estimates are derived from reported sales data, specific to California, from the California Air Resources Board's 2015 Consumer and Commercial Products Survey Data (CARB, 2019). These sales numbers are scaled upwards to a national-level by assuming equivalent per-capita product usage.

2.1.3 1st-Order and Organic Product Composition

Each sub-PUC features two composite profiles. The initial composite is the 1st-order product composition profile, which disaggregates the total mass of each sub-PUC into its water, inorganic, and organic fractions (Table 2). Total organics are further decomposed into non-evaporative and evaporative organics. The quantification and accounting of evaporative organics in this framework are necessary as CARB's organic profiles are processed to exclude organics that are not anticipated to evaporate on atmospherically relevant timescales. For ten sub-PUCS, the 1st-order product composition profile uses data from the California Air Resources Board's 2015 Consumer and Commercial Products Survey (CARB, 2019). Various product types are sorted into each sub-PUC and the 1st-order product composition profiles are calculated on a weighted basis using the reported sales from manufacturers and formulators in California. Due to omissions stemming from confidentiality concerns, not all sales and composition data from the survey are available. We utilize the publicly available portions of the data, which constitutes most of the survey and includes over 330 product types. For example, 126 product types and 20 product types were sorted into the General Cleaners and Adhesives & Sealants (Table S3) sub-PUCs, respectively.

For Architectural Coatings, Industrial Coatings, and Printing Inks, the 1st-order product composition profile is derived from data in the California Air Resources Board's 2005 Architectural Coatings Survey (CARB, 2007). The Architectural Coatings sub-PUC uses data from all profiles in the survey, which is dominated by flat paint, non-flat paints, and primers. Industrial Coatings and Printing Inks use the 1st-order product composition profiles of Industrial Maintenance coatings and Graphic Arts coatings, respectively. The 1st-order product composition profile for aerosol coatings uses data from the California Air Resources Board's 2010 Aerosol Coatings Survey (CARB, 2012), which includes more than 20 aerosolized product types. Only the evaporative organic composition of ~~these~~ aerosol coating products was ~~provided~~reported, so the remaining mass was evenly split between water and inorganics. For Dry Cleaning and Oil & Gas, as the product usage for these sub-PUCs were derived from the organic functional solvent mass usage, it is assumed that this mass is entirely evaporative organics.

The second composite is the organic composition profile. Again, the California Air Resources Board's 2015 Consumer and Commercial Products Survey (CARB, 2019) was used to derive a composite of product types~~the composition of organics~~ for ten sub-PUCs (Table S4). ~~Within each sub-PUC, all~~These product types are then mapped to an associated organic profile (CARB, 2018; see Table S3) and weighted based on their evaporative organic contributions to the total sub-PUC. For Architectural Coatings, a~~n~~ ~~88~~94% water-based and ~~12~~6% solvent-based paint (CARB, ~~2007~~2014) composite is generated. Aerosol Coatings are calculated on a weighted basis using the potentially evaporative organic contributions reported by CARB's 2010 Aerosol Coatings Survey (CARB, 2012). The organic composition profiles for Industrial Coatings, Printing Inks, and Dry Cleaning all utilize profiles (3149, 2570, 2422, respectively) from EPA's SPECIATEv5.0 database (EPA, 2019b). Approximately 65% of the solvents used in the Oil & Gas sector are alcohols and the remainder are a broad range of hydrocarbons (The Freedonia Group, 2016). Since detailed composition data for Oil & Gas solvents are sparse, all Oil & Gas alcohols are assumed to be methanol, as it is widely used in and emitted from Oil & Gas operations (Lyman et al., 2018; Stringfellow et al., 2017; Mansfield et al., 2018). The remaining 35% is allocated to naphtha, a blend of hydrocarbon solvents.

Several components within CARB profiles are lumped categories or complex mixtures. This includes naphtha, mineral spirits, distillates, Stoddard Solvent, fragrances, volatile methyl siloxanes, and a series of architectural coating and consumer product "bins." All naphtha, mineral spirits, distillates, and Stoddard Solvent occurrences in individual profiles are treated as a single mineral spirits profile (Carter, 2015). Volatile methyl siloxanes include several compounds (e.g. D_4, D_5, D_6), all of which are emitted in varying proportions (Janechek et al., 2017). Here, the lumped volatile methyl siloxane identity is preserved but the physiochemical properties of decamethylcyclopentasiloxane is applied to the surrogate. Fragrances are a diverse mixture of organic compounds that include many terpenes and alkenes (Nazaroff and Weschler, 2004; Sarwar et al., 2004; Singer et al., 2006b). However, since the proportion of these constituents are unknown, all fragrances are physically treated as d-limonene since it is the most prevalent terpene emitted from fragranced products (Sarwar et al., 2004; Singer et al., 2006b). Finally, for the architectural coating and consumer product "bins," we use the representative chemical compositions derived by Carter, 2015.

2.1.4 Controls

There are two methods for controlling organic emissions from VCPs. The first method is through product reformulation, which would occur prior to product usage. Strategies that fit this definition include switching from a hydrocarbon solvent-based ingredient to one that is water-based, replacing an organic component with a ~~increasing the proportion of~~ non-organic component~~s in a product~~, and reformulating a product with lower-volatility ingredients that are less likely to evaporate (Ozone Transport Commission, 2016). VCP emissions that stem from residential, commercial, and institutional settings rely on these pre-use controls to reduce emissions. ~~Regulations~~ Regulators often set VOC content limits for chemical products (e.g. national standards: Section 183(e) of the Clean Air Act; 40 CFR 59), with California (e.g. CARB – Title 17 CCR) typically setting some of the most stringent limits in the country (Ozone Transport Commission, 2016). As the 1^{st}-order and organic composition profiles utilized here are almost exclusively derived from product composition data, pre-use controls are implicitly represented. In fact, since the product composition data is from manufacturers and formulators in California, where product VOC content limits are typically more stringent than national regulations, applying these profiles nationally likely results in conservative assumptions.

The second pathway of controlling organic emissions from VCPs is through post-use controls. Strategies that fit this definition include add-on controls, manufacturing process modifications, and disposal techniques. Add-on control strategies and manufacturing process modifications are limited to industrial and commercial emission sources, such as Industrial Coating (U.S. EPA, 2007; U.S. EPA, 2008) and Printing Ink (U.S. EPA, 2006a; U.S. EPA, 2006b) facilities. Since adoption of these technologies vary widely in space and time, assigning ~~a single~~ post-use controls via these strategies ~~efficiency~~ is not considered here. As several of these industrial sources (e.g. coatings, printing inks, dry cleaning) feature controls, as required by Section 112 of the Clean Air Act (40 CFR 63), this assumption could lead to localized high bias and will be refined in future work. Here, we only consider post-use controls through disposal techniques for the Oil & Gas and Fuels & Lighter sub-PUCs. For Oil & Gas, we assume that the solvents used in these processes become entrained in the produced water at these sites. Since produced water is largely (~89-98%) reinjected for enhanced oil and gas recovery or disposal (Lyman et al., 2018; Liden et al., 2018), we apply a post-use control efficiency of 94% (i.e. average of reported reinjection rates) to this sub-PUC. However, it should be noted that reinjection frequency and solvent usage can vary regionally. For Fuels & Lighters, we assume 90% of the organics are destroyed through combustion upon use (CARB, 2019).

2.1.5 Evaporation and Use-Timescales

Fate-and-transport in the VCPy framework is a function of the predicted compound specific evaporation timescale and the assigned use timescale of each sub-PUC. It should be noted that this methodology explicitly results in the organic speciation of emissions differing from the organic composition of products from which they volatilize. For example, the composition of organics within a product may differ from the speciation of emitted organics if the product contains low-volatility compounds that do not evaporate on relevant timescales.

The evaporation timescale is the compound specific (i.e. independent of the sub-PUC of interest), characteristic timescale of emission from a surface layer and is calculated using previously published methods (Khare and Gentner, 2018; Weschler and Nazaroff, 2008). This timescale is defined as a relationship between the mass of a compound applied and the rate of its emission, which can be expressed by:

$$Evaporation\ Timescale\ [hr] = {M_{applied}}/{R_{emission}} = {K_{OA} \times d}/{v_e} \quad (1)$$

where K_{OA} is the octanol-air partitioning coefficient of the compound, d [m] is the assumed depth of the applied product layer, and v_e [m/hr] is the mass transfer coefficient of the compound from the surface layer into the bulk air, which is a function of aerodynamic and boundary layer resistances. Median values for d [0.1 mm] and v_e [30 m/hr] from Khare and Gentner (2018) are selected here. It should be noted that v_e can vary substantially based on outdoor vs. indoor atmospheric conditions and future work will incorporate a two-box model to better account for such differences. A compound's K_{OA} it is the ratio of an organic chemical's concentration in octanol to the organic chemical's concentration in air at equilibrium. It is often used to quantify the partitioning behaviour of an organic compound between air and a matrix. As experimental values of K_{OA} are sparse, modelled estimates from the quantitative structure-activity relationship (QSAR) model OPERA (Mansouri et al., 2018) are used here. All physiochemical properties, including OPERA results, are retrieved from the U.S. EPA's CompTox Chemistry Dashboard (https://comptox.epa.gov/dashboard; last access: August 31, 2020).~~ While simple in setup, the assumptions adopted here broadly capture the relevant characteristic evaporation timescale for each compound.~~

Use timescale is the timescale available for a sub-PUC to evaporate and is based on the length of its direct use phase (i.e. the elapsed time between application and any explicit removal process). As this value is subjective, broad values are applied to each sub-PUC (Table S5). For example, it is assumed that all products used in the bath and shower are quickly sequestered and washed down the drain, thus largely unavailable for emission (Shin et al., 2015). As such, Short Use Products are assigned a "Minutes" use timescale. In contrast, it is assumed that each person bathes once a day. Therefore, all Daily Use Products are assigned a "Days" use timescale.

Emissions are determined by comparing the calculated evaporation timescale for each component with the assigned use timescale for the sub-PUC. If the use timescale for the sub-PUC is greater than the evaporation timescale for a compound, the compound is emitted. Else, the compound is retained in the product or other condensed phase and permanently sequestered. Overall, organic emissions (E) for the complete sector are calculated as a summation over all organic compounds, i, and sub-PUCs, j, as follows:

$$E = \sum_{i,j} \begin{cases} 0 & if\ Use\ Timescale_j < Evaporation\ Timescale_i \\ U_j \times f_{E_j} \times f_{S_{i,j}} \times \left(1 - f_{C_j}\right) & if\ Use\ Timescale_j \geq Evaporation\ Timescale_i \end{cases} \quad (2)$$

where U is the product usage (Table 1), f_E is the evaporative organic fraction (Table 2), f_S is the fraction of an organic compound in the evaporative organics portion of a sub-PUC (Table S4), and f_C is the fraction of emissions that feature post-use controls on a mass basis. Application of Eqn. 2 determines the difference between organic product composition and organic emissions speciation.

2.2 Uncertainty Analysis

The sensitivity of emission estimates to a variety of input variables are tested through a systematic Monte Carlo analysis. We perform 10,000 simulations where product usage, evaporative organic proportions, variables associated with the characteristic evaporation timescale, the assigned use timescale, and post-use control assumptions are tested, both individually and ~~as a group~~collectively. For product usage, the primary sources of uncertainty are shipment values provided by the ASM, commodity prices, the balance of imports (including tourism) and exports, and unused product disposal. The ASM provides standard error estimates for most shipment values and are typically less than 5%. Uncertainty estimates are not provided for commodity prices and national-level exports generally outweigh traditional imports for most sub-PUCs (~2-15%; U.S. Census Bureau, 2016), but there are also imports of personal care products through tourism. Therefore, we ~~conservatively~~ assume there is a ±25% uncertainty (95% CI) ~~to~~ for all product usage estimates. CARB does not provide uncertainty estimates associated with the composition of product types or sales proportions. To account for these uncertainties, as well as the uncertainties associated with generating composites, we assume there is a ±25% uncertainty (95% CI) for all "Evaporative Organic" (Table 2) proportions. For the characteristic evaporation timescale, there are several layers of uncertainty. Application patterns vary by product type, which impacts assumptions regarding the depth of the chemical layer. In addition, indoor vs. outdoor product use and application of products to variable surface types (e.g. absorbing vs. non-absorbing) can impact mass transfer rates. As such, we apply broad uncertainties for variables associated with the characteristic evaporation timescale. We assume d (i.e. the depth of the applied chemical layer) is lognormally distributed with a median value of 0.1 mm (95% CI ~ [0.01 mm – 1 mm]) and v_e (i.e. the mass transfer coefficient) is normally distributed with a mean value of 30 m/hr (95% CI = [10 m/hr – 50 m/hr]). Since use timescales are categorical (e.g. minutes, days, years), we apply uncertainty by assuming the 95% CI of the assigned use timescale features a ±1 categorical uncertainty (e.g. mean: minutes; 95% CI = [seconds – hours]). Finally, for non-zero, post-use controls, we assume a ±25% uncertainty (95% CI) in the post-use control efficiency. It should be noted that ~~A~~additional avenues of uncertainty ~~may~~ likely persist but are difficult to quantify and therefore not included here. For example, due to the scarcity of large-scale product surveys, many of the 1st-order product composition profiles (e.g. Architectural Coatings) and organic profiles (e.g. Printing Inks) used in this analysis are more than a decade old. As a result, the proportion of organics in these product types and their organic components (i.e. the mean values applied here) may have changed in the interim period. Furthermore, the uncertainty associated with the evaporative organic composition of individual product types is not known or provided by the source data.

2.3 Spatial Allocation of National-Level Emissions

Emissions are calculated at the national-level and spatially allocated to the county-level using several proxies. Ten sub-PUCs, including all Cleaning Products and Personal Care Products, are allocated using population (Table S6; U.S. Census Bureau, 2020). Four sub-PUCs (Industrial Coatings, Allied Paint Products, Printing Inks, Dry Cleaning), all typically industrial in nature, are allocated using county-level employment statistics from the U.S. Census Bureau's County Business Patterns (U.S. Census Bureau, 2018). The employment mapping scheme for these four sub-PUCs utilize the methods from the 2017 NEI (U.S. EPA, 2020). On occasion, data in the County Business Patterns (CBP) is withheld due to confidentiality concerns. In those instances, we take the mid-point of the range associated with each data suppression flag. For Agricultural Pesticides, emissions are allocated based on county-level agricultural pesticide use and again taken from the 2017 NEI (U.S. EPA, 2020). Oil & Gas emissions are allocated using oil and gas well counts (U.S. EIA, 2019).

2.4 Inventory Evaluation

Previously published emission ratios from the Los Angeles basin during the summer of 2010 (de Gouw et al., 2018; de Gouw et al., 2017) are used to evaluate the VCPy emissions inventory (Table S7). Emissions ratios are generated by post-processing observed concentrations of organic gases, typically normalized to carbon monoxide (CO) or acetylene, to a period of "no chemistry" (Borbon et al., 2013; de Gouw et al., 2005; Warneke et al., 2007). As the air parcel is not photochemically aged (i.e. "no chemistry"), it is an ideal tool for evaluating an emissions inventory. An important caveat is that this method assumes the species being used for normalization (e.g. CO) is accurately inventoried and measured.

Since the emission ratios are not specific to a sector and represent total emissions, all other sectors must be quantified and speciated. For this purpose, all non-VCP anthropogenic emissions from the 2017 NEI (U.S. EPA, 2020) are collected and speciated using EPA's SPECIATEv5.0 database (EPA, 2019b; Table S8). This includes all on road, nonroad, nonpoint, and point sources. All VCP emission from the 2017 NEI are also collected and speciated for supplementary evaluation. In addition, biogenic emissions of ethanol, methanol, and acetone for May and June of 2016, as simulated by the Biogenic Emission Inventory System (Bash et al., 2016), were included to capture non-anthropogenic sources of these compounds. May and June were selected to coincide with the observational sampling months (de Gouw et al., 2018; de Gouw et al., 2017). As the observed emission ratios are specific to the Los Angeles basin, we derive all VCPy inventory emission ratios using data for Los Angeles County. Total CO emissions, including all on-road, non-road, non-point, and point sources, for Los Angeles County in 2017 are ~320 Gg. While the observed and VCPy inventory emission ratios are separated by 6-7 years, the ambient non-methane hydrocarbon to CO concentration ratio in Los Angeles has been consistent for several decades, indicating changes in emission controls feature similar improvements for both pollutants over time (McDonald et al., 2013). In addition, the magnitude of observed emission ratios for a given region do not appreciably change over marginal time horizons (Warneke et al., 2007).

2.5 Air Quality Impact Potential

Each organic compound is assigned a SOA yield and Maximum Incremental Reactivity (MIR) to facilitate an approximation of the potential air quality impacts of VCPs. For SOA, a wide collection of published yields, including both chamber results and prediction tools, were utilized (Fig. S1). These include: (1) all linear alkanes use a quadratic polynomial fit to the volatility basis set (VBS) data from Presto et al., 2010 at 10 μg/m^3; (2) all cyclic alkanes use linear alkane yields that are three carbons larger in size (Tkacik et al., 2012); (3) all branched alkanes use yields obtained from the Statistical Oxidation Model (SOM; Cappa and Wilson, 2012), as reported in McDonald et al. (2018); (4) benzene and xylenes use the average yields from Ng et al., 2007 under high-NO_x conditions; (5) toluene uses the average from Ng et al., 2007 under high-NO_x conditions and the VBS data from Hildebrant et al., 2009 at 10 μg/m^3; (6) all alkenes use yields obtained from SOM, as reported in McDonald et al. (2018); (7) volatile methyl siloxanes use the two-product model parameters from Janecheck et al., 2019, which includes additional SOA yields from Wu and Johnson 2017, at 10 μg/m^3; (8) all glycol ethers use chamber results and molecular structure relationships from Li and Cocker 2018 for reported and unreported glycol ethers, respectively; (9) benzyl alcohol uses the average of the ~~lower~~ lower-bound yields reported by Charan et al., 2020; (10) all remaining non-cyclic oxygenates, where available, use the arithmetic average of SOM results and a 1-D VBS approach, as reported by McDonald et al., 2018; (11) all remaining cyclic oxygenates, where available, use yields obtained from SOM, as reported by McDonald et al., 2018; (12) all halocarbons and compounds with less than five carbons are assigned a yield of zero; and (13) all remaining species are conservatively assigned a yield of zero if the effective saturation concentration (i.e. $C^* = (P^{vap} \times MW)/(R \times T)$) is $\geq 3 \times 10^6$ μg/m^3 and assigned the same yield as n-dodecane if the effective saturation concentration is $< 3 \times 10^6$ μg/m^3. The MIR of each compound, which measures the formation potential of ozone under various atmospheric conditions where ozone is sensitive to changes in organic compounds (Carter, 2010b), is calculated using the SAPRC-07 chemical mechanism (Carter, 2010a) and expressed as a mass of additional ozone formed per mass of organic emitted (Carter, 2010b).

3 Results and Discussion

3.1 National-Level PUC and sub-PUC Emissions

National-level, per-capita organic emissions from VCPs are 9.~~7~~5 kg person^{-1} year^{-1} (6.~~5~~4 kgC person^{-1} year^{-1}) for 2016 (Table 3), which translates to 3.~~12~~05 Tg (2.~~10~~06 TgC). When filtered to remove regulatory exempt organics, total emissions from VCPs are 2.6 Tg of VOC. In comparison, the 2017 NEI reports a combined total of 2.6 Tg of VOC emissions for on-road mobile, non-road mobile, and other mobile (i.e. aircraft, commercial marine vessels, and locomotives) sources, respectively. Therefore, when measured as VOC, the VCP sector is equal in magnitude to the sum of all mobile sources nationally, which is broadly consistent with the national-level emissions estimate from the 2017 NEI. Categorically, emission factors are largest for Paints & Coatings, which total 3.~~4~~1 kg person^{-1} year^{-1} (2.~~3~~2 kgC person^{-1} year^{-1}) and are approximately ~~35~~33% of the total sector (Table 3). The next largest PUCs are Personal Care Products and Cleaning Products, which contribute 2.1 kg person^{-1} year^{-1} (~~21~~22%) and 2.0 kg person^{-1} year^{-1} (~~20~~21%), respectively. Printing Inks, Adhesives & Sealants, and Pesticides each account for 6-~~8~~9% each, and the remaining PUCs contribute less than 2% in total.

For the complete sector (Fig. 2), the most abundantly emitted compound class are oxygenated species (53%), followed by alkanes (31%; including straight-chained, branched, and cyclic), aromatics (8%), alkenes (5%), and halocarbons (3%). Individually, organic emissions are dominated by ethanol (Daily Use Products, General Cleaners), acetone (Paints & Coatings, General Cleaners), isopropyl alcohol (Daily Use Products, General Cleaners), toluene (Paints & Coatings, Adhesives & Sealants), n-tetradecane (Printing Inks), fragrances (Daily Use Products, General Cleaners), propane (Aerosol Coatings, Industrial Coatings), and volatile methyl siloxanes (Daily Use Products, Adhesives & Sealants). Each of these species comprise > 3% of total VCP organic emissions (see Table S9 for the top-200 emitted compounds).

In terms of volatility classification (Donahue et al., 2012), as determined by the effective saturation concentration (i.e. C^*), total emissions are predominately VOCs ($C^* > 3 \times 10^6\ \mu g\ m^{-3}$), but there are also considerable contributions from IVOCs ($3 \times 10^2\ \mu g\ m^{-3} < C^* < 3 \times 10^6\ \mu g\ m^{-3}$; Fig. 2-3). IVOC emissions, which are efficient SOA precursors (Chan et al., 2009; Presto et al., 2010), are approximately 20% of total emissions. Of this 20% that are IVOCs, ~~55~~52% are oxygenated compounds (~~mainly~~ e.g. Texanol™, propylene glycol, ~~and~~ ethylene glycol, siloxanes, benzyl alcohol, and glycol ethers), ~~27~~30% are n-alkanes, and the rest are largely branched and cyclic alkanes. The prominence of oxygenated IVOC emissions ~~(e.g. siloxanes, benzyl alcohol, glycol ethers)~~ from VCPs is noteworthy, as SOA yields from these compounds have not historically been evaluated nor included as SOA precursors in model chemical mechanisms (Qin et al., 2020). However, work has been undertaken in recent years to better understand these compounds (e.g. Wu and Johnson 2017; Li and Cocker 2018; Janechek et al., 2019; Charan et al., 2020). Overall, Paints & Coatings is the largest source of IVOC emissions (~~~920~~ 760 g person^{-1} year^{-1}; Fig. 3), followed by Printing Inks (~350 g person^{-1} year^{-1}), Cleaning Products (~180 g person^{-1} year^{-1}), and Pesticides (~170 g person^{-1} year^{-1}). While Paints & Coatings emit more IVOCs by mass than all other PUCs, Printing Ink and Pesticide emissions both feature greater proportions of IVOCs to their total emissions (~44% and ~~~29~~28%, respectively).

These results also highlight how emissions from each PUC and sub-PUC are uniquely driven by mass of products used, organic composition, and use timescale. For example, the two largest sub-PUC sources are Daily Use Products and General Cleaners. Both are assigned a use timescale of 24-hr, but 40.6% of Daily Use Products are organic while General Cleaners are overwhelming composed of water (Table 2) and the annual mass usage of General Cleaners is ~3x higher than Daily Use Products (Table 1). As a result, net emissions of General Cleaners are within 10% of those from Daily Use Products (1.85 kg person^{-1} year^{-1} and 2.~~03~~04 kg person^{-1} year^{-1}, respectively). The emissions of Short Use Products, which is assigned a "Minutes" use timescale, can further illustrate the importance of considering fate-and-transport. Under these use timescale assumptions, only high volatility compounds (i.e. $C^* > 3 \times 10^7\ \mu g/m^3$) are emitted and a majority (~97%) of its organics are retained (Table 3). Besides Daily Use Products and General Cleaners, all remaining sub-PUCs emit ≤ 1.14 kg person^{-1} year^{-1}, with six emitting less than 0.1 kg person^{-1} year^{-1} (Table 3). Generally, sub-PUCs with low emissions stem from minimal use (e.g. Misc. Products), short use timescales (e.g. Short Use Products), or high control assumptions (e.g. Oil & Gas, Fuels & Lighter).

3.2 Uncertainty Analysis of National-Level Emission Factors

Uncertainty associated with product usage, proportion of evaporative organics, assumptions related to evaporation and use timescale, and post-use controls, where applicable, result in a total sector-wide emission uncertainty of ±15% (Fig. 4; 9.~~7~~5 kg person^{-1} year^{-1} [95% CI: 8.~~3~~1 – ~~11.2~~10.9]). Interestingly, the interaction of evaporation and use timescales can result in a threshold effect, where small changes in either do not necessarily translate into changes in the magnitude of emissions for a given sub-PUC (Fig. S2). For many PUCs, such as Paints & Coatings, Adhesives & Sealants, and Printing Inks, the use timescale is sufficiently long (i.e. years) for all evaporative organics to evaporate, regardless of the uncertainty associated with the evaporation and use timescales. Under such conditions, only uncertainty in product usage and product composition affect uncertainty in the emission magnitude. As a result, these two variables are the largest drivers of uncertainty for the complete sector (Fig. S2). However, uncertainties associated with evaporation and use timescale assumptions can be important for certain sub-PUCs with moderate to low use timescales (see Cleaning Products in Fig. S2). For example, Detergents & Soaps is assigned a "Minutes" use timescale, which results in a 0.12 kg person^{-1} year^{-1} emission factor (Table 3). If the use timescale for this sub-PUC was changed ~~to 1-hr~~"Hours," the emission factor would increase by a factor of 5.

From a national emissions perspective, these Monte Carlo results contain several important results. First, as mentioned above, the largest drivers of uncertainty are associated with a sub-PUC's usage and composition, not assumptions related to fate-and-transport (i.e. evaporation and use timescales). Second, the most uncertain PUCs are Cleaning Products, Personal Care Products, and Paints & Coatings, and their uncertainty generates a significant amount of emissions potential. The 95% confidence interval for all three span > 1.~~3~~24 kg person^{-1} year^{-1}, which is equivalent to > 400 Gg of organic emissions per year. Finally, the 95% confidence interval for the national level emissions from the complete sector for 2016 is 2.~~7~~6 – 3.~~6~~5 Tg (1.8 – 2.4 TgC), which is broadly consistent with the US EPA's 2017 NEI (2.8 Tg) and, largely due to differences in predicted evaporation, approximately half the emissions magnitude reported elsewhere (McDonald et al., 2018).

3.3 State and County-Level Emissions Allocation

The magnitude of VCP emissions varies substantially throughout the country, with the most populated states and counties featuring the highest ROC emissions (Fig. 5). California (~~358~~349 Gg), Texas (~~253~~247 Gg), and Florida (~~177~~173 Gg) are the largest state-level emitters and contribute ~25% of all VCP emissions. In contrast, the 30 smallest state-level emitters (plus Washington, DC) together emit $\sim$~~800~~780 Gg. At the county-level, Los Angeles County, Cook County (Chicago), and Harris

County (Houston) are the largest emitters. However, after normalizing by population, these three counties all feature per-capita emissions (8.~~42~~21, ~~9.09~~8.88, and 8.~~97~~ 76 kg person^{-1} year^{-1}, respectively) less than the national average (9.~~67~~ 45 kg person^{-1} year^{-1}) due to less industrial activity.

National spatial variability in per-capita emissions are largely driven by sub-PUCs tied to industrial and commercial activity (Fig. 5c). These sub-PUCs include Allied Paint Products (1.14 kg person^{-1} year^{-1}), Industrial Coatings (1.04 kg person^{-1} year^{-1}), Printing Inks (0.80 kg person^{-1} year^{-1}), Agricultural Pesticides (0.53 kg person^{-1} year^{-1}), and Oil & Gas (0.08 kg person^{-1} year^{-1}). The employment proxies for Allied Paint Products, Industrial Coatings, and Printing Inks are usually consistent with the underlying population (Fig. S3), with peaks in California, Texas, Florida, New York, and the industrial Midwest. In contrast, emissions from Agricultural Pesticides and Oil & Gas drive the large per-capita emissions in the Midwest and Great Plains (Fig. 5c). Emissions from these two sub-PUCs are heavily concentrated in the central United States (Fig. S3), including North Dakota, South Dakota, Iowa, Nebraska, Kansas, and Oklahoma. Collectively, these states contain < 4.5 % of the United States population but 24.1% and 17.5% of the Agricultural Pesticides and Oil & Gas VCP emissions, respectively. Both sub-PUCs also contribute to atypically high per-capita emissions in other States, such as Texas, Colorado, Idaho, and Wyoming.

While national VCP emissions from the 2017 NEI and the VCPy inventory are broadly consistent, county-level and categorical estimates can differ substantially between the two (Fig. S4). For example, VCPy reports > 35% lower emissions for 5% of all counties and > 55% higher emissions for ~~feature a decrease of > 35% and~~ another 5% ~~feature an increase of > 60%~~of all counties. When compared to the 2017 NEI, the states with the ~~largest~~ greatest emissions increases were Delaware, California, and Colorado, and the States with the greatest ~~largest~~ emissions decreases were North Dakota and South Dakota. There are also many spatial similarities between the two inventories. Both feature peaks in per-capita emissions over the Midwest and Great Plains (Fig. S4) and approximately half of all County-level emissions in the VCPy inventory are within ~~14~~15% of their value in the 2017 NEI. To compare the two inventories categorically, all product use categories are mapped to individual Source Classification Codes (SCCs; Table ~~S10~~S11). Categorically, VCPy reports higher emissions for Personal Care Products (150%) and Paints & Coatings (~~34~~25%) ~~feature the largest increases~~, whereas Pesticides (-54%) and Printing Inks (-13%) feature ~~the largest~~ emission decreases. The VCPy inventory also includes marginal increases in Cleaning Products and Adhesives & Sealants emissions, while also quantifying solvent-borne emissions in Oil & Gas operations (included as "Other" in Fig. S5).

3.4 Evaluation of Inventory Using Emission Ratios

Predicted per-capita VCP emissions in Los Angeles County are 8.~~42~~ 21 kg person^{-1} year^{-1} and consist of 250+ organic compounds. Observed emission ratios were available for 30 species (Table S7), including some of the most abundantly emitted (e.g. ethanol, acetone, isopropyl alcohol, toluene). In fact, of the 30 available emission ratios, 24 were for compounds that contributed more than 0.1% to total VCP emissions (Fig. 6), providing the opportunity to evaluate important markers. For most compounds, the VCPy estimate was well within a factor of 2 when compared to observations. Some important markers were marginally low biased (e.g. ethanol, isopropyl alcohol), while others were marginally high biased (e.g. acetone, methyl ethyl ketone, isobutane), illustrating the difficulty in precisely speciating organic emissions and uncertainties introduced by compositing. However, when considered as a whole, the complete VCPy inventory performs remarkably well with a correlation of 0.95. In total, the observed emission ratio for all 30 compounds was 0.259 g (g CO)$^{-1}$ and the inventory estimate is 0.226 g (g CO)$^{-1}$, indicating a 13% low bias. In addition, the VCPy inventory shows a marked improvement over the 2017 NEI, which reports 3.28 kg person^{-1} year^{-1} of VCP emissions in Los Angeles County. For the 30 compounds considered here, the 2017 NEI reports 0.143 g (g CO)$^{-1}$, which is 45% lower than observations (Fig. S6). Most notably, the emissions ratio of ethanol, acetone, isopropyl alcohol, and propane, all of which are emitted by VCPs in substantial quantities, were low by a factor of 2-3.

While the residual, 13% low bias could suggest that additional organic emissions might be missing from the VCPy inventory, several other factors could explain discrepancies. First, emission ratios are equally sensitive to both organic and CO emissions. While CO appears to be represented and modelled well in current inventories (Lu et al., 2020), a marginal, systematic bias in CO can affect the results presented here. For example, if the CO inventory were systematically high bias by 10%, the bias in the VCPy inventory emission ratios would be nearly eliminated. Second, since emission ratios are not sector-specific but reflect total emissions, missing organic emissions might be from other sources. Mobile sources, especially gasoline exhaust, is rich in small ($\leq C_6$) hydrocarbons, including ethene, n-butane, n-pentane, isopentane, methylpentanes, propene, and methylhexanes (Gentner et al., 2013). Except for n-butane, none of the remaining~~se~~ compounds appreciably come from VCP sources and all are low biased in the complete inventory (Fig. S6). Finally, while the ambient NMVOC to CO concentration ratio in Los Angeles has been consistent for several decades (McDonald et al., 2013), it is possible that trends for these two pollutants could have diverged in recent years.

3.5 Effective SOA Yields, O3 MIR, and Air Pollution Potential

Nationally, the effective SOA yield of the complete sector is 5.3% by mass (Table 4) and the most abundantly emitted SOA precursors are IVOC alkanes, aromatics, volatile methyl siloxanes, and fragrances. On a sub-PUC basis, the effective yield spans more than two-orders of magnitude, with Short Use Products and Printing Inks featuring an effective yield of 0.05% and 14.8%, respectively. For O_3, the effective MIR of the complete sector is 1.6 (g O_3) g^{-1} and, when compared to SOA yields, there is considerably less sub-PUC variability. While VCPs do emit aromatics and alkenes, both of which are photochemically reactive compound classes with high ozone potential, emissions are usually dominated by oxygenated compounds and alkanes, such as acetone, isopropyl alcohol, propane, and isobutane, which are minimally reactive. In fact, of the top fifteen highest emitting VCP compounds, seven feature a MIR < 1.0 (g O_3) g^{-1}.

While a sub-PUC may be a large source of organic emissions, this does not necessarily translate to a high potential impact on $PM_{2.5}$ and ozone. This is best highlighted by Industrial and Architectural Coatings. Together, these two sub-PUCs constitute ~20% of all VCP emissions (Table 3), but only ~10% of the total SOA potential due to their low effective yields (2.9~~4~~% and ~~2.42~~1.9%, respectively). Architectural Coatings emissions feature significant quantities of Texanol™ (a highly branched oxygenate) and small glycols, such as propylene and ethylene glycol. A < 1% ~~-~~and 0% SOA yield is assigned to Texanol™ and both glycols, respectively. Though, it should be noted that this may be a lower bound as Li et al., 2018 report moderate aerosol formation from propylene glycol. Similarly, Printing Inks contribute ~8% of all VCP emissions, which is nearly 2.5x less than Daily Use Products and General Cleaners nationally (Table 3). However, Printing Ink emissions are dominated by IVOC alkanes (C12-C16 hydrocarbons, represented by n-tetradecane here) and aromatics, resulting in a high effective SOA yield (14.~~4~~8%). As a result, Printing Inks contribute significantly to the total SOA potential nationally (Fig. 7). Paints & Coatings are nonetheless the dominant contributor to SOA potential, but this is more so due to the high emissions of the component sub-PUCs rather than their modest effective SOA yields (~~2.42~~1.9 – 6.~~5~~6%). Both General Cleaners and Daily Use Products also have moderate quantities of SOA precursors and high emissions, which translates to 17.~~2~~5% and 13.~~1~~3% of the national VCP SOA potential, respectively. Since the effective MIR of each sub-PUC is not highly variable, O_3 potential is highly correlated with emissions magnitude. Overall, the three highest emitting PUC, Paints & Coatings, Cleaning Products, and Personal Care Products, are also the highest contributors to O_3 potential (Fig. 7).

These results also demonstrate how fate-and-transport assumptions can impact estimates of SOA production. For example, a prior study reported that both laundry detergent and a general-purpose spray cleaner can form appreciable quantities of SOA (Li et al., 2018). Here, the VCPy inventory reports an effective yield of 0.0% by mass of organic emitted for Detergents & Soaps and 4.7% for General Cleaners (Table 4). While the organic content of both sub-PUCs, by mass, is ≥ 18% (Table 2), Detergents & Soaps feature a dramatically smaller use timescale (Minutes vs. Days). As a result, not only is the total mass of organic emissions from Detergents & Soaps smaller than General Cleaners, but the collection of compounds that are emitted feature systematically smaller evaporation timescales. Such compounds are highly volatile (i.e. $C^* > 1 \times 10^8\ \mu g\ m^{-3}$) and not SOA precursors. In contrast, General Cleaners are assigned a longer use timescale, which provides time for lower volatility organics (i.e. IVOCs) to evaporate and subsequently contribute to the formation of SOA.

3.6 Non-Evaporative Organic Assumptions

The composition and volatility distribution of the organics assumed to be non-evaporative, which is ~60% of all organics (Fig. S8), is unidentified and assumed to be entirely non-volatile for the main analysis. However, there is evidence that a non-negligible portion of this mass may be SVOCs ($0.3\ \mu g\ m^{-3} < C^* < 300\ \mu g\ m^{-3}$), which can evaporate on atmospherically relevant timescales (Khare and Gentner, 2018). SHEDS-HT, a near-field model used to prioritize human exposure to chemicals (Isaacs et al., 2014), reports that > 15%, > 5%, and > 2% of all organics found in residential personal care product, household product, and coatings, respectively, are composed of SVOCs (Qin et al., 2020). The treatment of non-evaporative organics and their potential emission can have a substantial impact on the modulation of SOA potential from VCPs. For example, if the assumption regarding evaporation of these organics is relaxed by assuming 1% of all non-evaporative organics eventually do evaporate, sector-wide emissions would increase by 0.18 kg person^{-1} year^{-1} (i.e. < 2% of the VCP emissions). Such a scenario is possible for products featuring long use timescales (e.g. paints, pesticides), if SVOCs are considered non-evaporative, or if products featuring shorter use timescales (e.g. Daily Use Products, Cleaning Products) are not fully sequestered. Since this increase in emissions is minor (i.e. < 2%), there would be negligible impacts on the total emission magnitude and O_3. However, these compounds, by definition, feature low vapor pressures, which makes them prime SOA precursor candidates. If these compounds were permitted to form SOA with 100% efficiency, the effective yield from the complete sector would increase from 5.3% to 7.0% by mass (Fig. S8). Correspondingly, if 2% of all non-evaporative organics were assumed to evaporate with similar SOA formation assumptions, the effective yield from the complete sector would increase to 8.~~6~~7% by mass.

4 Additional Uncertainties

The current VCPy framework assumes all evaporated organics reach the ambient atmosphere, regardless of origin. However, VCP emissions occur both indoors and outdoors (Farmer et al., 2019; Nazaroff and Weschler, 2004; Singer et al., 2006a). In fact, the indoor concentration of prevalent VCP markers and secondary pollutants often exceeds outdoor concentrations (Farmer et al., 2019; Patel et al., 2020). For ambient air emissions, consideration of VCP emissions indoors is important if there is a gas-phase loss mechanism occurring at a scale that is comparable to typical indoor air exchange rates (~0.5 hr^{-1}; Murray and Burmaster, 1995). Indeed, sorption of gas-phase organics (e.g. terpenes) into typical residential furnishing and dust has been shown to occur on relevant timescales (Singer et al., 2007; Singer et al., 2004; Weschler and Nazaroff, 2008). Organics emitted indoors can also react with oxidants, leading to the formation of lower-volatility organics that can form particulates (Nazaroff and Weschler, 2004; Singer et al., 2006b). These particulates can deposit before outdoor exhaust ~~can~~ occurs due to the high surface-to-volume ratio of indoor settings (Abbatt and Wang, 2020; Farmer et al., 2019). Planned future VCPy functionality includes the incorporation of a two-box model to capture these possible termination mechanisms and distinguish between near-field and far-field exposure pathways.

In addition, the efficiency of post-use controls for several sub-PUCs can be highly uncertain and vary both in space and time. In particular, this includes Oil & Gas, which is assigned a post-use control based on average reported reinjection rates of produced water (Liden et al., 2018; Lyman et al., 2018), as well as Industrial Coatings and Printing Inks, which occur at facilities capable of add-on controls (U.S. Environmental Protection Agency, 2006a; 2006b; 2007; 2008). Here, post-use controls are not assigned for Industrial Coatings or Printing Inks. As such, emissions from these sub-PUCs could feature localized high bias, depending on regional control requirements for facilities that use associated products. Similarly, the spatial allocation of nonpoint emissions features unique difficulties. For example, even if the allocation of nonpoint emissions was precisely matched to a quantifiable proxy, variation in the emission strength of individuals within that proxy (e.g. humans or employees) is often neglected (Li et al., 2020).

5 Conclusions

VCPy is a new framework to model organic emissions from volatile chemical products throughout the United States, including spatial allocation to regional and local scales. In VCPy, product volatilization is a function of the characteristic evaporation timescale of individual components and the use timescale for product-use categories. National, per-capita organic emissions from VCPs are 9.~~7~~5 kg person^{-1} year^{-1} (6.~~5~~4 kgC person^{-1} year^{-1}) for 2016, which translates to 3.~~12~~05 Tg (2.~~10~~06 TgC) for the U.S. Paints & Coatings, Personal Care Products, and Cleaning Products contribute most to these emissions. When filtered to remove regulatory exempt organics, ~~which enables a direct comparison to the EPA's NEI,~~ total emissions from VCPs are 2.6 Tg of VOC and equal in magnitude to the sum of all mobile sources nationally, thus highlighting the growing importance of the VCP sector. Organic emissions featured substantial (~20%) contributions from IVOCs, which are likely SOA precursors. Of this 20%, ~~55~~52% are oxygenated compounds, ~~27~~30% are n-alkanes, and the rest are largely branched and cyclic alkanes. Nationally, the effective SOA yield and MIR, two metrics that facilitate an approximation of the potential air quality impacts, of VCPs is 5.3% by mass and 1.~~59~~58 (g O_3) g^{-1}, respectively. This effective SOA yield indicates VCPs are likely a significant source of SOA in urban environments (Qin et al., 2020).

Uncertainty associated with this framework was tested through Monte Carlo analysis. Notably, the dominant drivers of uncertainty were associated with estimated product usage and the composition of products, and not assumptions related to fate-and-transport. SOA formation from VCP emissions is especially sensitive to assumptions regarding evaporation of low volatility species. If 1% of all non-evaporative organics eventually do evaporate, sector-wide emissions would increase by 0.18 kg person^{-1} year^{-1} and the effective SOA yield from the complete sector could increase by > 1.5%. The 95% confidence interval for the national level emissions from the complete sector for 2016 is 2.~~68~~61 – 3.~~60~~53 Tg (1.~~81~~76 – 2.~~42~~38 TgC). This is broadly consistent with the 2017 National Emission Inventory (2.84 Tg) and half the emissions magnitude reported elsewhere (McDonald et al., 2018).

While the national level emissions from the VCPy framework and the 2017 NEI are comparable, regional and localized differences can be significant. This is most clear when evaluating the VCPy inventory to published emission ratios. For Los Angeles County, the VCPy inventory performs well (normalized mean bias of -13% with r = 0.95) and is significantly improved over the reported 2017 NEI VCP emissions. Planned future work includes adoption of variable emission settings (indoor vs. outdoor) to account for loss mechanisms indoors (e.g. gas-phase sorption to surfaces), revisited mapping of VCP emissions to common chemical mechanisms for ease of research use in the chemical transport modelling community, estimation of SOA and ozone formation from VCPs using a chemical transport model and VCPy emissions inputs, and understanding the evolution of VCP emissions over time.

Data Availability

VCPy.v1.0 ~~will~~ is~~be~~ available on data.gov ~~following publication~~ (doi: 10.23719/1520157~~doi, to be provided~~). All data presented in this manuscript can be retrieved and/or generated by downloading VCPy.v1.0. ~~Additional instructions can be found in the main directory and g~~Guidance on using VCPy.v1.0 can be requested by contacting ~~one of~~ the corresponding author~~s~~.

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Author Contributions

KMS and HOTP designed the research scope. All authors participated in data curation and/or analysis. KMS and HOTP drafted the initial manuscript and all authors contributed to subsequent drafts.

Competing Interests

The authors declare that they have no conflicts of interest.

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Table 1: Description of all PUCs and sub-PUCs currently implemented in VCPy, their estimated mass usage for 2016, and product examples of each. See Table S2 for a derivation of all product usage estimates.

Product Use Categories (PUCs)	**Sub-Product Use Categories (sub-PUCs)**	**2016 Annual Usage [kg person^{-1} year^{-1}]**	**Product Examples**
Cleaning Products	Detergents & Soaps	40.58	Soaps, Detergents, Metal Cleaners, Scouring Cleaners
	General Cleaners	28.47	Disinfectants, Air Fresheners, Glass & Bathroom Cleaners, Windshield Washer Fluid, Hand Sanitizer, Automotive & Floor Polishes, Bleaches, Surfactants
Personal Care Products	Daily Use Products	8.83	Hair Products, Perfumes, Colognes, Cleansing & Moisturizing Creams, Sunscreens, Hand & Body Lotion and Oils, Cosmetics, Deodorants
	Short Use Products	3.16	Shampoo, Conditioners, Shaving Cream, Aftershave, Mouthwashes, Toothpaste
Adhesives & Sealants	Adhesives & Sealants	15.23	Glues and Adhesives, Epoxy Adhesives, Other Adhesives, Structural and Nonstructural Caulking Compounds and Sealants
Paints & Coatings	Architectural Coatings	13.27	Exterior/Interior Flat/Gloss Paints, Primers, Sealers, Lacquers
	Aerosol Coatings	0.39	Paint Concentrates Produced for Aerosol Containers
	Allied Paint Products	1.26	Thinners, Strippers, Cleaners, Paint/Varnish Removers
	Industrial Coatings	7.42	Automotive, Appliance, Furniture, Paper, Electrical Insulating, Marine, Maintenance, and Traffic Marking Finishes and Paints
Printing Inks	Printing Inks	3.20	Letterpress, Lithographic, Gravure, Flexographic, Nonimpact/Digital Inks
Pesticides & FIFRA Products	FIFRA Pesticides	1.46	Lawn and Garden Pesticides and Chemicals, Household and Institutional Pesticides and Chemicals
	Agricultural Pesticides	10.32	Agricultural and Commercial Pesticides & Other Organic Chemicals
Dry Cleaning	Dry Cleaning	0.03	Dry Cleaning Fluids
Oil & Gas	Oil & Gas	1.32	Cleaners, Deicers
Misc. Products	Misc. Products	0.18	Pens, Markers, Arts and Crafts, Dyes
Fuels & Lighter	Fuels & Lighter	2.80	Lighter Fluid, Fire Starter, Other Fuels

Table 2: 1st-Order product composition profiles and evaporative organics proportion for all sub-PUCs.

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	Water	Inorganic	Non-Evaporative Organics[a]	Evaporative Organics[a]
Cleaning Products	Detergents & Soaps[b]	67.8%	13.9%	15.4%	2.9%
	General Cleaners[b]	73.3%	8.6%	11.1%	6.9%
Personal Care Products	Daily Use Products[b]	48.8%	10.7%	16.9%	23.7%
	Short Use Products[b]	72.2%	5.8%	17.7%	4.3%
Adhesives & Sealants	Adhesives & Sealants[b]	12.8%	53.2%	29.0%	5.0%
Paints & Coatings	Architectural Coatings[c]	~~41.9~~45.5%	~~51.1~~49.6%	0.0%	~~6.8~~5.0%
	Aerosol Coatings[d]	12.7%	12.7%	0.0%	74.7%
	Allied Paint Products[b]	5.1%	3.5%	0.6%	90.8%
	Industrial Coatings[e]	15.0%	70.0%	0.0%	14.0%
Printing Inks	Printing Inks[f]	8.0%	67.0%	0.0%	25.0%
Pesticides & FIFRA Products	FIFRA Pesticides[b]	74.8%	4.9%	15.1%	5.1%
	Agricultural Pesticides[b]	74.8%	4.9%	15.1%	5.1%
Dry Cleaning	Dry Cleaning[g]	0.0%	0.0%	0.0%	100%
Oil & Gas	Oil & Gas[g]	0.0%	0.0%	0.0%	100%
Misc. Products	Misc. Products[b]	27.1%	14.6%	48.8%	9.5%
Fuels & Lighter	Fuels & Lighter[b]	0.0%	92.9%	0.0%	7.1%

[a]: "Non-Evaporative Organics" and "Evaporative Organics" sum to total product organics. "Evaporative Organics" represent the potentially evaporative organic fraction of the total product and excludes assumed "non-evaporative" (i.e. assumed non-volatile) organics, which are not included in the California Air Resource Board's organic profiles.

[b]: Source: California Air Resources Board 2015 Consumer and Commercial Products Survey Data (CARB, 2019).

[c]: Source: California Air Resources Board 2005 Architectural Coatings Survey (CARB, 2007). VOC + Exempts is used for both organic and evaporative organics. Non-evaporative organic proportions not provided. Sales proportions of water vs. solvent-based architectural coatings based on California Air Resource Board 2014 Architectural Coatings Survey (CARB 2014).

[d]: Source: California Air Resources Board 2010 Aerosol Coatings Survey (CARB, 2012). Only evaporative organics is provided. Remainder (~25%) is split evenly between water and inorganics.

[e]: Source: Industrial Maintenance composition data from California Air Resources Board 2005 Architectural Coatings Survey (CARB, 2007).

[f]: Source: Graphic Arts composition data from California Air Resources Board 2005 Architectural Coatings Survey (CARB, 2007).

[g]: All product usage is composed of organic functional solvents (The Freedonia Group, 2016). Therefore, all mass is assumed to be potentially evaporative.

Table 3: National-level emissions, volatilization fraction, and proportion of all usage that is emitted for all sub-PUCs.

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	ROC Emissions		Organic Volatilization Fraction [%][a]	Total Product Emitted [%]
		[kg person^{-1} year^{-1}]	[kgC person^{-1} year^{-1}]		
Cleaning Products	Detergents & Soaps	0.12	0.06	1.6%	0.3%
	General Cleaners	1.85	1.25	36.0%	6.5%
Personal Care Products	Daily Use Products	2.~~03~~04	1.12	56.~~7~~9%	23.~~0~~1%
	Short Use Products	0.02	0.01	3.3%	0.7%
Adhesives & Sealants	Adhesives & Sealants	0.76	0.56	14.7%	5.0%
Paints & Coatings	Architectural Coatings	0.~~89~~67	0.~~51~~37	100%[b]	~~6.7~~5.0%
	Aerosol Coatings	0.29	0.22	100%[b]	74.7%
	Allied Paint Products	1.14	0.80	99.2%	90.6%
	Industrial Coatings	1.04	0.79	100%[b]	14.0%
Printing Inks	Printing Inks	0.80	0.65	100%[b]	25.0%
Pesticides & FIFRA Products	FIFRA Pesticides	0.07	0.06	25.2%	5.1%
	Agricultural Pesticides	0.53	0.41	25.2%	5.1%
Dry Cleaning	Dry Cleaning	0.01	0.01	34.5%	34.5%
Oil & Gas	Oil & Gas	0.08	0.04	6.0%	6.0%
Misc. Products	Misc. Products	0.02	0.01	16.3%	9.5%
Fuels & Lighter	Fuels & Lighter	0.02	0.02	10.0%	0.7%
Total		9.~~67~~45	6.~~52~~38	~~32.0~~31.5%	~~7.0~~6.9%

[a]: Volatilization fraction represents the fraction of the total organic content of products that volatilize/emit to ambient air.

[b]: The "Organic" portion of these sub-PUCs is entirely composed of "Evaporative Organics" (see Table 2). Only data from the California Air Resources Board's 2015 Consumer and Commercial Products Survey featured the disaggregation of evaporative and non-evaporative organics. Prior surveys typically combined the non-evaporative organic portion of each profile with solids/inorganics.

Table 4: The national effective SOA yield and MIR for all sub-PUCs. These results are plotted in Fig. S7.

Product Use Categories (PUCs)	**Sub-Product Use Categories (sub-PUCs)**	**Effective SOA Yield [%]**	**Effective MIR [(g O_3) g^{-1}]**
Cleaning Products	Detergents & Soaps	0.00	1.48
	General Cleaners	4.74	1.88
Personal Care Products	Daily Use Products	3.~~27~~26	1.38
	Short Use Products	0.05	1.27
Adhesives & Sealants	Adhesives & Sealants	6.19	1.51
Paints & Coatings	Architectural Coatings	~~2.42~~1.92	1.~~89~~92
	Aerosol Coatings	3.26	1.66
	Allied Paint Products	6.56	1.27
	Industrial Coatings	2.94	1.71
Printing Inks	Printing Inks	14.81	1.93
Pesticides & FIFRA Products	FIFRA Pesticides	8.10	1.01
	Agricultural Pesticides	8.10	1.01
Dry Cleaning	Dry Cleaning	3.47	1.13
Oil & Gas	Oil & Gas	2.21	1.03
Misc. Products	Misc. Products	1.94	2.26
Fuels & Lighter	Fuels & Lighter	5.35	1.15
Total		5.~~26~~29	1.~~59~~58

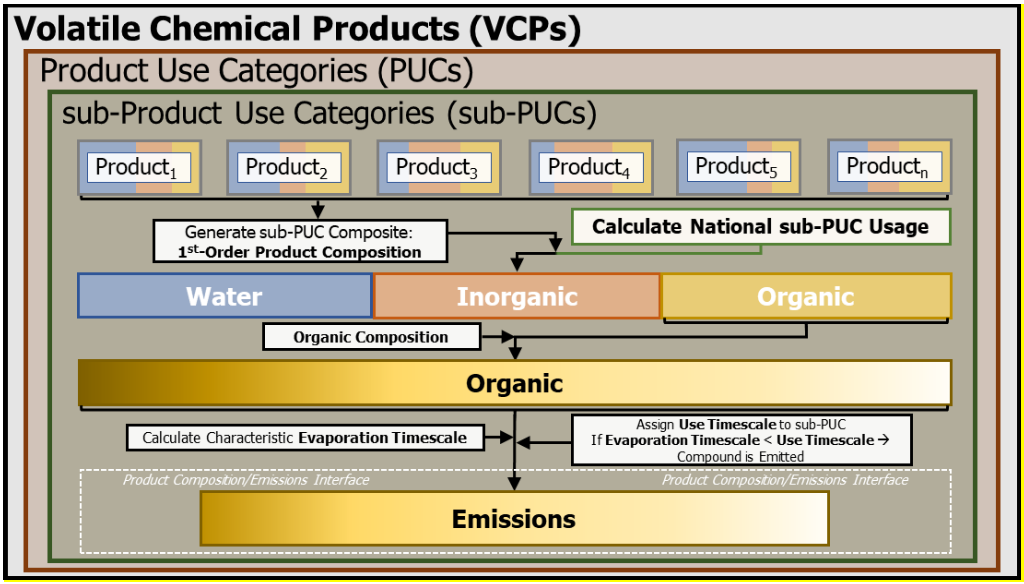


Figure 1: Conceptual overview of the VCPy framework. Note: PUC = Product Use Category.

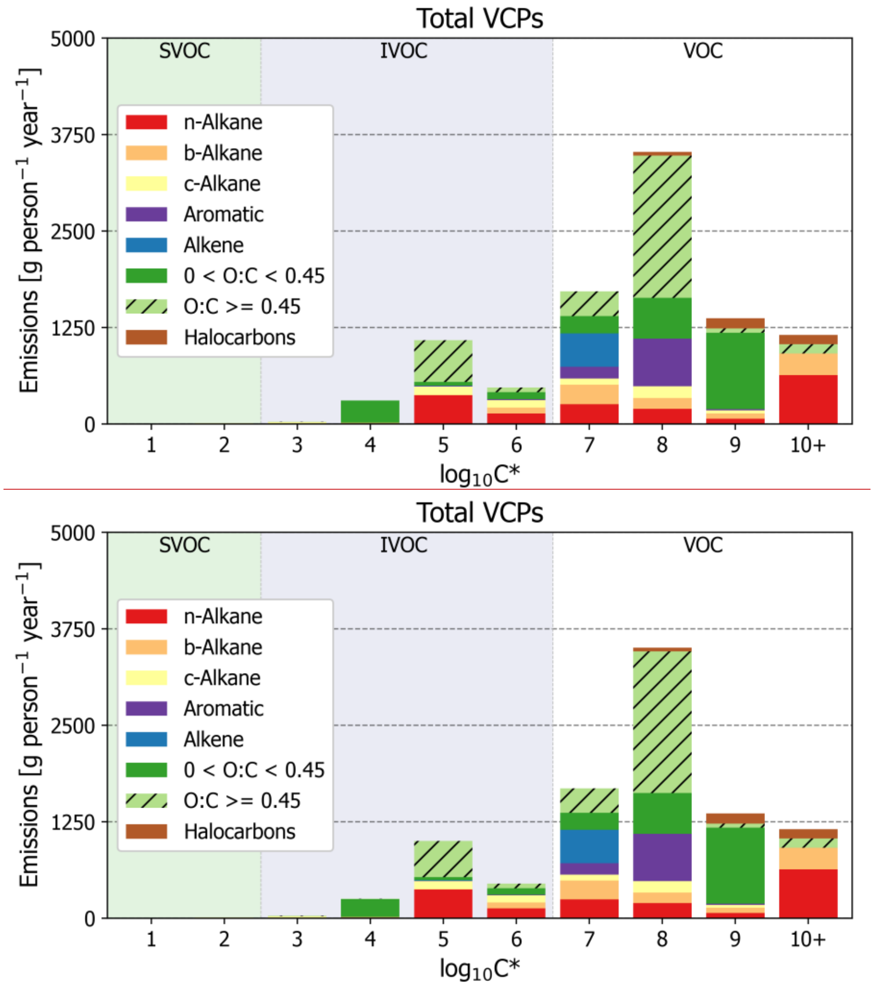
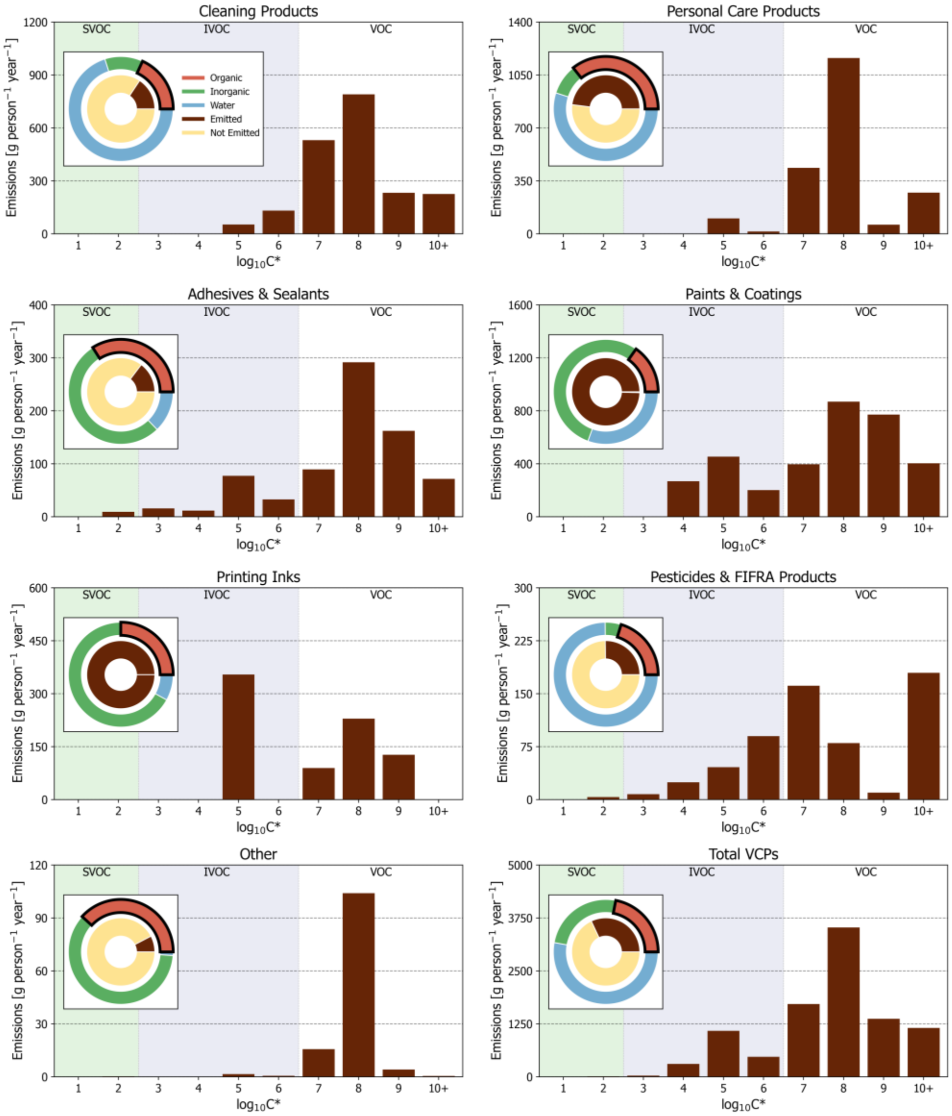


Figure 2: Sector-wide volatility distribution of emissions by compound class.


Cleaning Products
Personal Care Products
Adhesives & Sealants
Paints & Coatings
Printing Inks
Pesticides & FIFRA Products
Other
Total VCPs
SVOC
IVOC
VOC
Organic
Inorganic
Water
Emitted
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Emissions [g person−1 year−1]
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1 2 3 4 5 6 7 8 9 10+

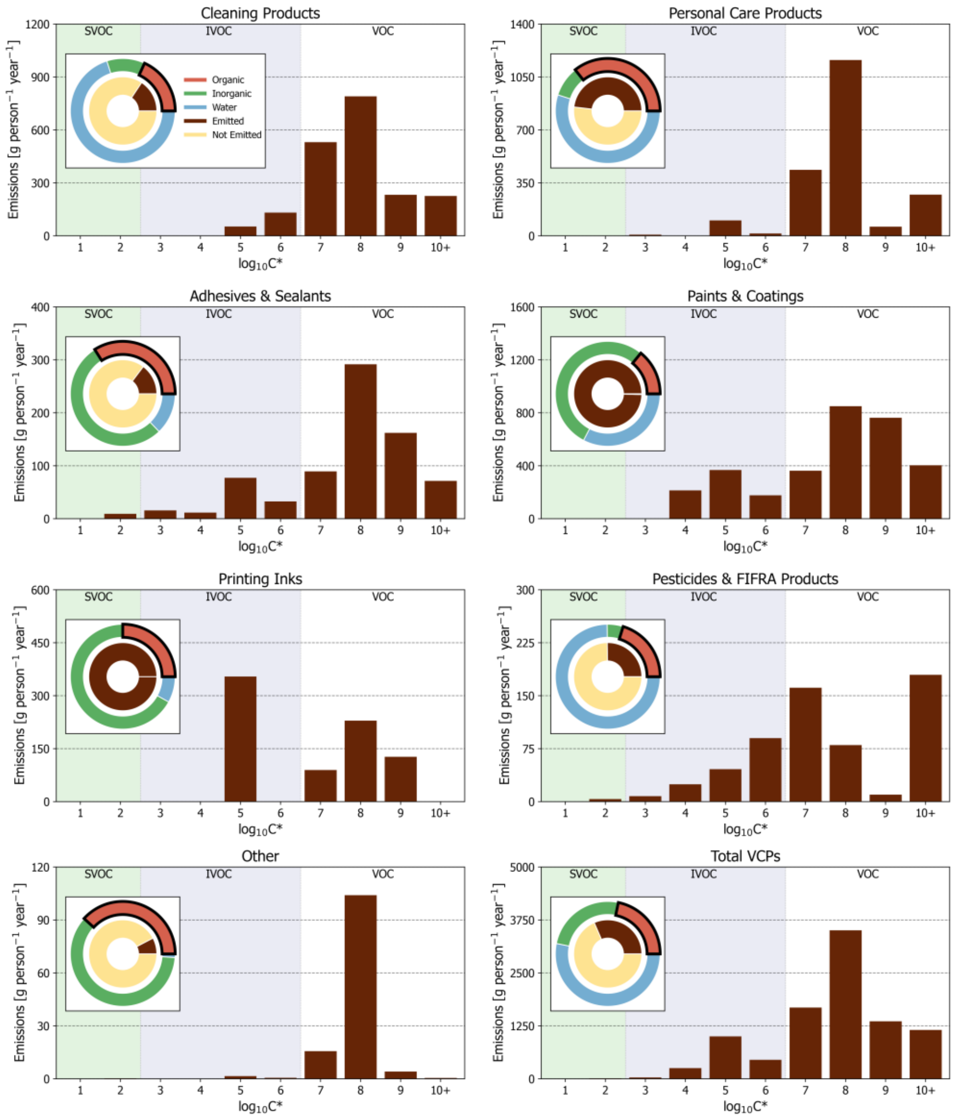


Figure 3: PUC and sector-wide volatility distribution of organic emissions. Other is summation of Dry Cleaning, Oil & Gas, Misc. Products, and Fuels & Lighter. Pie charts are 1st-order product composition and organic emission proportions for PUCs and the complete sector. Note: The "Organic" portion of all Paints & Coatings and Printing Inks pie charts is entirely composed of "Evaporative Organics" (see Table 2).

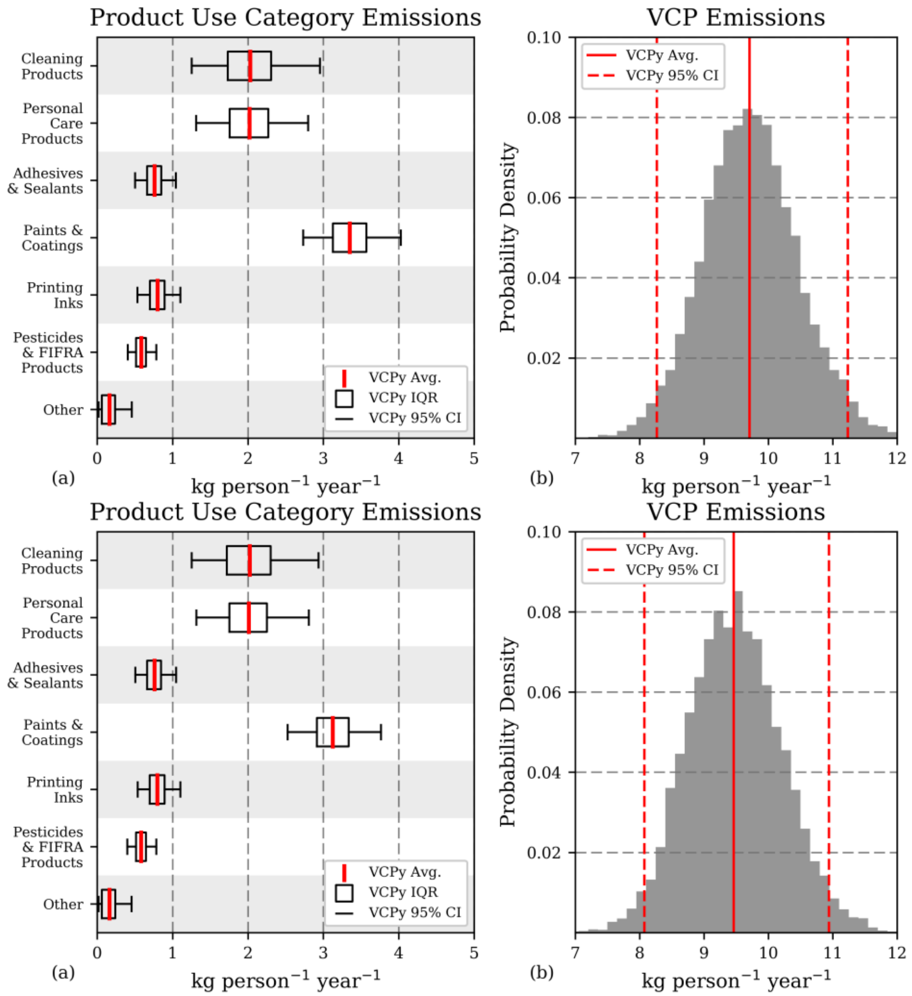


Figure 4: Monte Carlo sensitivity results for organic emissions. (a) Mean, interquartile range, and 95% confidence intervals for six PUCs and a combination of the remaining four (Dry Cleaning, Oil & Gas, Misc. Products, and Fuels & Lighter). (b) Probability distribution of sector-wide emission estimates. See Table ~~S9~~ S10 for a tabulation of this figure.

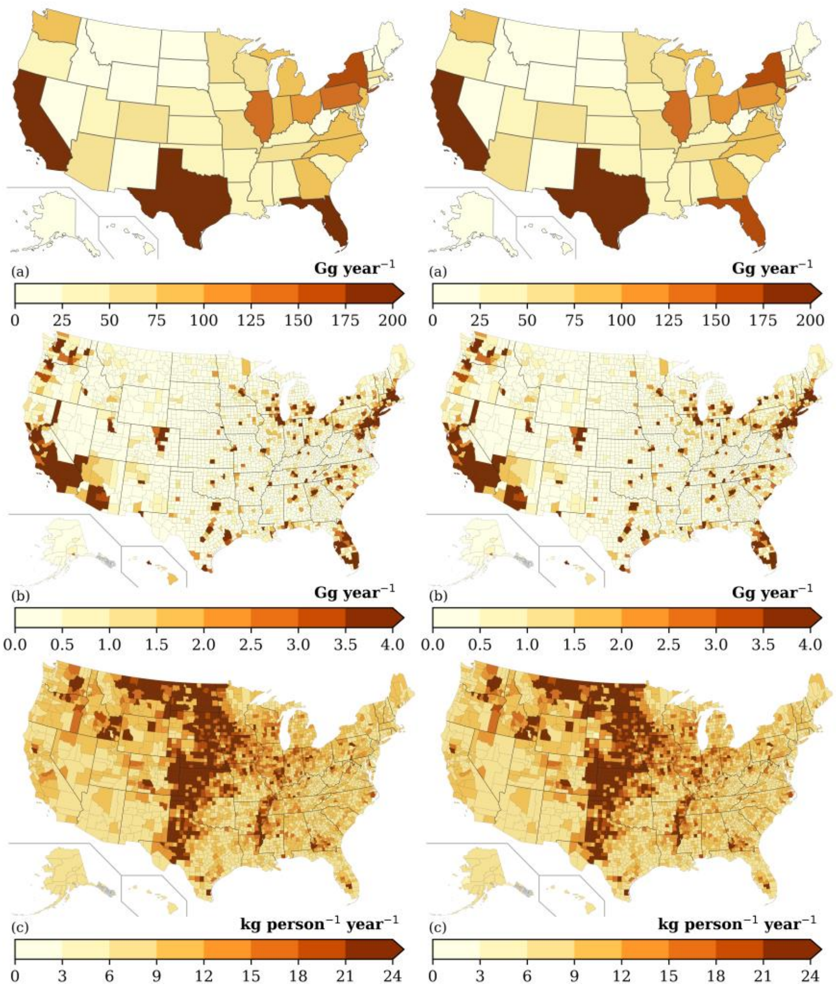
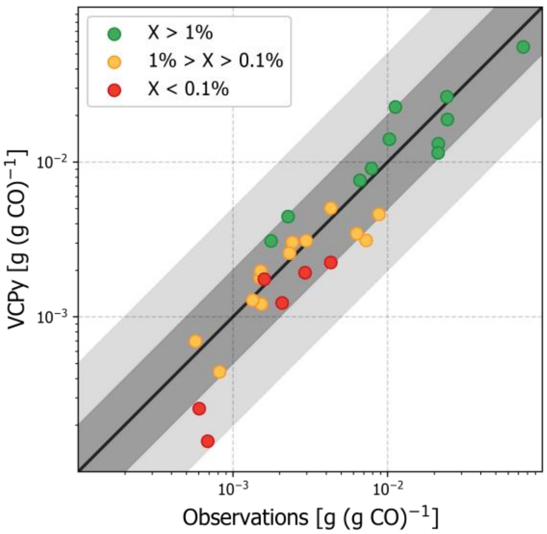


Figure 5: (a) State-level, (b) County-level, and (c) County-level per-capita VCP emissions.



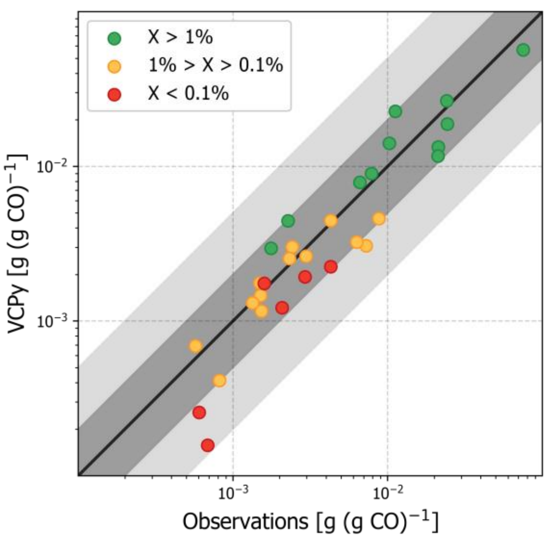


Figure 6: Evaluation of organic emission ratios in Los Angeles County using observed emission ratios from summer 2010. VCPy inventory ratios utilize VCPy predicted emissions for VCPs and the 2017 NEI for all other sources. The scatter point colors represent the relative abundance of each compound (represented as "X" in the figure legend) in the complete VCP sector. For example, all green points represent compounds that are > 1% of the total VCP emissions in Los Angeles County. Black line – 1:1; Dark grey shading – 2:1; Light grey shading – 5:1. Values available in Table S7.

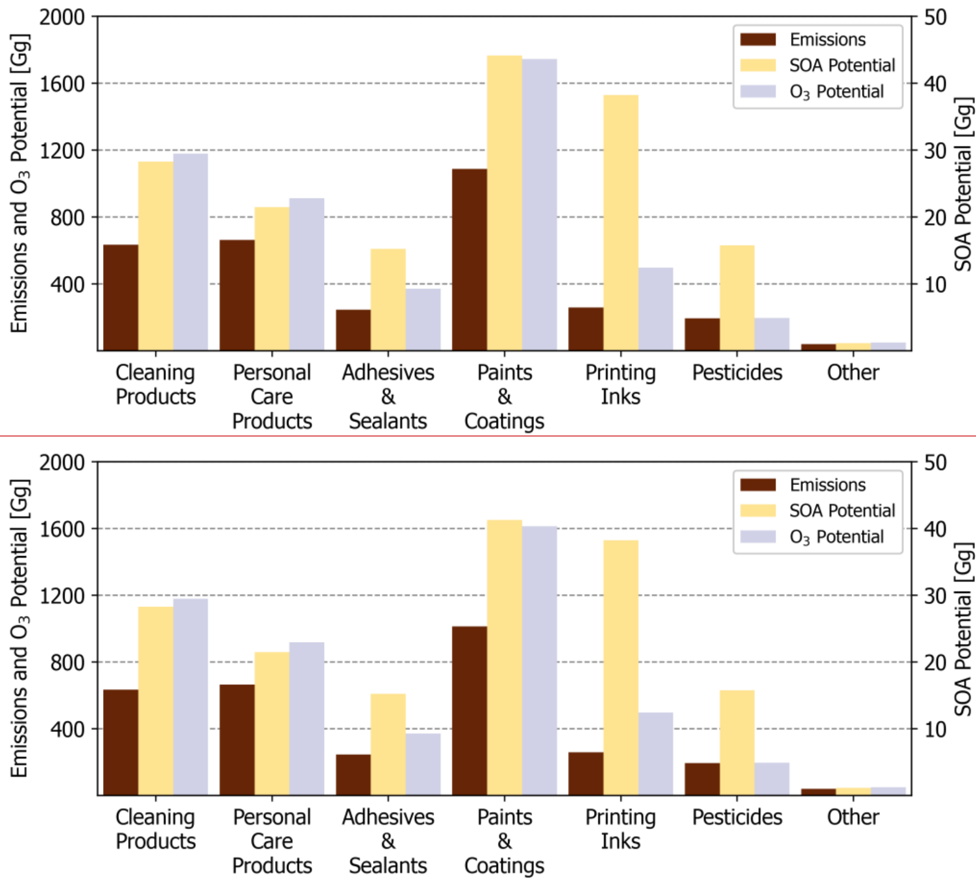


Figure 7: National-level emissions, SOA potential, and O_3 potential by PUC. Other is summation of Dry Cleaning, Oil & Gas, Misc. Products, and Fuels & Lighter.

Supporting Information:
Reactive Organic Carbon Emissions from Volatile Chemical Products

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Table S1: PUCs, sub-PUCs, NAICS codes, and SCTG codes for all sub-PUCs.

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	NAICS Codes[a]	SCTG Code[b]	Producer Price Index Category[c]
Cleaning Products	Detergents & Soaps	3256111, 3256114, 3256117, 325611W	233	Soap and Other Detergent Manufacturing
	General Cleaners	3256125, 2356127, 2356121, 235611A, 2356130, 325612W	233	Polish and Other Sanitation Good Manufacturing; Soap and Other Detergent Manufacturing; Surface Active Agent Manufacturing;
Personal Care Products	Daily Use Products	3256204, 325620D, 325620G, 325620W, 3256207 (25%)[d]	232	Toilet Preparation Manufacturing
	Short Use Products	3256201, 325620A, 325611D, 3256207 (75%)[d]	232	Toilet Preparation Manufacturing; Soap and Other Detergent Manufacturing
Adhesives & Sealants	Adhesives & Sealants	3255201, 3255204, 3255207, 305520A, 325520W	239	Adhesive Manufacturing
Paints & Coatings	Architectural Coatings	3255101, 325510W	[f]	Paint and Coating Manufacturing
	Aerosol Coatings	3255107 (10%)[e]	[f]	Paint and Coating Manufacturing
	Allied Paint Products	325510B	[f]	Paint and Coating Manufacturing
	Industrial Coatings	3255104, 3255107 (90%)[e]	[f]	Paint and Coating Manufacturing
Printing Inks	Printing Inks	3259101, 3259104, 3259107, 325910A, 325910E, 325910H, 325910W	231	Printing Ink Manufacturing
Pesticides & FIFRA Products	FIFRA Pesticides	3253204, 3253207	235	Pesticide and Other Agricultural Chemical Manufacturing
	Agricultural Pesticides	3251994, 3253201, 325320W	235	All Other Basic Organic Chemical Manufacturing; Pesticide and Other Agricultural Chemical Manufacturing
Dry Cleaning	Dry Cleaning	[g]		
Oil & Gas	Oil & Gas			
Misc. Products	Misc. Products	[h]		
Fuels & Lighter	Fuels & Lighter			

[a]: NAICS Codes used for mapping U.S. Census Bureau (2016) ASM statistics to individual sub-PUCs.
[b]: SCTG Codes used for mapping U.S. Department of Transportation (2015) calculated commodity values to individual sub-PUCs. All values ($) and mass (tons) quantities retrieved from Table 6 of that report.
[c]: Category used when retrieving Produce Price Indices from the U.S. Bureau of Labor Statistics, FRED, Federal Reserve Bank of St. Louis.
[d]: NAICS code 3256207 includes all hair preparation products. Based on sales data by California Air Resources Board 2015 Consumer and Commercial Products Survey Data, we estimate ~75% of all hair preparation products are short-use (e.g. shampoos, conditioners) and ~25% are daily-use (e.g. hair spray, other leave-in products).
[e]: NAICS code 3255107 includes all special-purpose coating materials (e.g. automotive finishing, traffic markings, aerosolized painting products). Based on shipment data from U.S. Census Bureau, Paint and Allied Products - 2010, MA325F(10), Issued July 2011, we estimate ~10% of all special-purpose coating materials in this NAICS are aerosolized painting products, with the residual consisting of special-purpose coating materials used exclusively in industrial settings.
[f]: All Paints & Coatings commodity values retrieved from: U.S. Census Bureau, Paint and Allied Products - 2010, MA325F(10), Issued July 2011.
[g]: sub-PUC usage estimated from solvent usage as reported by The Freedonia Group; Industry Study #3429; Solvents; July 2016.
[h]: sub-PUC usage estimated from reported sales data by California Air Resources Board 2015 Consumer and Commercial Products Survey Data: https://ww3.arb.ca.gov/consprod/survey/2015_cp_survey_summary_data_2019-12-09.xlsx; last access: August 28, 2020

Table S2: PUCs, sub-PUCs, NAICS codes, shipment values, commodity price, producer price index, and product usage for all sub-PUCs (2016). Per-capita product usage calculated using the U.S. Census Bureau's 2016 estimate of the U.S. population (~323,000,000).

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	NAICS Codes	ASM Shipment Values [$1000][a]	Commodity Price [$/kg][b]	2016 Producer Price Index[c]	2012 Producer Price Index[c]	Annual Usage [kg/person/year][d]	
Cleaning Products	Detergents & Soaps	3256111	7849011	1.615	1.08	1.07	14.92	40.58
		3256114	10137793		1.08	1.07	19.27	
		3256117	2740950		1.08	1.07	5.21	
		325611W	625339		1.08	1.07	1.19	
	General Cleaners	3256125	5391225	1.615	1.11	1.05	9.85	28.47
		2356127	1024203		1.11	1.05	1.87	
		2356121	1067746		1.11	1.05	1.95	
		235611A	290791		1.08	1.07	0.55	
		2356130	7173431		1.18	1.16	13.48	
		325612W	417549		1.11	1.05	0.76	
Personal Care Products	Daily Use Products	3256204	3140488	9.290	1.07	1.03	1.01	8.83
		325620D	7912985		1.07	1.03	2.54	
		325620G	13192677		1.07	1.03	4.23	
		325620W	1562269		1.07	1.03	0.50	
		3256207 (25%)[e]	1734011		1.07	1.03	0.56	
	Short Use Products	3256201	281282	9.290	1.07	1.03	0.09	3.16
		325620A	3574170		1.07	1.03	1.15	
		325611D	789937		1.08	1.07	0.26	
		3256207 (75%)[e]	5202032		1.07	1.03	1.67	
Adhesives & Sealants	Adhesives & Sealants	3255201	1405299	2.602	1.11	1.10	1.65	15.23
		3255204	7967659		1.11	1.10	9.34	
		3255207	694706		1.11	1.10	0.81	
		305520A	2405554		1.11	1.10	2.82	
		325520W	513470		1.11	1.10	0.60	
Paints & Coatings	Architectural Coatings	3255101,	11253306	2.694[g]	1.14	1.00[h]	11.32	13.27
		325510W	1933890		1.14	1.00[h]	1.95	
	Aerosol Coatings	3255107 (10%)[f]	671641	4.616[g]	1.14	1.00[h]	0.39	0.39
	Allied Paint Products	325510B	1383897	2.987[g]	1.14	1.00[h]	1.26	1.26
	Industrial Coatings	3255104	6157635	4.310[g]	1.14	1.00[h]	3.87	7.42
		3255107 (90%)[f]	6044765	4.616[g]	1.14	1.00[h]	3.55	
Printing Inks	Printing Inks	3259101	265917	4.217	1.17	1.07	0.18	3.20
		3259104	1050099		1.17	1.07	0.71	
		3259107	271753		1.17	1.07	0.18	
		325910A	837641		1.17	1.07	0.56	
		325910E	1233022		1.17	1.07	0.83	
		325910H	608350		1.17	1.07	0.41	
		325910W	498817		1.17	1.07	0.34	
Pesticides & FIFRA Products	FIFRA Pesticides	3253204	905062	4.552	1.05	1.04	0.61	1.46
		3253207	1273582		1.05	1.04	0.86	
	Agricultural Pesticides	3251994	1475958	4.552	1.04	1.12	1.08	10.32
		3253201	12842492		1.05	1.04	8.63	
		325320W	911325		1.05	1.04	0.61	
Dry Cleaning	Dry Cleaning	i						0.03
Oil & Gas	Oil & Gas							1.32
Misc. Products	Misc. Products	j						0.18
Fuels & Lighter	Fuels & Lighter							2.80

[a]: All values ($) retrieved via the U.S. Census Bureau ASM's API tool.
[b]: All quantities retrieved from Table 6 of U.S. Department of Transportation (2015) and representative of 2012 values.
[c]: U.S. Bureau of Labor Statistics, Producer Price Index by Industry, retrieved from FRED, Federal Reserve Bank of St. Louis. All re-indexed to 2010.
[d]: Annual usage $[\mathbf{kg/person/year}] = (\boldsymbol{ASM\ Shipment\ Values}) \div \left(\boldsymbol{Commodity\ Price} \times \left(\boldsymbol{2016\ Index}/\boldsymbol{2012\ Index}\right)\right) \div (\boldsymbol{Population})$

[e]: NAICS code 3256207 includes all hair preparation products. Based on sales data by California Air Resources Board 2015 Consumer and Commercial Products Survey Data, we estimate ~75% of all hair preparation products are short-use (e.g. shampoos, conditioners) and ~25% are daily-use (e.g. hair spray, other leave-in products).
[f]: NAICS code 3255107 includes all special-purpose coating materials (e.g. automotive finishing, traffic markings, aerosolized painting products). Based on shipment data from U.S. Census Bureau, Paint and Allied Products - 2010, MA325F(10), Issued July 2011, we estimate ~10% of all special-purpose coating materials in this NAICS are aerosolized painting products, with the residual consisting of special-purpose coating materials used exclusively in industrial settings.
[g]: Commodity values retrieved from: U.S. Census Bureau, Paint and Allied Products - 2010, MA325F(10), Issued July 2011.
[h]: To be consistent with U.S. Census Bureau, Paint and Allied Products – 2010, producer price indices from 2010 used here.
[i]: sub-PUC usage estimated from solvent usage as reported by The Freedonia Group; Industry Study #3429; Solvents; July 2016.
[j]: sub-PUC usage estimated from reported sales data by California Air Resources Board 2015 Consumer and Commercial Products Survey Data (CARB, 2019).

Table S3: Derivation of complete (1st-order and organic) product composition profiles for Adhesives & Sealants. A similar composite tables was generated for all sub-PUCs.

Product Type	CARB Profile[a]	Sales [tpd]	Sales [%]	Water [%]	Inorganic [%]	Organic [%]	Evaporative Organics [%][b]	Evaporative Organics Profile Composite [%][c]
Other adhesives	3096	58.95	38.6%	3.97%	61.47%	34.56%	1.09%	16.81%
Sealant or Caulking Compound -- Nonchemically Curing	3005	30.34	19.9%	18.44%	51.43%	30.14%	2.14%	16.99%
Spackling Compound	3005	12.43	8.1%	19.58%	51.06%	29.36%	1.56%	5.07%
Carpet and Tile Adhesive	3003	11.69	7.7%	17.45%	52.61%	29.93%	1.29%	3.95%
Construction, Panel, or Floor Covering Adhesive	3001	8.81	5.8%	26.41%	44.08%	29.51%	5.52%	12.73%
Sealant or Caulking Compound -- Chemically Curing	3005	7.31	4.8%	1.05%	60.67%	38.28%	5.25%	10.04%
Other sealants and caulks	3005	6.85	4.5%	8.57%	55.92%	35.51%	5.07%	9.10%
Woodworking Glue	1510	6.00	3.9%	47.26%	33.39%	19.35%	1.17%	1.84%
General Purpose Adhesive	3002	4.40	2.9%	28.14%	41.66%	30.20%	7.52%	8.67%
Insulating and Sealing Spray Foam	3084	2.48	1.6%	0.10%	55.03%	44.87%	14.91%	9.69%
Wood Filler	1521	1.27	0.8%	12.63%	54.66%	32.70%	2.95%	0.98%
Arts and Crafts Adhesive	3105	0.66	0.4%	68.53%	18.91%	12.55%	2.26%	0.39%
Contact Adhesive - General Purpose	3002	0.50	0.3%	42.69%	34.97%	22.34%	3.30%	0.43%
Floor Seam Sealer	3001	0.24	0.2%	47.11%	30.29%	22.60%	6.11%	0.39%
Specialty Automotive Adhesive	1503	0.24	0.2%	0.13%	54.13%	45.75%	16.28%	1.02%
Pipe Thread Sealant/Pipe Joint Compound	3004	0.14	0.1%	0.00%	61.25%	38.74%	5.40%	0.20%
Contact Adhesive - Special Purpose	2513	0.08	0.1%	1.04%	22.16%	76.80%	64.73%	1.37%
Thread Locking Compound*	3002	0.06	0.0%	0.18%	58.07%	41.75%	10.14%	0.16%
Tile and Grout Sealer	3097	0.06	0.0%	70.33%	18.07%	11.61%	1.77%	0.03%
Household Glues and Paste	3002	0.05	0.0%	53.23%	24.11%	22.66%	9.54%	0.14%
Adhesives & Sealants[d]	**--**	**152.6**	**--**	**12.80%**	**53.22%**	**33.99%**	**5.02%**	**--**

[a]: Assigned organic profile for each product type. Retrieved from California Air Resources Board and available: https://ww2.arb.ca.gov/speciation-profiles-used-carb-modeling; last access: August 28, 2020

[b]: "Evaporative Organics" is a component of "Organic." This represents the potentially evaporative organic fraction and excludes "non-evaporative" (i.e. non-volatile) organics, which are not included in the California Air Resource Board's organic profiles.

[c]: Percent of "Evaporative Organic," weighted by sales abundance:

$$\boldsymbol{Evaporative\ Organic\ Profile\ Composite}\ [\%] = (\boldsymbol{Sales})_i \times (\boldsymbol{Evaporative\ Organic})_i \div \left(\sum_{i=1}^{n} (\boldsymbol{Sales})_i \times (\boldsymbol{Evaporative\ Organics})_i\right)$$

Where i is the Product Type index and n = 20 (i.e. all Product Types).

[d]: All water, inorganic, organic, and evaporative organics percentages for the complete sub-PUC are derived on a weighted basis from the reported sales abundance.

Table S4: Organic composition profile source/method summary for all sub-PUCs.

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	Organic Composition Source/Methods
Cleaning Products	Detergents & Soaps	Product type ~~C~~composite derived from CARB's 2015 Consumer and Commercial Products Survey[b] and speciated using CARB organic profiles[c]
	General Cleaners	
Personal Care Products	Daily Use Products	
	Short Use Products	
Adhesives & Sealants	Adhesives & Sealants	
Paints & Coatings	Architectural Coatings	Product type ~~Composite~~ composite derived from CARB's 2005 Architectural Coatings ~~Survey[c]~~Survey[d]~~;~~ (Assumes ~~88~~94% is water-based, ~~12~~6% is solvent-based[e]) and speciated using CARB organic profiles[c].
	Aerosol Coatings	Product type composite ~~Composite~~ derived from CARB's 2010 Aerosol Coatings ~~Survey[d]~~Survey[f] and speciated using CARB organic profiles[c]
	Allied Paint Products	Product type composite derived from CARB's 2015 Consumer and Commercial Products Survey[b] and speciated using CARB organic profiles[c]~~Composite derived from CARB's 2015 Consumer and Commercial Products Survey~~
	Industrial Coatings	SPECIATEv5.~~0[e]~~ 0[g] Profile: 3149
Printing Inks	Printing Inks	SPECIATEv5.0[g] Profile: 2570
Pesticides & FIFRA Products	FIFRA Pesticides	Product type composite derived from CARB's 2015 Consumer and Commercial Products Survey[b] and speciated using CARB organic profiles[c]~~Composite derived from CARB's 2015 Consumer and Commercial Products Survey~~
	Agricultural Pesticides	
Dry Cleaning	Dry Cleaning	SPECIATEv5.0[g] Profile: 2422
Oil & Gas	Oil & Gas	[a]
Misc. Products	Misc. Products	Product type composite derived from CARB's 2015 Consumer and Commercial Products Survey[b] and speciated using CARB organic profiles[c]~~Composite derived from CARB's 2015 Consumer and Commercial Products Survey~~
Fuels & Lighter	Fuels & Lighter	

[a]: According to The Freedonia Group; Industry Study #3429; Solvents; July 2016: ~65% of all solvents used in O&G operations are alcohols, with the residual largely consisting of "hydrocarbons." We allocate all alcohols to methanol, as it is widely used in and emitted from O&G operations (Stringfellow, et al., 2017; Lyman et al., 2018; Mansfield et al., 2018). We treat the remaining 35% as naphtha, a blend of hydrocarbon solvents.

[b]: Ref: CARB, 2019

[c]: Ref: CARB, ~~2007~~2018

[d]: Ref: CARB, 2007

[e]: Ref: CARB, 2014

[~~d~~f]: Ref: CARB, 2012

[~~e~~g]: Ref: EPA, 2019b

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Table S5: Assigned use timescales for all sub-PUCs.

Product Use Categories (PUCs)	Sub-Product Use Categories (sub-PUCs)	Use Timescale
Cleaning Products	Detergents & Soaps	Minutes
	General Cleaners	Days
Personal Care Products	Daily Use Products	Days
	Short Use Products	Minutes
Adhesives & Sealants	Adhesives & Sealants	Years
Paints & Coatings	Architectural Coatings	Years
	Aerosol Coatings	Years
	Allied Paint Products	Years
	Industrial Coatings	Years
Printing Inks	Printing Inks	Years
Pesticides & FIFRA Products	FIFRA Pesticides	Weeks
	Agricultural Pesticides	Weeks
Dry Cleaning	Dry Cleaning	Minutes
Oil & Gas	Oil & Gas	Years
Misc. Products	Misc. Products	Years
Fuels & Lighter	Fuels & Lighter	Years

Table S6: Methods and Data Sources for Allocating National Emissions to County-level.

Sub-Product Use Categories (sub-PUCs)	Allocation Proxy	Employment NAICS[a]	NAICS Description
Detergents & Soaps	Population	--	--
General Cleaners	Population	--	--
Daily Use Products	Population	--	--
Short Use Products	Population	--	--
Adhesives & Sealants	Population	--	--
Architectural Coatings	Population	--	--
Aerosol Coatings	Population	--	--
Allied Paint Products	Employment	236///	Construction of Buildings
Industrial Coatings	Employment	811121	Automotive Body, Paint, and Interior Repair and Maintenance
		4411//	Automobile Dealers
		4412//	Other Motor Vehicle Dealers
		336411	Aircraft Manufacturing
		3365//	Railroad Rolling Stock Manufacturing
		3366//	Ship and Boat Building
		488390	Other Support Activities for Water Transportation
		339///	Miscellaneous Manufacturing
		3369//	Other Transportation Equipment Manufacturing
		811///	Repair and Maintenance
		3133//	Textile and Fabric Finishing and Fabric Coating Mills
		332812	Metal Coating, Engraving
		2373//	Highway, Street, and Bridge Construction
		321///	Wood Product Manufacturing
		337110	Wood Kitchen Cabinet and Countertop Manufacturing
		337121	Upholstered Household Furniture Manufacturing
		337122	Non-upholstered Wood Household Furniture Manufacturing
		337211	Wood Office Furniture Manufacturing
		337212	Custom Architectural Woodwork and Millwork Manufacturing
		337124	Metal Household Furniture Manufacturing
		337214	Office Furniture (except Wood) Manufacturing
		337215	Showcase, Partition, Shelving, and Locker Manufacturing
		322///	Paper Manufacturing
		325992	Photographic Film, Paper, Plate, and Chemical Manufacturing
		33243/	Metal Can, Box, and Other Metal Container Manufacturing
		333///	Machinery Manufacturing
		3352//	Household Appliance Manufacturing
		331318	Other Aluminum Rolling, Drawing, and Extruding
		3314//	Nonferrous Metal (except Aluminum) Production and Processing
		33592/	Communication and Energy Wire and Cable Manufacturing
		335311	Power, Distribution, and Specialty Transformer Manufacturing
		3361//	Motor Vehicle Manufacturing
		3362//	Motor Vehicle Body and Trailer Manufacturing
		3363//	Motor Vehicle Parts Manufacturing
Printing Inks	Employment	32311/	Printing
		3222//	Converted Paper Product Manufacturing
FIFRA Pesticides	Population	--	--
Agricultural Pesticides	Pesticide Use	[b]	
Dry Cleaning	Employment	812320	Dry Cleaning and Laundry Services
Oil & Gas	O&G Well Count	[c]	
Misc. Products	Population	--	--
Fuels & Lighter	Population	--	--

[a]: All employment mapping, expect Allied Paint Products, follows the NAICS mapping from the 2017 NEI (U.S. EPA, 2017). For Allied Paint Products, mapping is allocated based on construction employment.
[b]: Allocation of Agriculture Pesticides emissions follows the mapping from the 2017 NEI (U.S. Geological Survey, Pesticide National Synthesis Project, https://water.usgs.gov/nawqa/pnsp/usage/maps/county-level/; last access: August 31, 2020).
[c]: U.S. Energy Information Administration, The Distribution of U.S. Oil and Natural Gas Wells by Production Rate, Washington, DC, 2019.

Table S7: Observed emission ratios (de Gouw et al., 2017; de Gouw et al., 2018) and inventory emission ratios for Los Angeles County. VCPy: All emissions retrieved from the 2017 NEI, except VCPs, which are replaced using the emissions derived in this study (representative of 2016). 2017 NEI: All emissions retrieved from the 2017 NEI. Emissions consist of all on-road, non-road, non-point, and point sources, as well as biogenic ethanol, methanol, and acetone. Total CO emissions (~320 Gg) include all on-road, non-road, non-point, and point sources.

Compound	Observed [g /g CO]	VCPy [g /g CO]	2017 NEI [g /g CO]
Ethanol	0.0752	0.0567~~0.0555~~	0.0301
Acetone	0.0241	0.0265~~0.0264~~	0.0116
i-Propanol	0.0212	0.0134~~0.0131~~	0.0070
Toluene	0.0112	0.0227~~0.0227~~	0.0169
Propane	0.0211	0.0117~~0.0115~~	0.0074
i-Butane	0.0066	0.0079~~0.0076~~	0.0060
(m + p)-Xylenes	0.0078	0.0090~~0.0091~~	0.0041
n-Butane	0.0103	0.0141~~0.0141~~	0.0116
Methyl Ethyl Ketone	0.0023	0.0044~~0.0044~~	0.0017
Methanol	0.0243	0.0187~~0.0188~~	0.0172
Undecane	0.0018	0.0030~~0.0031~~	0.0012
Octanes	0.0072	0.0031~~0.0031~~	0.0014
Heptane	0.0030	0.0026~~0.0031~~	0.0021
Nonane	0.0015	0.0018~~0.0018~~	0.0006
Hexane	0.0043	0.0045~~0.0051~~	0.0039
Methylcyclohexane	0.0015	0.0015~~0.0020~~	0.0007
Trimethylbenzenes	0.0063	0.0032~~0.0035~~	0.0022
Ethyltoluenes	0.0024	0.0030~~0.0030~~	0.0020
Decane	0.0015	0.0012~~0.0012~~	0.0004
Ethylbenzene	0.0023	0.0025~~0.0026~~	0.0019
n-Pentane	0.0088	0.0046~~0.0046~~	0.0043
Dimethylcyclohexanes	0.0008	0.0004~~0.0004~~	0.0001
Styrene	0.0013	0.0013~~0.0013~~	0.0013
Propylbenzenes	0.0006	0.0007~~0.0007~~	0.0004
o-Xylene	0.0029	0.0019~~0.0019~~	0.0018
Methylacetate	0.0006	0.0003~~0.0003~~	0.0001
2-Methylhexane	0.0021	0.0012~~0.0012~~	0.0011
n-Propanol	0.0007	0.0002~~0.0002~~	0.0001
3-Methylpentane	0.0043	0.0022~~0.0022~~	0.0022
Cyclohexane	0.0016	0.0018~~0.0018~~	0.0017

Table S8: SCC – SPECIATEv5.0 (EPA, 2019b) profile mapping for all non-point sources in the 2017 NEI. A similar mapping scheme was used to additionally speciate 53 on-road SCCs, 57 non-road SCCs, and > 4,500 point SCCs.

SCC	SPECIATE Profile	SCC	SPECIATE Profile	SCC	SPECIATE Profile	SCC	SPECIATE Profile
2401001000	95513	2104006000	0195	2620030000	3002	2810005001	5560
2401005000	2402	2104007000	0195	2102004001	0002	2810035000	5560
2401008000	3135	2302002100	4553	2102004002	0002	2104002000	1185
2401090000	3149	2302002200	4553	2102007000	0003	2601020000	0122
2401100000	3138	2302003000	4652	2304000000	1089	2103002000	1178
2401200000	3138	2302003100	4651	2308000000	1008	2810005000	5560
2420000000	2422	2302003200	4651	2309000000	2466	2830000000	0000
2425000000	1191	2501011011	8870	2312000000	0000	2505020180	2488
2460100000	95509	2501011012	8870	2399000000	0000	2302070000	1188
2460200000	95508	2501011013	8870	2510000000	0000	2862000000	0000
2460400000	95510	2501011014	8870	2620000000	3002	2301000000	2462
2460500000	95512	2501011015	8870	2650000000	3002	2301030000	2462
2460600000	95507	2501012011	8870	2660000000	8870	2635000000	8870
2460800000	95511	2501012012	8870	2810030000	0000	2840000000	2402
2460900000	95512	2501012013	8870	2810040000	5565	2851001000	0000
2461021000	1007	2501012014	8870	2810050000	0000	2501070053	DIESEVP
2461022000	1007	2501012015	8870	2830001000	0000	2810003000	4659
2461850000	CARB3103	2501060053	8870	2102001000	1185	2601000000	122
2415000000	8745	2501060201	8870	2102011000	0004	2601010000	122
2401015000	2405	2501080050	8869	2501060052	8870	2630010000	3003
2401020000	2405	2501080100	8869	2103004000	0002	2302070010	1188
2401025000	2406	2505030120	8870	2301010000	2462	2680002000	0000
2401055000	3149	2610000100	0121	2302000000	4553	2306010100	0026
2401075000	2414	2610000400	0121	2302050000	1188	2620010000	3002
2401080000	2415	2610000500	0121	2302070005	1188	2102010000	0004
2401070000	3131	2610030000	0121	2302080000	1188	2310010100	0003
2401030000	2552	2630020000	3003	2306010000	0026	2310010200	2487
2401040000	2408	2680003000	8933	2505010000	2489	2310011001	1011
2401060000	2411	2810025000	4553	2505020000	0305	2310011201	2487
2401065000	3138	2810060100	0000	2610000300	0121	2310011501	1011
2401085000	2416	2810060200	0000	2640000000	0000	2310011502	1011
2440000000	95512	2104004000	0002	2680001000	0000	2310011503	1011
2401005700	2402	2104011000	0002	2102005000	0001	2310011505	1011
2401010000	3137	2501050120	8869	2302002000	4553	2310021010	2487
2420000999	2422	2501055120	8869	2305000000	0000	2310021030	2487
2461023000	1007	2505040120	8869	2307000000	2405	2310021100	0003
2401050000	3127	2102002000	1185	2325030000	0000	2310021300	8949
2420000055	0085	2102006000	0003	2501995120	8762	2310021302	1001
2401045000	2409	2102008000	4642	2302070001	1188	2310021351	1001
2425010000	2543	2103004001	0002	2301020000	1092	2310021400	0003
2425020000	2544	2103004002	0002	2801520000	3161	2310021501	8949
2425030000	2545	2103007000	0003	2103005000	0001	2310021502	8949
2425040000	1086	2103008000	4642	2103001000	1178	2310021503	8949
2461020000	1007	2103011000	0002	2520010000	0000	2310021505	8949
2461800001	CARB3103	2302010000	4553	2501080201	8762	2310021506	8949
2461800002	CARB3103	2501060051	8870	2505020030	0305	2310021603	8949
2440020000	95507	2102004000	0002	2505020060	0305	2310023300	8950
2460000000	95512	2103006000	0003	2505020090	2488	2310023302	1001
2401035000	3137	2311030000	0000	2505020120	8869	2310023351	1001

2461100000	0000
2310023511	8950
2310023512	8950
2310023513	8950
2310023515	8950
2310023516	8950
2310000220	0008
2310111100	1011
2310111700	1011
2310000660	0008
2310023600	8950
2310121700	8949
2310001000	8949
2310010300	8949
2310021251	1001
2310000553	1207
2310011600	1001
2310021202	1001
2310111401	1011
2310121401	8949
2310121100	8949
2310021303	1001
2310022420	8949
2310002401	8949

2325000000	0000
2310000552	1207
2310023100	0003
2310023202	1001
2310023251	1001
2310023310	8950
2310023603	8950
2310011500	95399
2310020000	8949
2310022000	8949
2310010700	DJVNT_R
2310011450	DJVNT_R
2310021310	DJVNT_R
2310021509	DJVNT_R
2310021700	1001
2310030220	FLR99
2310030300	1207
2310030400	2487
2310111701	FLR99
2310321603	DJVNT_R
2310400220	0008
2310022010	95109a
2310022090	0003
2310022506	1010

2505020150	100
2310421100	0003
2310011020	2487
2310021109	1001
2310021209	1001
2310021309	1001
2310021600	SSJCO_R
2310021602	SSJCO_R
2310000551	1207
2310023010	2487
2310023030	2487
2310021102	1001
2310023606	SSJCB_R
2310023509	8950
2310023102	1001
2310021500	FLR99
2310300220	0008
2310321010	2487
2310321100	0003
2310321400	0003
2310421010	2487
2310021802	95417
2310021801	95417
2310021803	FLR99

2310023400	0003
2310421603	8949
2310421400	0003
2310020600	1001
2310011504	1011
2310011506	1011
2310021103	1001
2310021402	1001
2310021403	1001
2310021450	8949
2310021504	8949
2310021101	1001
2310021203	1001
2310021301	1001
2310002000	8949
2310002421	8949
2310012000	1011
2310012020	95087a
2310012526	1011
2310022105	0008
2310112401	1011
2310011100	0003
2310000230	0008

Table S9: National-level emission rates [kg $person^{-1}$ $year^{-1}$] for the top-200 compounds emitted from VCPs, as predicted by VCPy.

Compound	Emissions	Compound	Emissions	Compound	Emissions	Compound	Emissions
Ethanol	1.6519	C6 Cycloalkanes	0.0238	C10 Trialkylbenzenes	0.0052	cis-1,cis-3,5-trimethylcyclohexane	0.0022
Acetone	0.8506	d-Limonene	0.0235	Aliphatics	0.0052	trans,cis-1,2,4-trimethylcyclohexane	0.0022
Isopropyl Alcohol	0.4274	C15 Cycloalkanes	0.0223	Isobutyl Acetate	0.0051	1,1,3-trimethylcyclohexane	0.0022
Toluene	0.3704	Other, Misc. VOC Compounds Aggregated In Profile	0.0219	C11 Tetrasubstituted Benzenes	0.0051	1,1,3-trimethylcyclopentane	0.0022
n-Tetradecane	0.3632	Branched C10 Alkanes	0.0208	2,6-dimethylnonane	0.0050	4-methyldecane	0.0022
Fragrances	0.3444	C10 Cycloalkanes	0.0208	Methyl Acetate	0.0050	1,2,3-Trimethylbenzene	0.0021
Propane	0.3365	Witch Hazel	0.0197	3-methylheptane	0.0049	5-methyldecane	0.0021
Volatile Methyl Siloxanes	0.3024	2-Amino-2-Methyl-1-Propanol	0.0188	2-methylhexane	0.0049	trans,trans-1,3,5-trimethylcyclohexane	0.0021
Isobutane	0.2814	n-Tridecane	0.0186	1-Tetradecene	0.0049	Butylcyclohexane	0.0021
Propylene Glycol	0.2488	n-Heptane	0.0167	C15 Branched Alkanes	0.0048	4-methylheptane	0.0021
2,2,4-Trimethyl-1,3-Pentanediol Isobutyrate (Texanol)	0.2195	Hexane	0.0162	Other, Lumped VOCs, Individually < 2% Of Category	0.0047	2,3-Dimethylbutane	0.0021
Ethylene Glycol	0.2144	Propylene Glycol Monomethyl Ether Acetate	0.0159	1,2,4-trimethylcyclopentane	0.0046	C11 Tetralin or Indane	0.0020
Xylenes	0.1941	Benzene	0.0155	1,2-dimethylcyclopentane	0.0046	Trichloroethylene	0.0020
n-Butane	0.1916	C7 Cycloalkanes	0.0151	Turpentine	0.0046	Diisopropyl Adipate	0.0020
Methanol	0.1772	C14 Cycloalkanes	0.0150	Diethylene Glycol	0.0045	1,3-diethylbenzene (meta)	0.0019
Ethylene Glycol Monobutyl Ether	0.1588	n-Pentane	0.0149	C12 Naphthalenes	0.0045	2,2-Dimethylbutane	0.0018
Branched C12 Alkanes	0.1568	Branched C7 Alkanes	0.0146	Propylene Glycol Monomethyl Ether (1-Methoxy-2-propanol)	0.0045	Dihydroxyacetone	0.0018
n-Undecane	0.1461	Ethyl Cyanoacrylate	0.0139	n-Propyl Alcohol	0.0044	Dipropylene Glycol	0.0017
Methylene Chloride (Dichloromethane)	0.1291	C13 Cycloalkanes	0.0129	trans,trans-1,2,4-trimethylcyclohexane	0.0043	Dimethyl Succinate	0.0016
Methyl Ethyl Ketone (2-Butanone)	0.1287	Isopropyl acetate	0.0126	Diethylene Glycol Monoethyl Ether	0.0043	Ethylene Glycol Monopropyl Ether	0.0015
Dimethyl Ether	0.1186	Propylene Glycol N-Propyl Ether	0.0124	C5 Branched Alkanes	0.0042	Triethanolamine	0.0015
n-Dodecane	0.1100	1,1,1,2-Tetrafluoroethane (HFC-134a)	0.0124	Hexadecane	0.0040	p-Xylene	0.0015
1,1-Difluoroethane (HFC-152a)	0.1082	Hydrocarbon Propellant (LPG)	0.0122	m-Xylene	0.0040	C11 Dialkyl Benzenes	0.0014
Hydrocarbon Propellant (LPG, Sweetened)	0.0914	Cyclopentane	0.0118	Branched C17 Alkanes	0.0040	C14 Branched Alkanes	0.0014
C11 Cycloalkanes	0.0898	N,N-Diethyl-M-Toluamide	0.0115	Methyl Amyl Ketone	0.0040	Methyl Ethyl Ketoxime	0.0014
n-Octane	0.0803	Styrene	0.0112	1,2,4-Trimethylbenzene	0.0038	2,2,4,6,6-Pentamethylheptane	0.0014
C12 Cycloalkanes	0.0758	C16 Cycloalkanes	0.0109	Misc. Oxygenated Compounds	0.0037	(2-methylpropyl)benzene (or isobutylbenzene)	0.0014
C13 Branched Alkanes	0.0747	1,3,5-trimethylbenzene	0.0107	C10 Dialkyl Benzenes	0.0035	Toluene	0.0013
Branched C9 Alkanes	0.0700	N-Methylpyrrolidinone	0.0107	Dipropylene Glycol Monopropyl Ether	0.0034	Methyl Propyl Ketone (2-Pentanone)	0.0013

C8 Cycloalkanes	0.0617	Aggregated Vocs < 1.0%	0.0104	Phenoxyethanol	0.0033	Methyltriacetoxysilane	0.0013
Pine Oil	0.0605	C11 Trialkyl Benzenes	0.0101	Diethanolamine	0.0033	Ethyltriacetoxysilane	0.0012
n-Nonane	0.0551	Benzyl Alcohol	0.0099	m-Xylene	0.0032	Hexylene Glycol (2-Methyl-2,4-Pentanediol)	0.0012
Branched C11 alkanes	0.0538	C12 Trisubstituted Benzenes	0.0092	Diisobutyl Ketone	0.0031	Isobutyl Alcohol	0.0012
Ethyl Acetate	0.0481	cis-1,3-dimethylcyclohexane	0.0090	3-Methylpentane	0.0030	Ethylbenzene	0.0012
Perchloroethylene (Tetrachloroethene)	0.0481	White Mineral Oil	0.0089	Methyl Methacrylate	0.0030	1,2-diethylbenzene (ortho)	0.0012
C9 Cycloalkanes	0.0474	Diethylene Glycol Monomethyl Ether	0.0086	1-Ethyl-2-Propyl Cyclohexane	0.0029	Diacetone Alcohol	0.0012
n-Decane	0.0445	o-Xylene	0.0085	Dimethyl Adipate	0.0028	Tetramethylbenzenes	0.0012
n-Heptane	0.0422	n-Pentadecane	0.0083	Ethylcyclohexane	0.0028	1,3,5-Trimethylbenzene	0.0011
Branched C8 Alkanes	0.0407	2-Methylheptane	0.0076	trans-1,4-dimethylcyclohexane	0.0027	Misc. Hydrocarbon Propellants	0.0011
Ethanolamine	0.0381	C16 Branched Alkanes	0.0072	Cumene	0.0027	1,4-diethylbenzene (para)	0.0010
Methylcyclohexane	0.0369	o-Ethyltoluene	0.0072	trans-1,3-dimethylcyclohexane	0.0027	Diisopropylene glycol	0.0010
Dipropylene Glycol Monomethyl Ether	0.0307	2-Methylpentane	0.0071	4-methylnonane	0.0026	C13 Naphthalenes	0.0010
Branched C6 Alkanes	0.0306	Voc Ingredients < 0.1%	0.0068	Cyclohexane	0.0026	Ethylene Glycol Monohexyl Ether	0.0010
n-Hexane	0.0302	Glycerol	0.0067	2-methyldecane	0.0024	Tetrahydrofuran	0.0010
Diethylene Glycol Monobutyl Ether	0.0297	n-Propylbenzene	0.0065	2-Ethylhexyl Benzoate	0.0024	Cyclohexanol	0.0010
Ethyl Benzene	0.0286	Propylene Glycol Butyl Ether (1-Butoxy-2-Propanol)	0.0063	trans 1-methyl-3-propyl cyclohexane	0.0024	Methylindans	0.0010
m-Ethyltoluene	0.0281	1,2,3-trimethylbenzene	0.0059	3-methyldecane	0.0023	Diethyl Phthalate	0.0010
1,2,4-Trimethylbenzene	0.0270	Ethyl-3-Ethoxypropionate	0.0056	2,6-dimethylheptane	0.0023	Isopentane	0.0010
N-Butyl Acetate	0.0253	Glycol Ether Dpnb (1-(2-Butoxy-1-Methylethoxy)-2-Propanol)	0.0056	Misc. Esters	0.0023	Triethylene Glycol	0.0009
Methyl Isobutyl Ketone (Hexone)	0.0248	n-Butyl Alcohol	0.0055	Butyl Acrylate	0.0022	Pentanedioic Acid, Dimethyl Ester	0.0009

Table S10: Tabulation of Fig. 4 from main text.

Product Use Categories (PUCs)	**2.5th**	**25th**	**Mean**	**75th**	**97.5th**
Cleaning Products	1.25~~1.25~~	1.72~~1.73~~	2.02~~2.03~~	2.3~~2.31~~	2.94~~2.96~~
Personal Care Products	1.32~~1.31~~	1.76~~1.76~~	2.01~~2.02~~	2.25~~2.27~~	2.81~~2.80~~
Adhesives & Sealants	0.51~~0.50~~	0.66~~0.66~~	0.76~~0.76~~	0.85~~0.85~~	1.05~~1.04~~
Paints & Coatings	2.53~~2.73~~	2.92~~3.13~~	3.12~~3.35~~	3.34~~3.57~~	3.77~~4.03~~
Printing Inks	0.54~~0.53~~	0.7~~0.70~~	0.8~~0.80~~	0.89~~0.89~~	1.1~~1.10~~
Pesticides & FIFRA Products	0.40~~0.41~~	0.52~~0.52~~	0.58~~0.58~~	0.65~~0.65~~	0.78~~0.79~~
Other	0.02~~0.02~~	0.06~~0.06~~	0.16~~0.16~~	0.24~~0.24~~	0.46~~0.46~~
Total	8.07~~8.27~~	8.98~~9.23~~	9.46~~9.71~~	9.96~~10.20~~	10.94~~11.23~~

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Table S11: Mapping of all sub-PUCs to equivalent Source Classification Codes (SCCs).

PUC	SCC	SCC Description	VCPy sub-PUC
Cleaning Products	2460200000	All Household Products	Detergents & Soaps
	2415000000	Degreasing	General Cleaners
	2460400000	All Automotive Aftermarket Products	
Personal Care Products	2460100000	All Personal Care Products	Daily Use Products
			Short Use Products
Adhesives & Sealants	2460600000	All Adhesives and Sealants	Adhesives and Sealants
Paints & Coatings	2401001000	Architectural Coatings	Architectural Coatings
	2460500000	All Coatings and Related Products	Aerosol Coatings
	2401005000	Auto Refinishing	Industrial Coatings
	2401008000	Traffic Markings	
	2401015000	Factory Finished Wood	
	2401020000	Wood Furniture	
	2401025000	Metal Furniture	
	2401030000	Paper	
	2401040000	Metal Cans	
	2401055000	Machinery and Equipment	
	2401060000	Large Appliances	
	2401065000	Electronic and Other Electrical	
	2401070000	Motor Vehicles	
	2401075000	Aircraft	
	2401085000	Railroad	
	2401080000	Marine	
	2401090000	Misc. Manufacturing	
	2401100000	Industrial Maintenance Coatings	
	2401200000	Other Special Purpose Coatings	
	2402000000	Paint Strippers	Allied Paint Products
Printing Inks	2425000000	Graphic Arts, employment	Printing Inks
Pesticides & FIFRA Products	2460800000	All FIFRA Related Products	FIFRA Pesticides
	2461850000	Pesticide Application: Agricultural	Agricultural Pesticides
Dry Cleaning	2420000000	Dry Cleaning	Dry Cleaning
Oil & Gas	n/a	n/a	Oil & Gas
Misc. Products	2460900000	Miscellaneous Products: NEC	Misc. Products
Fuels and Lighter	n/a	n/a	Fuels and Lighter

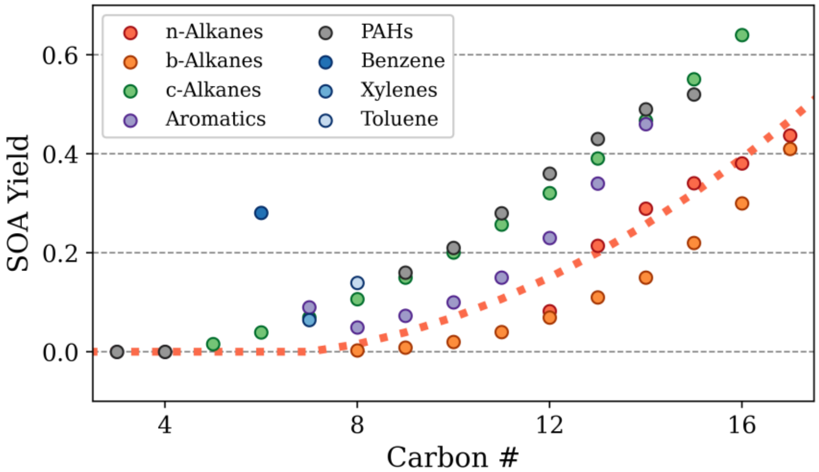

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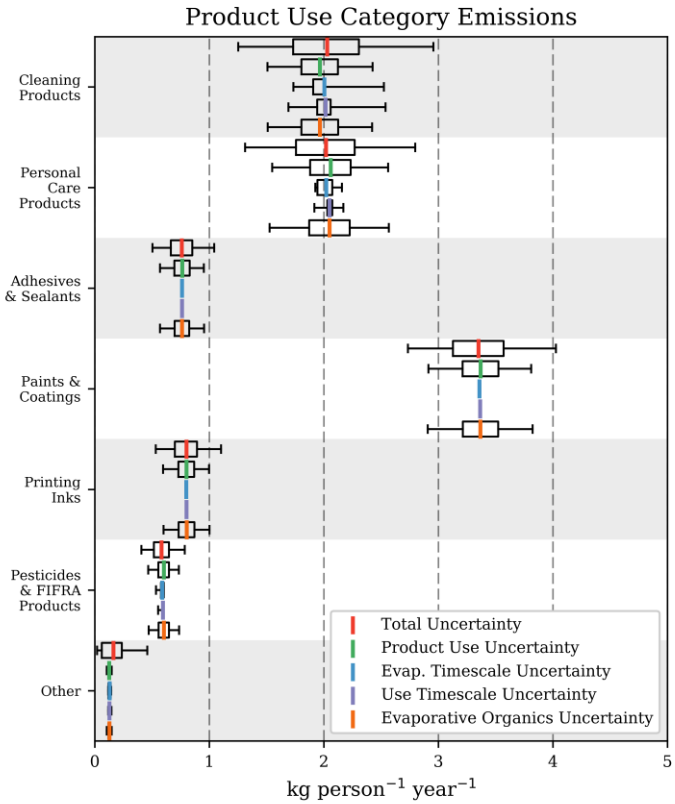
Figure S1: Summary of SOA yields by compound class. Several references are used to construct these values and are described in the main text.


Product Use Category Emissions
Cleaning Products
Personal Care Products
Adhesives & Sealants
Paints & Coatings
Printing Inks
Pesticides & FIFRA Products
Other
Total Uncertainty
Product Use Uncertainty
Evap. Timescale Uncertainty
Use Timescale Uncertainty
Evaporative Organics Uncertainty
0
1
2
3
4
5
kg person^{-1} year^{-1}

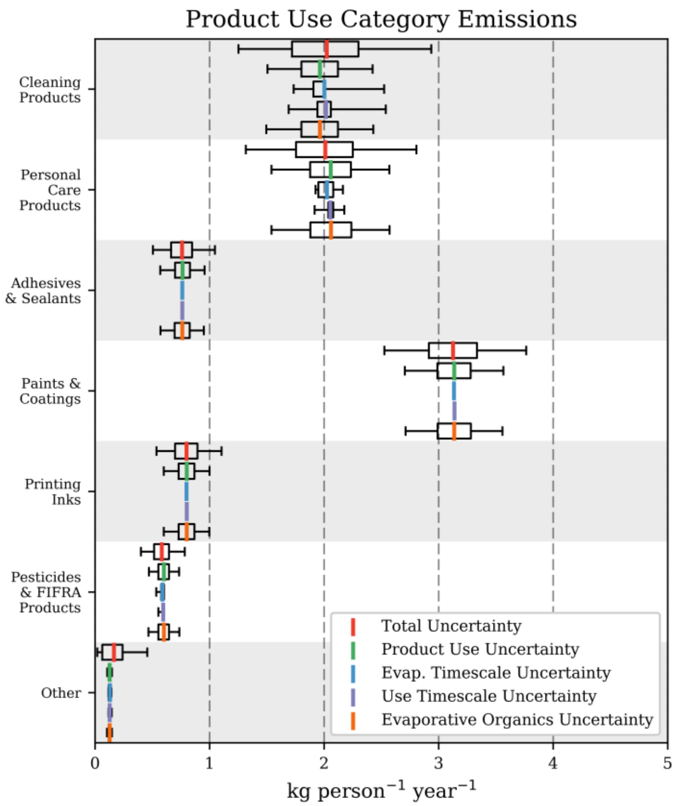
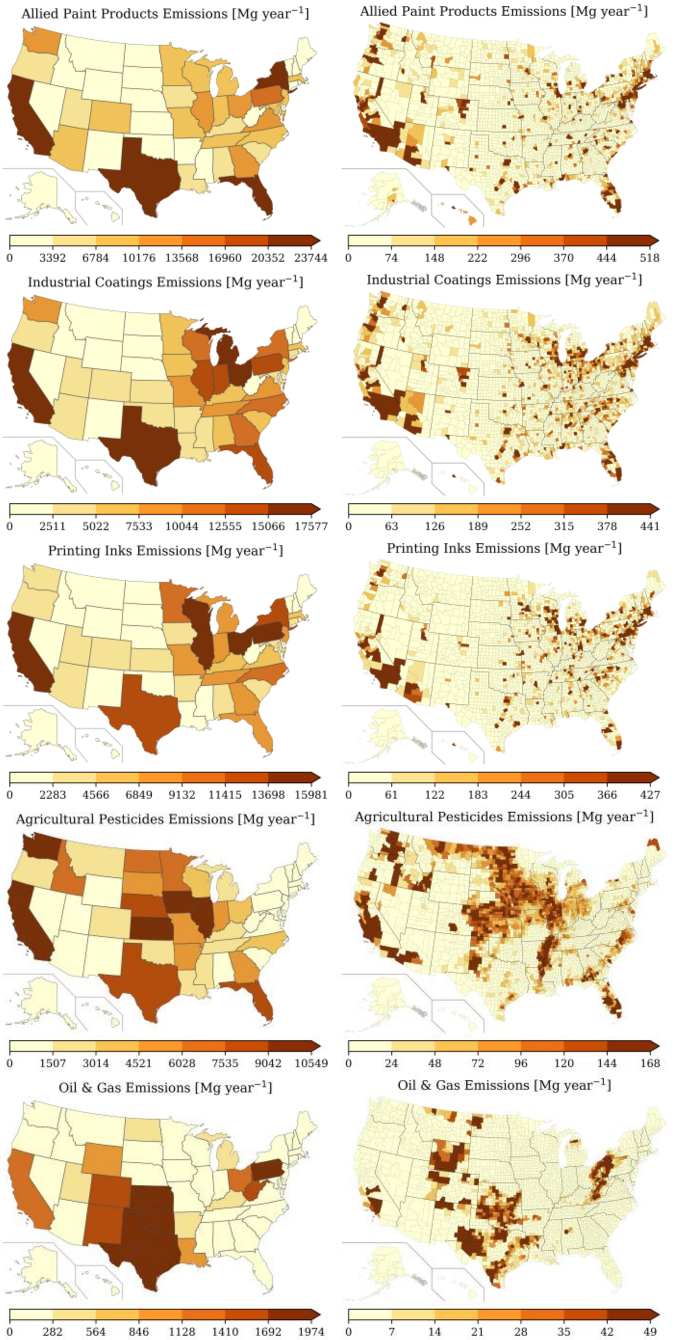


Figure S2: Monte Carlo sensitivity results: mean, interquartile range, and 95% confidence intervals for emission rates for six major PUCs and the sum of all others. Red: Estimate considering uncertainty in product usage, evaporation timescale, use timescale, and controls. Green: MC simulations that only perturb product usage uncertainties. Blue: MC simulations that only perturb evaporation timescale uncertainties. Purple: MC simulations that only perturb use timescale uncertainties. Orange: MC simulations that only perturb evaporative organic uncertainties.


Allied Paint Products Emissions [Mg year^{-1}]
0 3392 6784 10176 13568 16960 20352 23744
Allied Paint Products Emissions [Mg year^{-1}]
0 74 148 222 296 370 444 518
Industrial Coatings Emissions [Mg year^{-1}]
0 2511 5022 7533 10044 12555 15066 17577
Industrial Coatings Emissions [Mg year^{-1}]
0 63 126 189 252 315 378 441
Printing Inks Emissions [Mg year^{-1}]
0 2283 4566 6849 9132 11415 13698 15981
Printing Inks Emissions [Mg year^{-1}]
0 61 122 183 244 305 366 427
Agricultural Pesticides Emissions [Mg year^{-1}]
0 1507 3014 4521 6028 7535 9042 10549
Agricultural Pesticides Emissions [Mg year^{-1}]
0 24 48 72 96 120 144 168
Oil & Gas Emissions [Mg year^{-1}]
0 282 564 846 1128 1410 1692 1974
Oil & Gas Emissions [Mg year^{-1}]
0 7 14 21 28 35 42 49

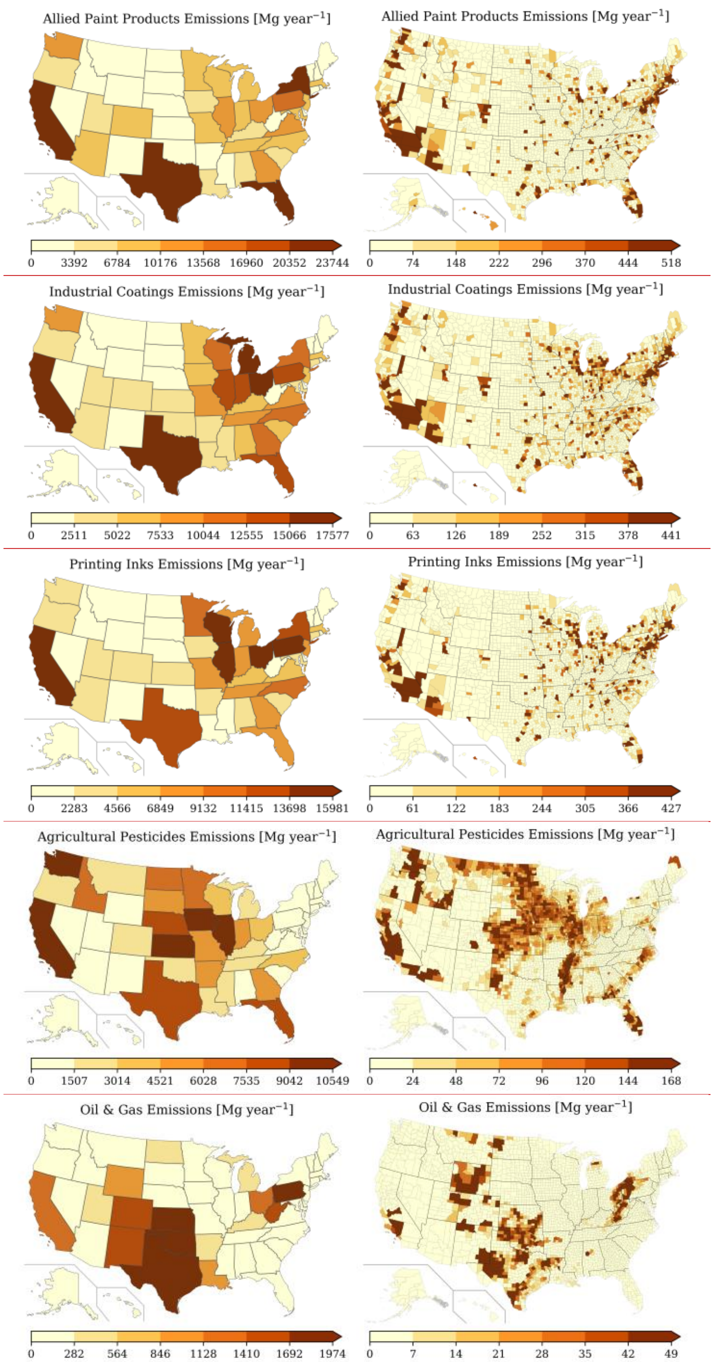
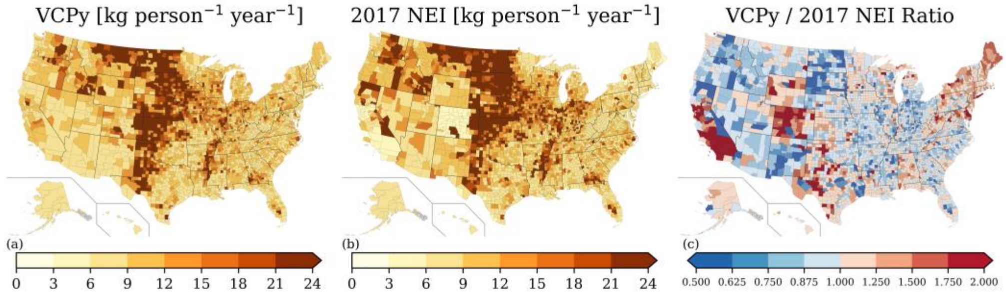

Allied Paint Products Emissions [Mg year^{-1}]
0 3392 6784 10176 13568 16960 20352 23744
Allied Paint Products Emissions [Mg year^{-1}]
0 74 148 222 296 370 444 518
Industrial Coatings Emissions [Mg year^{-1}]
0 2511 5022 7533 10044 12555 15066 17577
Industrial Coatings Emissions [Mg year^{-1}]
0 63 126 189 252 315 378 441
Printing Inks Emissions [Mg year^{-1}]
0 2283 4566 6849 9132 11415 13698 15981
Printing Inks Emissions [Mg year^{-1}]
0 61 122 183 244 305 366 427
Agricultural Pesticides Emissions [Mg year^{-1}]
0 1507 3014 4521 6028 7535 9042 10549
Agricultural Pesticides Emissions [Mg year^{-1}]
0 24 48 72 96 120 144 168
Oil & Gas Emissions [Mg year^{-1}]
0 282 564 846 1128 1410 1692 1974
Oil & Gas Emissions [Mg year^{-1}]
0 7 14 21 28 35 42 49

Figure S3: State and County-level emissions for select sub-PUCs.



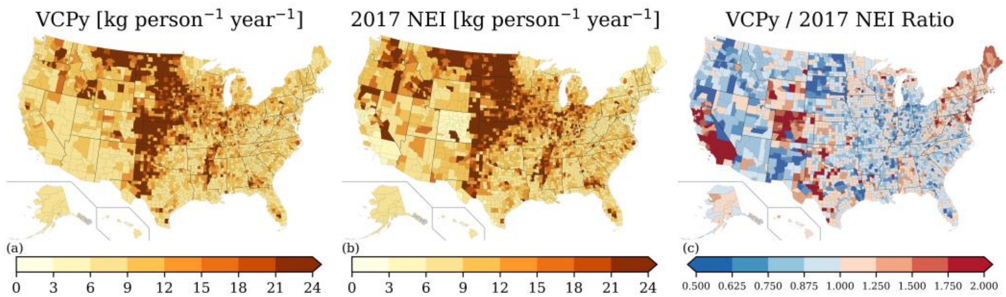
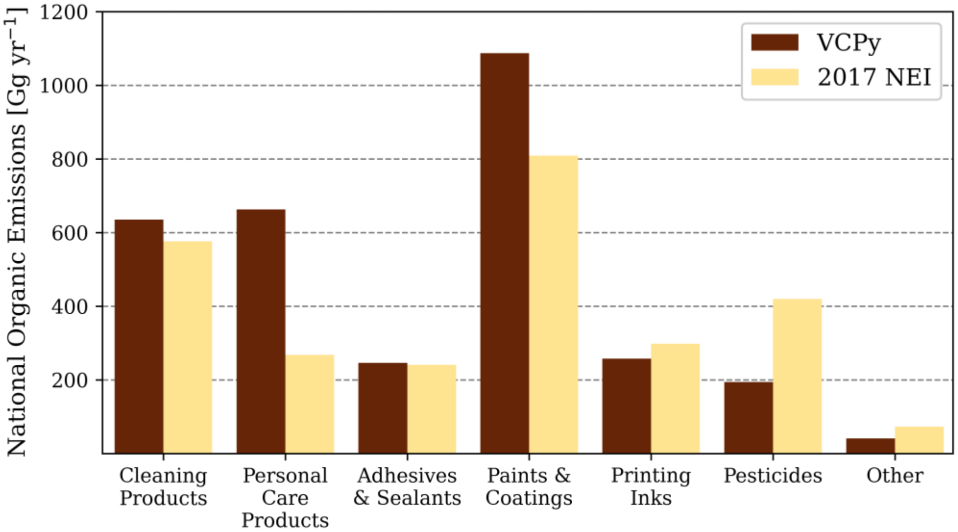


Figure S4: (a) County-level per-capita VCP emissions from the VCPy inventory (same as right panel of Fig. 5 in main text), (b) County-level per-capita VCP emissions from the 2017 NEI, and (c) County-level ratio of VCPy / 2017 NEI VCP emissions.



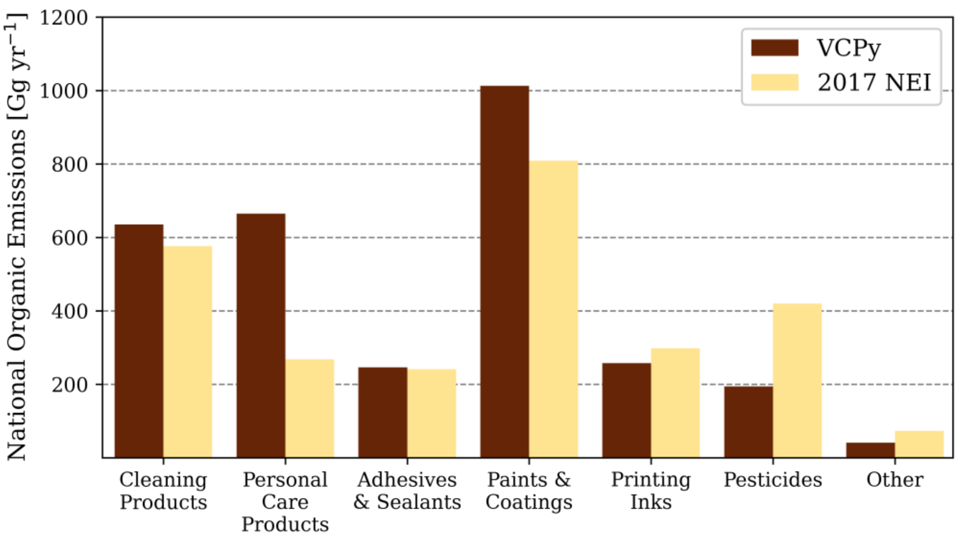
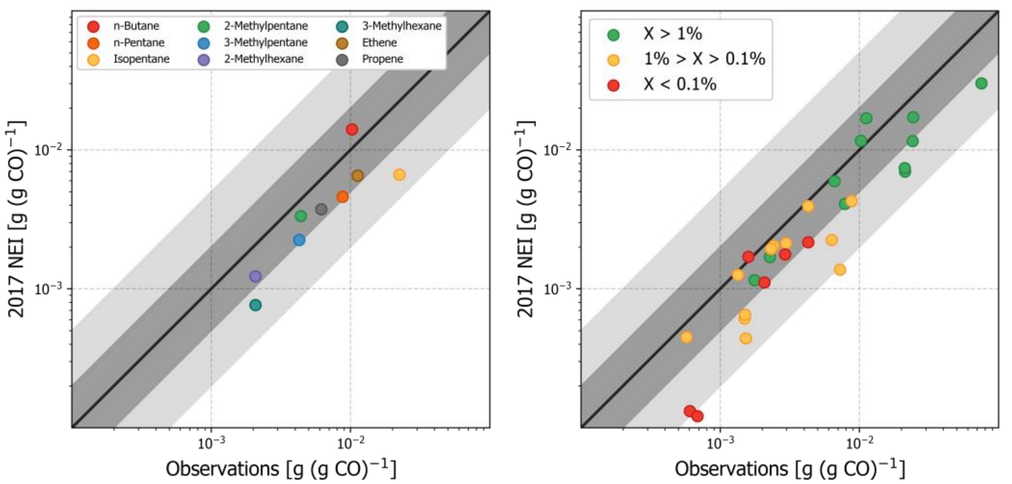


Figure S5: Product Use Category comparison of national-level emissions from the VCPy and 2017 NEI inventories for VCPs. For "Other," asphalt emissions in the 2017 NEI are excluded as those emissions are not quantified in the VCPy inventory.



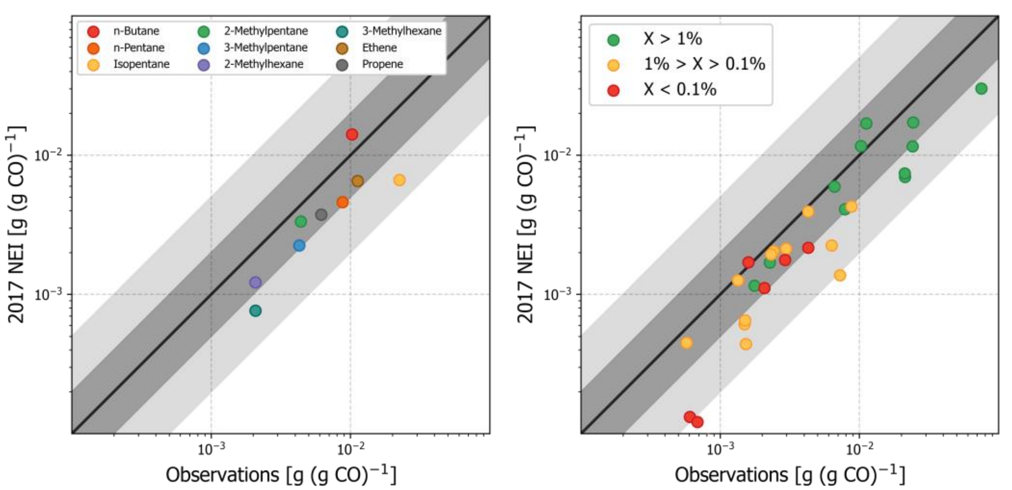
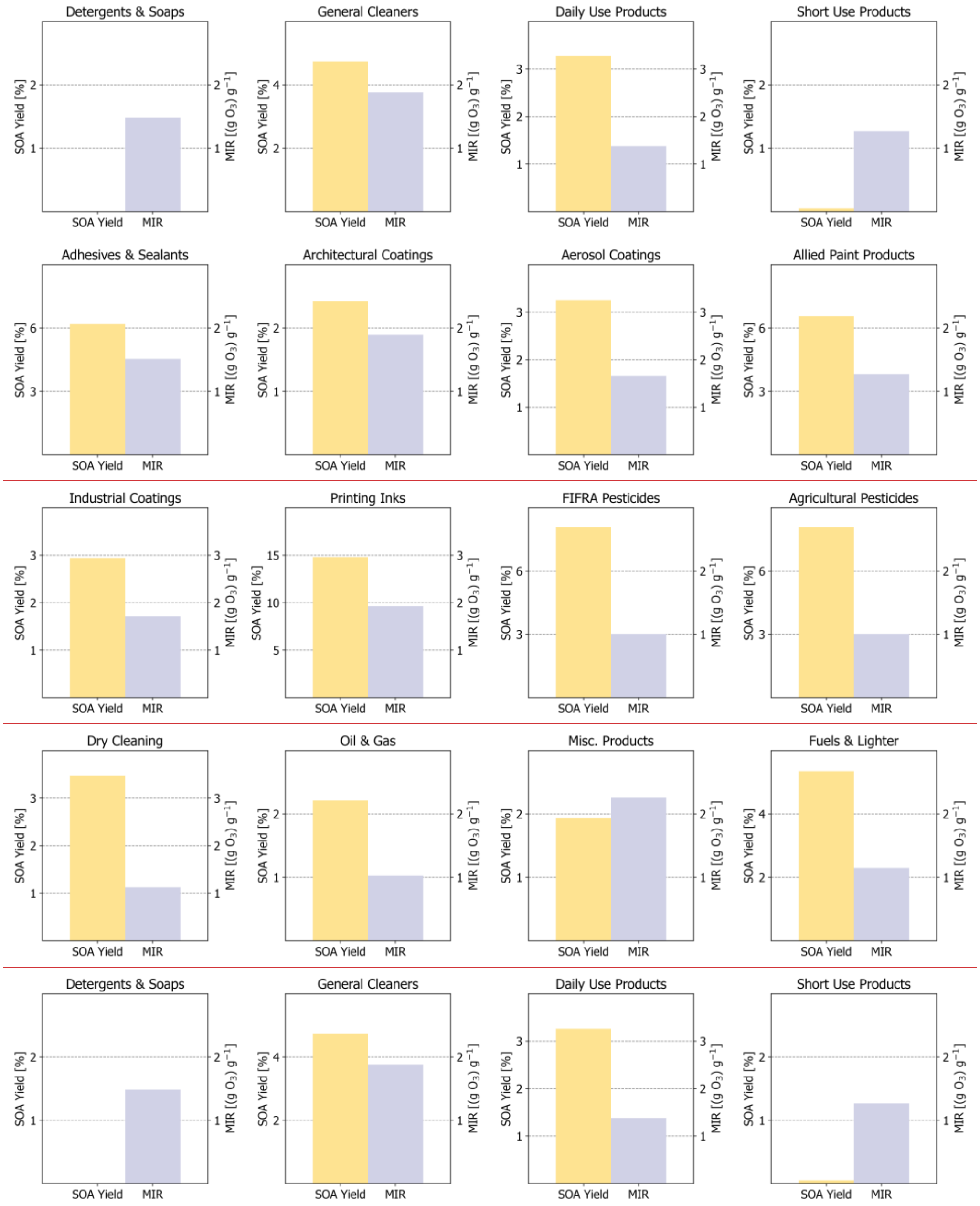


Figure S6: (Left) Evaluation of organic emission ratios of species that feature high emission factors from mobile sources in Los Angeles County using observed emission ratios from summer 2010. (Right) Evaluation of 2017 NEI organic emission ratios in Los Angeles County using observed emission ratios from summer 2010. The scatter point colors represent the relative abundance of each compound in the complete VCP sector. For example, all green points represent compounds that are > 1% of the total VCP emissions in Los Angeles County. Black line – 1:1; Dark grey shading – 2:1; Light grey shading – 5:1.


Detergents & Soaps
General Cleaners
Daily Use Products
Short Use Products
Adhesives & Sealants
Architectural Coatings
Aerosol Coatings
Allied Paint Products
Industrial Coatings
Printing Inks
FIFRA Pesticides
Agricultural Pesticides
Dry Cleaning
Oil & Gas
Misc. Products
Fuels & Lighter
Detergents & Soaps
General Cleaners
Daily Use Products
Short Use Products
SOA Yield [%]
MIR [(g O$_3$) g^{-1}]
SOA Yield
MIR

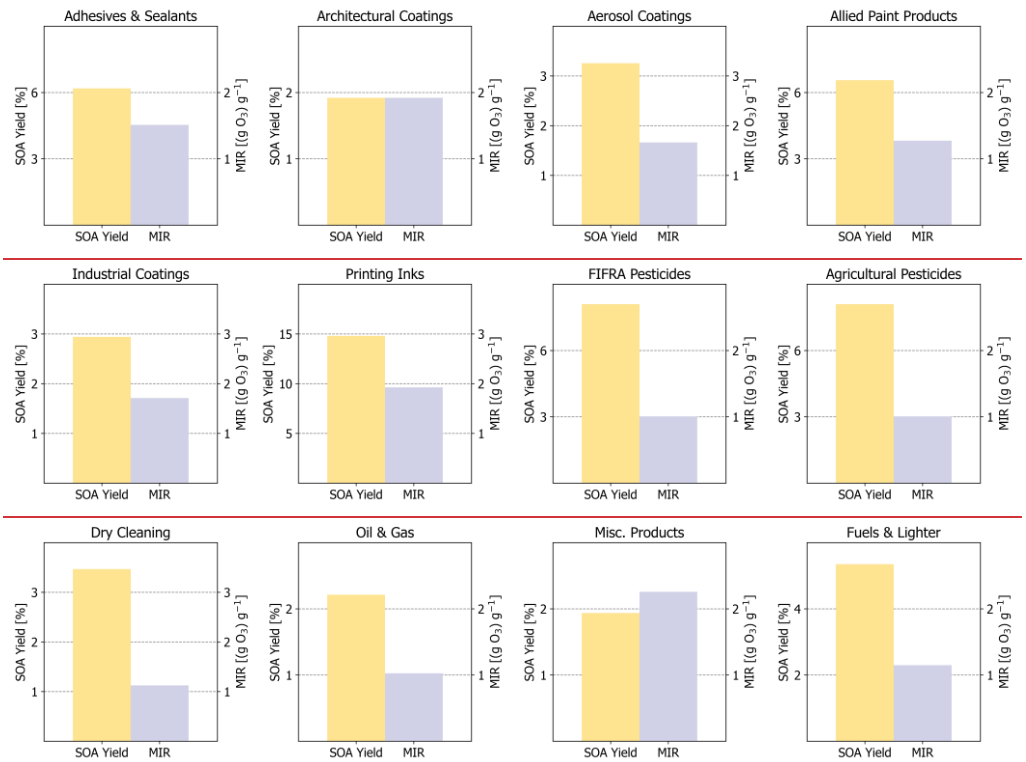
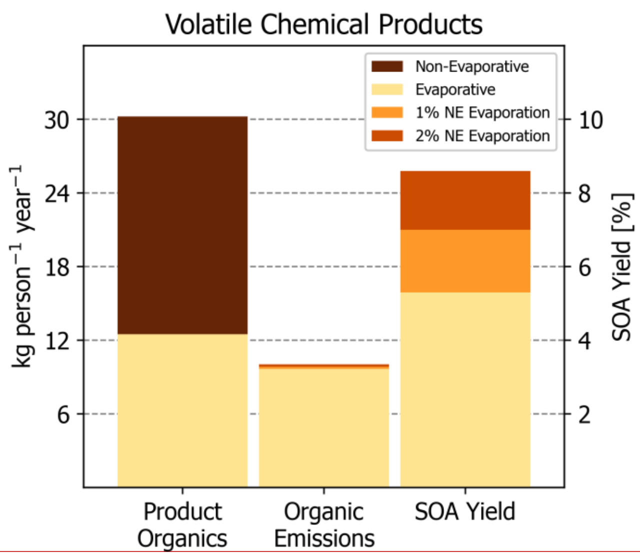


Figure S7: Effective SOA yield and MIR for all sub-PUCs.



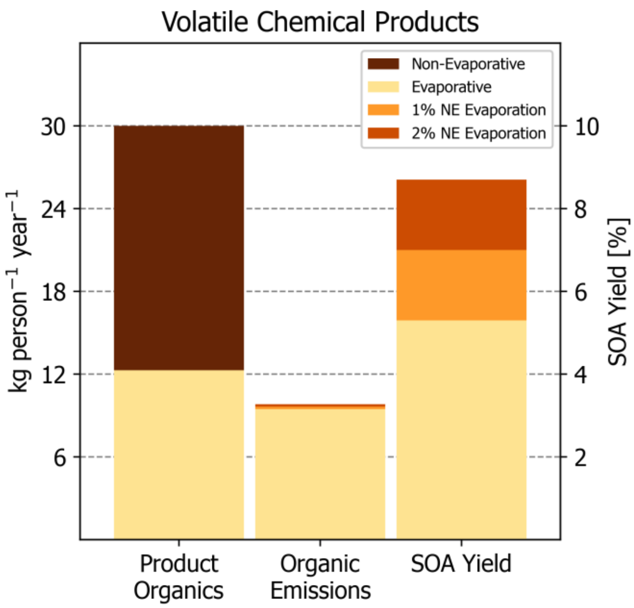


Figure S8: Total product organics, organic emissions, and sector-wide effective SOA yields resulting from adjusted non-evaporative assumptions. The two sensitivity tests are assuming 1% and 2% of all non-evaporative organic mass in VCPs evaporates and forms SOA with 100% efficiency.