

February 27, 2018

Re: Manuscript ID: acp-2017-761, “Considering the future of anthropogenic gas-phase organic compound emissions and the increasing influence of non-combustion sources on urban air quality”

We sincerely thank the reviewer and editor for supporting our manuscript, and insights which have helped us in improving our work. After having gone through the comments in great detail, we are writing to present our revisions/replies. The reviewer comments are presented below and our responses are shown in italics.

Reviewer 1:

This paper presents a detailed look at VOCs, specifically delving into their representation in emission inventories, what details are (or are not reported) relating to composition, and their ozone and SOA formation potential. The authors spend some time as well to investigate intermediate-volatility and semivolatile organic compounds that are typically not included in standard emission inventories and are exempt from emissions targets, but in need of greater attention in the future. Finally, the misattribution of some of the VOCs to fossil fuel from e.g., vehicles, while some likely originates from e.g., petrochemical processes, which also have a fossil fuel feedstock, needs to be considered in source attribution.

This paper adds interesting and novel points to the literature and is generally well written and well organized, if a bit long and drawn out at points. I would recommend publication after addressing some minor points for revision.

Comments:

L116-118: the use of the ‘pp’ prefix seems unnecessary given that it is then only used ca. 20 pages later and only twice. When it is initially explained here the point seems odd and unnecessary. When it is then applied much later, as a reader you have to go back and figure out where that came from so that you are clear what is meant. It would be clearer to just remove the initial text and just explain the ‘products and processes’ source when discussed at that one point later in the text.

Response: We agree that the prefix ‘pp’ could be dispensed with given its minimal use in the manuscript. It is now completely removed from the revised manuscript. In places where it previously appeared, ‘pp’ is replaced with the term ‘products/process-related’ which has been frequently used in the manuscript.

Overall, the paper has both appendixes and SI. It is not clear what the criteria are for having information in an appendix versus in the SI. I would suggest to combine these all into the SI.

Response: We understand the reviewer's concern and reviewed the ACP author guidelines on the matter. Appendixes A and B in our manuscript provide valuable and directly relevant information to the average reader regarding the historical context and significance of product/process-related emissions over long timescales, but don't seem to qualify as SI based on the author guidelines. They are shown in the appendixes rather than the main text to help maintain a reasonable length for the main body of the manuscript. Hence, we prefer to keep the two appendixes with the published manuscript while carefully complying with the ACP author guidelines. We defer to the editor's judgement on this matter.

Furthermore, there are a number of figures and tables in the SI that are referred to quite often in the text. I would suggest that the authors consider moving some of these to the main text. Supplemental information should really only be for information that provides a level of detail that most readers will not be interested in, but is important for those who e.g., might want to apply some of the methods to their own analysis or really dig into the details. For example, I would suggest moving e.g., Table S2 or Figure S3 to the main text. If there is a concern that there are too many figures/tables in the main text, you could move figure 4 to the SI, which I find adds less than some of the other figures.

Response: We agree with the reviewer that any SI figures and tables that are referenced frequently in the main text should appear in the main text, but we reviewed the manuscript's references to SI figures/tables, and we wish to highlight that tables and figures listed in the SI are individually cited once or twice at most in the main manuscript. The emission timescales shown in Figure 4 are very important for expanding understanding of the timescales of emissions and long-term I/SVOC emissions from products and processes, and their lack of consideration in existing inventories/models, which is part of the core theme of our manuscript. Additionally, Figure 4 is referenced 5 times in the manuscript. Therefore, considering these reasons and reviewer's concern regarding the manuscript length, we have decided to keep the figures and tables as it is.

L245-246: the SOA yield estimate – what is this value based on? And is this in general or specifically for the unstudied compounds? More information would be good here to clarify what is meant and where these numbers come from.

Response: We understand that this is an important concern. We have clarified this point in the text and directed the reader to Table S6 where the rules that we used for assigning the SOA yield values are listed in great detail. These yield values are applicable to unstudied organic compounds and are based on trends observed in published yields of such organic compounds that are comparable in terms of molecular structure. In cases of compounds where exact SOA yields are straight up available from scientific literature, they are directly used.

L248-250: can you give some information as to the magnitude of this? Are we talking an order of magnitude or 10%? A rough indication is fine.

Response: Unfortunately, it is difficult to generalize and constrain based on the current state of knowledge, especially relevant to urban air quality. There are too many different precursors to estimate since the impact is likely to be precursor-dependent and site-dependent.

L269-270: Please add some more information to clarify how the composition is calculated. The source categories are based on the CARB inventory, but where is the speciation coming from? Or are you explaining what you will be doing with the speciate database and then MSDSs etc that is explored in the following sections. This isn't clear.

Response: We thank the reviewer for pointing this out. We include a more detailed explanation, found in lines 223-226 (and there is additional detail in lines 269-325) of the (revised) manuscript. We combine source categories from the CARB Almanac emissions inventory with the chemical profiles for those sources listed in the EPA SPECIATE 4.4 database to calculate compound-specific emissions in the south coast air basin (SoCAB). We have also added an SI figure (figure S5) for comparison using an alternative speciation profile (applicable to limited years), with very similar results. The MSDSs were not used to establish source profiles. We have made revisions to clarify the primary data source in the section of concern, and the relevant section that follows "Analysis of emissions inventories" (lines 269-325).

L313-315: Can you provide some more information as to the comparison to the speciate profiles? Percent of single-ring aromatic content is listed in the text for a couple of products and for more in Table 2, but a comparison percent is not provided for the speciate source profiles. There is a comparison listed, but this seems to be from a different source and not speciate. If this is the comparison to speciate, please be explicit about this. One could also consider adding information to the table.

Response: In lines 310-311, we state that ~21% of emissions from paints are single-ring aromatics. However, we understand the need for more clarity and information in the sentence in question. Figure 3(b) and figure S4 are now cited in line 317 of the revised manuscript to point the reader to relevant SPECIATE data to help compare with our findings of the MSDS surveys.

L315, but also more generally (L337, L342, L371 etc.): sometimes the term solvents is used, at other times consumer products, sometimes product/process-related VOCs. How these terms are used and where the overlap is or is not, needs to be defined. This would really help the clarity of the manuscript. In some cases (not necessarily how it is used here), solvents is an umbrella classification for consumer products, paints, etc. But it could also be a more limited definition.

Response: We agree with the reviewer that the mix of terms used by the field has been confusing and sometimes misleading. We use the products/processes as the general inclusive term for this diverse range of sources. However, historically, research and reporting has been focused narrowly on either just “solvents” or “consumer products”. At the lines in question, we purposefully report prior work and data with the classification they used for the sake of consistency and accuracy. We have tried to improve the wording in line 318 to prevent confusion.

L390: is there any indication of how much is missing. You cite how much is included, but can the missing amount be estimated at all from your analysis? Even if it is only for a couple of products or is just an order of magnitude range?

Response: In an attempt to constrain the potential IVOC content of current products, we provide this information in the ‘IVOC content’ column in tables 1 and 2 via different approaches, and discuss it later in the manuscript.

L509/Figure 5: as stated in the text and in the figure caption, the product emissions have SOA yields and ozone formation potentials on par with other major urban sources as a function of mass emitted. What about overall. It would be good to explicitly include this information based on your EI estimates here, also with a comment on whether you find the sources to be underestimated or not, as these are really some of the important implications of this work.

Response: We agree that this would also provide useful information. Yet, we present this figure as shown/described since it is conventional to report SOA yields as a function of mass of precursor species reacted, which is directly connected to mass emitted, and less to total mixture mass of product used/applied. The emissions inventories also only report mass emitted and not mass used/applied so it is not possible to back-calculate the requested information from the emissions inventory analysis.

As to whether or not these yields are underestimated relative to potential emissions, direct emissions and their potential SOA via pathways #1 and #2 (i.e. solvents + solute evaporation) is included, but emissions and potential SOA from degradation byproducts (pathway #3) are not since the larger precursors to degradation b-products have little- to no-volatility and were not eluted from the column. So in that the respect, these are conservative estimates. We acknowledge this pathway inclusion/exclusion in the paper.

The last two italicized sub-sections in section 4.2 seem a bit disjointed from the rest. With the first one on off-road combustion, I see what you are getting at with this, to just show the importance of off-road in the broader context, also for products/processes, but it is quite long and drawn out. I think this could be shortened significantly and the same points made (also because this really isn't the main aim of the paper at all). Following on from that the section on

modifying factors then seems pretty random and short. Could these points be integrated elsewhere? Possibly in the beginning of 4.2 as part of an intro/context for SOA/ozone formation?

Response: We understand the reviewer's comments, and have tried to shorten the section on off-road emission sources. However, we feel that these two subsections are key points of discussion as they play central roles in emissions and atmospheric chemistry in urban areas worldwide. Off-road mobile sources are important factors in urban air quality that have been mitigated at different timelines to on-road mobile sources. After reviewing and editing the section, we have decided to leave them in place since they are extremely important considerations that future researchers or policy-makers need to take into account when applying the methods or interpreting our findings in the context of specific urban areas outside the scope of our study.

L671/Inclusion of IVCOs... section: This section could be shortened a bit and made more concise.

Response: At the reviewer's request, we have revised and shortened this section in lines 689-713 of the (revised) manuscript.

Edits:

L144: evaporation of a solvent from a product or during a process – this is a bit awkward. From a product, is it meant here during storage? Or during use? Can this be clarified?

Response: Done. It is rephrased in line 143 of the revised manuscript for clarity.

L168: why is 'curing' in quotes and none of the others are?

Response: Edited. Quotes removed in line 168.

L196-197: 'other additional methods details' such as what? It would be good to mention what those are that are relevant to the text just outlined.

Response: Edited in line 197. Part of the line in question now reads "...and other additional methods details pertaining to potential SOA and ozone estimation can be found in the supporting information."

L230: ‘...the U.S. National Emissions Inventory and emission inventories from the Global Emissions Initiative (GEIA) were used for...’

Response: Thank you for pointing this out. A correction has been made in line 231 of the (revised) manuscript.

L236: write out EMFAC

Response: In lines 237-238, ‘EMFAC’ has been changed to ‘Emissions Factor (EMFAC)’.

L237: write out LEV

Response: In line 239, ‘LEV’ has been changed to ‘low-emission vehicles (LEV)’.

L275: ‘...such as those discussed in section 4.’ We are in section 4 and it has loads of subsections and examples, etc. so can you be more specific?

Response: Done. Revised in line 278.

L281: here, as well as some other cases (e.g., L450), please add a ‘from’ before listing a % or concentration range. E.g., ‘... from 3-100% ...’

Response: We thank the reviewer for mentioning this. A correction has been made in line 284 now.

L308: reference Fig 3b as well as S4 because they essentially show the same thing. The only thing that S4 adds is the uncertainty (?) or standard deviation (?) associated with the compound classes. Please specify in the figure caption for S4 what the error bars indicate.

Response: In line 311, figure 3(b) is now cited along with figure S4. The figure caption of figure S4 is edited to include that the error bars represent standard deviation in emissions from paints between the years 2000 and 2020.

L318: add ‘...in the U.S.’ at the end of the sentence unless this is inappropriate, in which case please specify the region.

Response: In line 322, ‘in the U.S.’ is added at the end of the sentence.

L326: ‘...where provided’ (currently just provide)

Response: Correction made in line 329.

L426: 18-29 Gg, of how many total Gg VOC emissions per year in CA? Is this a lot or not? Context would be good to add here (even if the info is available elsewhere).

Response: We have added more information in lines 436-439 to show the importance of pesticide emissions as a source of air pollution.

L442: larger asphalts? What is meant by this? Larger VOCs from asphalts that degrade to form smaller VOCs?

Response: In this sentence, we indeed point toward the non-solvent emissions that are caused by the degradation (i.e. fragmentation) of larger organic compounds from asphalts to form smaller compounds. The sentence has been edited in line 455 of the revised manuscript.

L459-460: please define 'cutback asphalt'

Response: Cutback asphalt is now defined in line 473 of the revised manuscript.

L461-463: how are these references also relevant for the EU inventories?

Response: We thank the reviewer for pointing this out. We have now added a reference relevant to the EU inventories in line 475 of the revised manuscript.

L516: is the additional 25% reduction relative to 1990, or relative to the 2015 amount?

Response: As stated in line 530, by 2020, a 25% reduction in potential SOA from on-road gasoline vehicles is expected relative to the year 2015's value of 3.3 tons day⁻¹.

L535: only 5% for which year?

Response: This is true for the year 2015. The sentence is now edited for clarity in line 550.

L605: 'their emissions' can you please be explicit and mention which ones? it is not clear here.

Response: In line 620, 'their emissions' is replaced with 'products/process-related emissions' for clarity.

L621: it would be good to give an e.g., and mention some of the other anthropogenic sources explicitly.

Response: We have revised lines 634-638 of the (revised) manuscript to clarify the conclusions of the cited work.

L636-637: This sentence is awkward and needs to be rewritten.

Response: Done. The sentence is now revised in lines 652-654.

L663: might be good to reiterate the three pathways from section 2

Response: Done. The sentence is rewritten in lines 679-681 to reiterate the three emission pathways.

March 1, 2018

We thank the reviewer for supporting our manuscript and their insightful comments which have helped us further improve our work. After having gone through the comments in detail, we present our revisions/replies below:

Reviewer 2:

Following on from the detailed reviewer comments added on the 15th of February, I have only a few things to add. I think this paper is a valuable contribution to the literature and nicely complements the paper by McDonald et al. recently published in Science (I was surprised there was no reference to this paper here).

-Line 124: Should furnishings be added to your list? There are emissions of VOCs from e.g. carpets that I'm not entirely clear are included in your list. Is it also worth somewhere making the distinction between primary and secondary emissions from surface sources, as these will vary over time – so primary emissions from furnishings/building materials are high in the first instance and then decrease over time, whilst secondary emissions are ozone dependent and may even increase over time if outdoor ozone concentrations (and hence indoor concentrations) increase.

Response: We agree with the reviewer that furnishings are a major potential source of emissions that belong as part of this work. We had broadly included these things with building materials but acknowledge that greater detail could be provided. To address this concern, we have modified the bullet points in lines 131-132 to “Building materials (e.g. carpeting, flooring, insulation, wood, gypsum)” and “Furnishings (e.g. furniture)”. Secondly we also agree that primary versus secondary emissions are an important differentiation. We include them both in our three emission pathways described in section 2, lines 144-149. To address the reviewer's concern, we have changed emission pathway #3 in lines 148-149 to also include emissions via degradation of “a solid product/material”. We also refer the author to lines 156-159 where we specifically discuss emissions via ozone oxidation, and have now added a comment about emission timescales for these emissions.

-Line 261: The products you investigated have ‘hidden’ ingredients (varying from 30-60% of the total). Is it similar for similar types of products, so you could potentially estimate the amount that is missing from your inventory, or is it manufacturer rather than product dependent or random?

Response: Our survey of MSDSs demonstrated very wide ranges of reported product composition for proprietary reasons. The reported ranges tended to vary at random, and could not be reliably constrained by product type.

-Line 498: There is some literature that suggests siloxanes may have health impacts - see review of such by Tran and Kannan in Science of the Total Environment 511 (2015) 138–144.

Response: We thank the reviewer for pointing out the Tran and Kannan (2015) paper. We acknowledge potential health effects from a wide range of compound classes that might not be fully understood. We intended “low” to include things that are not part of well-established hazardous compounds such as those on the air toxics list. We have now edited the sentence in lines 514-516 of the (revised) manuscript for clarity, and removed “siloxanes” to resolve the reviewer’s concern.

-Section 4.2: I agree with the first reviewer that this section is too long. I wasn’t clear from your response whether you had shortened this section significantly, but if not, would urge you to make it shorter and more focused.

Response: We have revisited the last 2 sub-sections of section 4.2 again, and based on the comments from the two reviewers, we have significantly shortened the “Contributions from off-road combustion-related sources” sub-section to a single, more concise paragraph. We have also incorporated the sub-section on modifying factors for SOA and ozone into earlier sub-sections (as suggested by Reviewer #1).

-Table 1: I was a bit confused by some of the footnotes. You need to tie them more specifically to the information in the table so we know which fraction the footnotes refer to (e.g. footnote f refers to the emitted fraction presumably?).

Response: We agree with the reviewer and have rephrased the “Note” at the bottom of Table 1 footnotes to prevent confusion. The “Note” now reads, “All fractions given in the footnotes refer to the emittable fraction of a product. Emittable fraction is confirmed with MSDS where possible.”

-The authors might also want to consider the increasing use of so-called ‘green’ materials indoors that typically have lower emission rates than more traditional materials. If such materials become more widely adopted, it may be that indoor sources of VOCs decrease over the considered period of time as well (or at least those from some of the sources considered).

Response: We agree with the reviewer that this is a very interesting area of development within the market and research field, but it is unclear if a holistic consideration of emissions across all three described pathways will yield significantly different emissions/conclusions, since modifications have primarily targeted more direct emissions historically. For example, a study

by Toftum et al. 2008 (cited in the manuscript in line 161) reports SOA formation from ozonation of a green paint. We agree that this is a very interesting area of research and have acknowledged it in the conclusions section with future research needs.

Edits

Line 322: Sentence too long and grammar needs improving.

Response: Done. The sentence is split into two sentence and rephrased for clarity.

Line 703: grammar needs improving.

Response: Done

1 **Considering the future of anthropogenic gas-phase organic compound** 2 **emissions and the increasing influence of non-combustion sources on urban air** 3 **quality**

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8 **Keywords:** Anthropogenic emissions, volatile and semivolatile organic compounds (VOCs,
9 SVOCs), ozone, secondary organic aerosol (SOA), urban air quality, consumer products,
10 building materials

12 **Abstract**

13 Decades of policy in developed regions has successfully reduced total anthropogenic emissions of
14 gas-phase organic compounds, especially volatile organic compounds (VOCs), with an intentional,
15 sustained focus on motor vehicles and other combustion-related sources. We examine potential
16 secondary organic aerosol (SOA) and ozone formation in our case study megacity (Los Angeles),
17 and demonstrate that non-combustion-related sources now contribute a major fraction of SOA and
18 ozone precursors. Thus, they warrant greater attention beyond indoor environments to resolve
19 large uncertainties in their emissions, oxidation chemistry, and outdoor air quality impacts in cities
20 worldwide. We constrain the magnitude and chemical composition of emissions via several
21 bottom-up approaches using: chemical analyses of products, emissions inventory assessments,

theoretical calculations of emission timescales, and a survey of consumer product material safety datasheets. We demonstrate that the chemical composition of emissions from consumer products, and commercial/industrial products, processes, and materials is diverse across and within product/material-types with a wide range of SOA and ozone formation potentials that rivals other prominent sources, such as motor vehicles. With emission timescales from minutes to years, emission rates and source profiles need to be included, updated, and/or validated in emissions inventories, with expected regional/national variability. In particular, intermediate-volatility and semivolatile organic compounds (IVOCs and SVOCs) are key precursors to SOA but are excluded or poorly represented in emissions inventories, and exempt from emissions targets. We present an expanded framework for classifying VOC, IVOC, and SVOC emissions from this diverse array of sources that emphasizes a lifecycle approach over longer timescales and three emission pathways that extend beyond the short-term evaporation of VOCs: (1) solvent evaporation, (2) solute off-gassing, and (3) volatilization of degradation by-products. Furthermore, we find that ambient SOA formed from these non-combustion-related emissions could be misattributed to fossil fuel combustion due to the isotopic signature of their petroleum-based feedstocks.

1. Introduction

Anthropogenic emissions of gas-phase organic compounds, including volatile organic compounds (VOCs), are of direct concern as toxic or carcinogenic air pollutants in indoor and outdoor environments (Cohen et al., 2005; Nazaroff and Weschler, 2004; Weschler and Nazaroff, 2008). Often they are more important for air quality as reactive precursors to the formation of outdoor tropospheric ozone and secondary organic aerosol (SOA) as well as indoor SOA, and thus play a central role in the elevated mortality and morbidity rates caused by fine mode particulate matter

(i.e. PM_{2.5}) and ozone in both developed and developing regions (Destailats et al., 2006; Jerrett et al., 2009; Lim et al., 2012; Nazaroff and Weschler, 2004; Pope and Dockery, 2006; Sarwar et al., 2004; Singer et al., 2006; Weschler, 2011). In urban and downwind areas globally, 20-70% of PM_{2.5} is organic aerosol (OA), with SOA comprising 58% of OA in urban areas and 82% downwind on average (Zhang et al., 2007).

Yields of SOA and ozone are strongly dependent on precursor molecular size, volatility, structure, and moieties/functionalities as well as environmental conditions (Gentner et al., 2012; Kroll and Seinfeld, 2008). SOA models have struggled to reproduce observations due to incomplete knowledge on SOA precursors and their sources (Hallquist et al., 2009; Kroll and Seinfeld, 2008). Gas-phase organic compound measurements and emissions data have historically focused on VOCs (i.e. C₂-C₁₂ alkanes and VOCs with equivalent volatilities), but research has demonstrated the prevalence and importance of larger, intermediate-volatility and semivolatile organic compounds (IVOCs and SVOCs, respectively) (Gentner et al., 2012; Kroll and Seinfeld, 2008; Robinson et al., 2007; Zhao et al., 2014). With greater SOA mass yields, IVOCs and SVOCs are key for modeling and mitigating SOA but are less-studied due to measurement difficulties (Gentner et al., 2012; Goldstein and Galbally, 2007). While their emissions from motor vehicles have received more attention, emissions from most other sources are poorly constrained.

In recent history, motor vehicles, power plants, and residential combustion have been dominant drivers of detrimental air quality (e.g., VOCs, nitrogen oxides (NO_x), PM_{2.5}, ozone, sulfur dioxide (SO₂)), with automobiles dominating anthropogenic VOC emissions in major cities (Gentner et

67 al., 2017; Hao et al., 2007; McDonald et al., 2013, 2015; Warneke et al., 2012; Zhou et al., 2003).
68 Yet due to the success of combustion-related emissions control policies over the past 60 years,
69 motor vehicle VOC (and other pollutant) emission factors have decreased by orders of magnitude
70 in route to today's diesel and gasoline vehicles, albeit with some recent on-road diesel NO_x
71 compliance issues (Hao et al., 2007; Kirchstetter et al., 1999; U.S. Environmental Protection
72 Agency, 2011; Warneke et al., 2012). In no other place has this been more studied than Los
73 Angeles, CA, where PM_{2.5} and ozone still exceed standards, and OA comprises 41% of PM₁, 66%
74 of which is SOA (Hayes et al., 2013; Warneke et al., 2012). Recent work has suggested that motor
75 vehicle emissions in L.A. cannot fully explain observations of reactive precursors and SOA
76 (Ensberg et al., 2014; Hayes et al., 2015; McDonald et al., 2015; Zhao et al., 2014). In L.A. and
77 beyond, the role of motor vehicles in degrading air quality will decline further with the newest on-
78 road emissions standards (e.g. Tier 3/LEV-III, Euro 6/VI) (Figure 1), use of electric-powered
79 vehicles, and stricter regulations on emissions from off-road vehicles and other engines (Gentner
80 et al., 2017; Giannouli et al., 2011; Gordon et al., 2013; Platt et al., 2014; Tessum et al., 2014;
81 Weiss et al., 2011).

82
83 The main objective of this paper is to evaluate anthropogenic sources of gas-phase organic
84 compounds that are gaining prominence as motor vehicles and other combustion-related sources
85 become cleaner in the developed and developing world. We demonstrate that consumer products
86 and commercial/industrial products, processes, and materials are important, widely-distributed
87 area sources of VOCs, IVOCs, and SVOCs with variable source profiles making their emissions
88 and impacts difficult to observe and constrain. Our demonstration builds on relevant indoor air
89 quality studies observing emissions and substantial SOA formation from ozone-initiated reactions

with emissions from consumer products, building materials, and cleaning products (Chang et al., 2011; Destailats et al., 2006; Gold et al., 1978; Lewis, 2001; Mitro et al., 2016; Nazaroff and Weschler, 2004; Singer et al., 2006; Weschler, 2011; Weschler and Nazaroff, 2008; Wilke et al., 2004) as well as secondary emissions from oxidative degeneration of indoor paints, varnishes, and materials (Knudsen et al., 1999; Poppendieck et al., 2007a, 2007b; Salthammer and Fuhrmann, 2007).

We outline a holistic framework to assist future research in the field, and then use a multi-dataset approach to constrain emissions, which includes the most detailed emissions inventory available, laboratory analysis of product composition, a survey of consumer product material safety datasheets (MSDSs), and calculations of emissions timescales from theory. Specifically, we (a) evaluate the pathways, chemical composition, and magnitude of VOC, IVOC, and SVOC emissions from prominent types of products, processes, and materials; (b) calculate their potential to form SOA and ozone; and (c) compare their emissions and potential air quality impacts to other prominent sources. We identify major knowledge gaps in emission rates and composition, including I/SVOCs, which we explore via two examples: pesticides and asphalt-related emissions.

2. Defining a comprehensive framework for non-combustion-related emissions

There is a wide body of work on some aspects of emissions from products, processes, and materials in indoor or outdoor environments, but outdoor-focused emissions inventories are mostly focused on evaporative emissions of VOC solvents at the time of application. Here we outline the current

111 scope of emissions and expand on it to define a comprehensive framework to guide future studies
112 on this class of emissions.

113
114 **Volatility:** Emissions of gas-phase organic compounds can and should be differentiated by their
115 volatility: VOCs, IVOCs, and SVOCs, or further by their n-alkane-equivalent volatility (Murphy
116 et al., 2014). One can broadly refer to compounds emitted from products and processes ~~via the~~
117 ~~prefix ‘pp’ (e.g. pp VOCs), which is consistent with existing conventions for volatility basis set~~
118 ~~and aerosol abbreviations (e.g. pp SOA) as products/process-related emissions.~~

119
120 **Source categories:** While the spectrum of sources is very diverse, they are often grouped by use.
121 The following source categories are consistent with the most detailed VOC inventories for
122 consumer products, but are extended (shown with * below) to include incorporate a broader array
123 of materials/ and products, including those used in commercial and industrial processes ~~(shown~~
124 ~~with * below):~~

- 125 • Paints (indoor and outdoor),
- 126 • Industrial adhesives,
- 127 • Consumer adhesives,
- 128 • Sealants,
- 129 • Pesticides (consumer and agricultural),
- 130 • General cleaning products,
- 131 • Targeted cleaners,
- 132 • Personal products (e.g. beauty and hygiene),

- Building materials; (e.g. carpeting, flooring, insulation, wood, gypsum).*
- Furnishings (e.g. furniture).*
- Paving and roofing asphalt-related materials,
- Solid consumer products and packaging (e.g. plastics, elastomers), and their additives (e.g. plasticizers, flame retardants).*

However, the volatility range of most of the existing categories needs to be expanded to completely include I/SVOCs. We discuss I/SVOC-containing pesticides and asphalt-related products/processes as examples that are not sufficiently included in inventories (see Section 4.1.3). Notes: we exclude gasoline evaporation (from vehicles or fuel stations) and fossil fuel extraction/processing given their close connection to combustion. We briefly discuss food-derived cooking emissions for comparison, which are important and classified separately.

Emission pathways: We propose three general pathways of product/process-related emissions:

- 1) Evaporation of solvent from a product while it is in storage or being used, or during a process;
- 2) Volatilization of solute or active compounds from an applied coating or a solid product/material (i.e. off-gassing); and
- 3) Volatilization of by-products from the degradation or transformation of solute or active compounds; or a solid product/material.

The first is the principal pathway previously considered and often occurs over faster timescales (minutes-days) from liquids like cleaners, paints, and other solvents. The second acts over longer timescales (weeks-years), is not always included in inventories, and is a key potential source of

I/SVOCs (see Section 4.1.3). SVOC pesticides, flame retardants, and off-gassing plastics are examples studied for indoor air quality (Batterman et al., 2009; Brodzik et al., 2014; Clausen et al., 2004; Faber et al., 2013; Kemmlein et al., 2003; Lewis, 2001; Wensing et al., 2005; Weschler and Nazaroff, 2008). The third is due to the generation of degradation by-products via thermal or photochemical stress, exposure to oxidants (i.e. OH, O₃, or NO₃), or other reactive chemical processes. These may have longer emission timescales depending on oxidant concentrations and the chemical and physical properties of degradation precursors. These emissions are largely understudied with the exception of asphalt-related emissions and ozonation of indoor materials (Poppendieck et al., 2007a, 2007b; Salthammer and Fuhrmann, 2007; Toftum et al., 2008).

A lifecycle approach: Emissions during application or immediate use are most commonly studied, but many emissions related to products, process, and materials occur over much longer timescales. In the future, emissions studies need to include all three potential emissions pathways across full lifecycles of:

- Storage,
- Transport,
- Application,
- ~~Curing~~²²₁₂,
- Active use,
- Weathering,
- Restoration,
- Removal and disposal.

177 There are too many facets within the lifecycles of each source category to discuss in this paper,
178 but our case study on asphalt-related emissions in Section 4.1.3 demonstrates several aspects.
179 Production methods are not explicitly included here since they may fall under the purview of
180 industrial point sources, but with small, distributed production it may be appropriate to consider
181 the industrial/commercial processes as area-wide sources. Chemical transformations of materials
182 or products across their lifetimes need to be considered since they can influence the chemical
183 composition and rates of emissions. For example, transformations can occur during a wide variety
184 of in-use conditions or during the storage of unused or partially-used products/materials, both of
185 which can be over long storage periods in a variety of environmental conditions. Application
186 methodology is a determining factor in both emission rates and composition during application.
187 For example, common methods include direct liquid application or aerosolization, either via a
188 pressurized can or a nozzle and air compressor, and aerosolization provides a direct emission
189 pathway for all components and subsequent evaporation of aerosol droplets. Finally, emissions in
190 latter parts of lifecycles (i.e. weathering/deterioration, restoration, removal, and disposal) all need
191 to be considered.

192

193 **3. Methods**

194 We constrain the magnitude, chemical composition, and potential air quality impacts of
195 emissions using multiple bottom-up approaches: chemical characterization of a selection of
196 consumer products via gas chromatography-mass spectrometry and carbon isotope mass
197 spectrometry; a detailed assessment of emissions inventories and estimation of source
198 contributions to potential SOA and ozone formation; theoretical calculations of emission
199 timescales; and a survey of reported chemical composition in consumer product MSDSs. Details

on the emission timescale calculations can be found in Appendix B, and other additional methods details [pertaining to potential SOA and ozone estimation](#) can be found in the supporting information.

Chemical speciation of consumer products: We selected 17 consumer products across a range of product types with attention to those with unresolved alkane or aromatic mixtures (Table 1). Top-selling products from major commercial providers were selected in order to make a realistic assessment of products that are currently in significant public use, and to avoid biasing the analysis towards less-common products. With a limited sample size, only 1 water- and 1 ethanol-based product were included. Diluted samples were prepared at a concentration of 1000 ng μL^{-1} in either hexane or toluene depending upon the product composition provided in their MSDS. The chemical composition of the emittable fraction of products was determined using gas chromatography with electron ionization-mass spectrometry (GC-EI-MS) (Agilent 7890B/5977). A 1 μL syringe (Hamilton) was used to directly inject the sample onto the GC column through an inlet held at 320 °C. During each injection, the column was initially held at 40 °C for 2 minutes, then ramped at 10 °C min^{-1} to 325 °C and lastly held at 325 °C for 5 minutes. Mass spectra of background subtracted individual ion peaks were used to identify compounds in a sample using the NIST mass spectra library. Calibration curves were drawn for five different concentrations of authentic standards (AccuStandard) for diesel range alkanes ($\text{C}_{10}\text{-C}_{28}$), purgeable aromatics, and terpenes. The emittable organic fraction of raw products is defined by volatility and approximated as VOCs + IVOCs via our chemical analysis and reported MSDS data (Note: SVOCs excluded in this analysis to focus on compounds that are fully partitioned to the gas-phase at equilibrium under typical conditions). A carbon isotopic analysis was also carried out at the Keck Carbon Cycle Accelerator

223 Mass Spectrometer (AMS) facility at U.C. Irvine to measure carbon-14 ($\text{‰ } \Delta^{14}\text{C}$) and carbon-13
224 ($\text{‰ } \text{d}^{13}\text{C}$) relative to carbon-12 via a 0.5MV Compact AMS (National Electrostatics Corp.), and
225 estimate the fossil carbon content in individual products.

226
227 ***Urban emissions inventories:*** We used the California Air Resources Board (CARB) Almanac
228 emissions inventory and the U.S. EPA SPECIATE 4.4 source profiles to generate the total and
229 compound-specific emissions for ~~our~~ California and Los Angeles (Cox et al., 2013; U.S.
230 Environmental Protection Agency, 2014). The CARB emissions inventory is the most detailed
231 available with respect to source categories and basin-level rates. Los Angeles is our case study
232 megacity given its historical role in air quality research and policy with a multi-decadal record of
233 emissions data, ambient measurements, field campaigns, and publications (Bishop and Stedman,
234 2008; Fortin et al., 2005; Neligan, 1962; Warneke et al., 2012). In addition, the U.S. National
235 Emissions Inventory and emission inventories from the Global Emissions Initiative (GEIA)
236 ~~inventories~~ were used for nationwide and worldwide comparison of VOC emissions respectively
237 from solvents and on-road motor vehicles.

238
239 We estimated total daily potential ozone and SOA formation in greater Los Angeles from the
240 product/process-related emissions included in the CARB inventory. Values were also calculated
241 for exhaust and evaporative emissions from on-road motor vehicles using CARB's Emission
242 Factors (EMFAC) model database and literature ozone and SOA yields for each source pathway
243 and low-emission vehicles (LEV) generation (Gentner et al., 2013, 2017; Zhao et al., 2017).
244 Potential ozone formation values are based on maximum ozone incremental reactivity (MOIR)

values from the SAPRC-07 inventory (Carter, 2007; Gentner et al., 2013). The very diverse range of compound classes used in products, materials, and processes remain largely understudied with respect to their SOA yields (e.g. esters, siloxanes). Hence, [specific](#) literature SOA yields were used wherever possible (Algrim and Ziemann, 2016; Chacon-Madrid et al., 2010; Chan et al., 2010; Gentner et al., 2012; Kwok and Atkinson, 1995; Ng et al., 2006; Pankow and Asher, 2008; Sadezky et al., 2006; Tsimpidi et al., 2010), and estimated for other unstudied compounds ~~(without existing~~ [SOA yields \(see Table S6 for details\)](#). SOA yields are estimated at 10 $\mu\text{g OA m}^{-3}$ in urban, “high- NO_x ” conditions (approx. >5 ppb). Ozone and SOA yields for each analyzed consumer product and product categories are compared to other key sources. The SOA yields provide a conservative, lower estimate of potential SOA without aqueous SOA despite studies showing that aqueous pathways to SOA increase SOA yields for small oxidized precursors or their oxidation by-products (Daumit et al., 2016; Jia and Xu, 2014). For the case study city Los Angeles (and Mexico City), aqueous SOA formation was relatively small during major field studies (Dzepina et al., 2009; Hayes et al., 2015; Washenfelter et al., 2011). However, future work in other cities should consider aqueous SOA given the magnitude of oxygenated fraction in product/process-related emissions (see section 4.1.2).

Survey of material safety datasheets (MSDS): We obtained the MSDS data by surveying a set of 88 MSDS entries from the websites of major home improvement stores focusing on their top-selling products. Five product categories were chosen including paints, adhesives, cleaning products, sealants and pesticides. Chemical composition information was extracted from the “composition/information on ingredients” section reported in the datasheets. MSDS entries for commercial products frequently report 30% to 60% of product composition as “proprietary

mixtures”, so this survey only identifies the presence and establishes general ranges for current product types (Table 2).

4. Results and Discussion

4.1 Composition and magnitude of product/process-related emissions

The organic composition of consumer products and commercial/industrial products, materials, and processes are very diverse, which leads to similar diversity in emissions, and further region/nation-specific heterogeneity can be expected. We calculate the magnitude and average chemical composition of emissions from source categories included in the CARB [emissions](#) inventory [and SPECIATE database](#) (Figure 2a) with the goal of assessing the distribution of emissions across organic compound classes and product types (i.e. source profiles) in a megacity with the most representative inventory available. We find that the consumer products and commercial/industrial processes that comprise product/process-related sources are large emitters of a diverse suite of VOCs, but view these results as a lower estimate given likely missing emissions, such as those discussed in [Section 2](#) and [4.1.3](#).

4.1.1 Chemical composition

Laboratory analysis of consumer products: Our results summarized in Table 1 demonstrate the prevalence of non-benzene, single-ring aromatics and C₆-C₁₂ alkane mixtures as solvents, and the presence of IVOCs and SVOCs in consumer products. The emittable fraction of products ranged [from](#) 3-100%, and the single-ring aromatic and IVOC content ranged [from](#) 3.5-93% and 0.77-95%,

respectively. Another key result of this analysis is the frequency of many functionalized aliphatic or aromatic VOCs and IVOCs that are not traditionally measured in atmospheric monitoring (e.g. esters, acetates, siloxanes), which is echoed in the MSDS survey results. More detailed speciation results with a breakdown of alkanes and single-ring aromatic compounds can be found in Table S1. Validation of real-world product/process-related emission rates/timescales is necessary to advance the field but will require examining a wide range of products and geographic conditions.

Analysis of emissions inventories: Figure 3(a) shows the estimated overall composition of product/process-related emissions in Los Angeles averaged over the years 2005 to 2020 determined by combining SPECIATE source profiles with the CARB emissions inventory for the South Coast Air Basin (SoCAB). Alcohols and miscellaneous emissions together make up 50% of the total product/process-related emissions with ethanol as one of the largest individual species emitted, which is consistent with SoCAB ethanol observations that were substantially greater than what would be expected with gasoline vehicles as the only source (Gouw et al., 2012). 70% of the miscellaneous emissions are made up of mineral spirits whose composition varies greatly with application but are generally comprised of acyclic and cyclic C₇₋₁₂ alkanes with variable amounts of aromatic content. The remaining 30% includes unresolved asphalt mixtures, oxygenates, fragrances, and undefined petroleum distillates/oils/spirits, some of which also fall into the I/SVOC category discussed below. Single-ring aromatics are estimated at 13% of total emissions with a mix of 43% toluene, 38% C₈, 3% C₉, and 1.5% C₁₀ aromatics, and minor PAH emissions. Carbonyls represent 10% of total emissions, 36% of which is acetone. Anthropogenic terpenoid emissions in the inventory, while highly reactive, are small relative to other sources and biogenic contributions (see section S.4 in SI).

312

313 Figure 3(b) shows the distribution of organics in the SoCAB emissions inventory from major
314 product/process-related source categories. Paints emerged as the highest VOC emitter, ~21% of
315 which is single-ring aromatics (Figure 3(b) and S4). Targeted cleaners, pesticides, and
316 general cleaners were the next largest source types in the inventory. A more detailed breakdown
317 of emissions broken up by source category and compound class can be found in Table S3. We used
318 the EPA SPECIATE database for source profiles in this study, which had a national scope, some
319 inclusion of I/SVOCs, and greater chemical detail (i.e. number of species). We examined changes
320 in the chemical speciation of total emissions with CARB's 2010 adjustment to SPECIATE in the
321 final years of our study period. We found minor differences in the overall emissions (Figure S5)
322 that are largely attributable to changes in the assignment of unspciated (i.e. miscellaneous) VOC
323 and I/SVOC emissions that do not affect our conclusions. Differences in potential SOA and ozone
324 in the SoCAB for 2010-2020 (Section 4.2) are not clear due to uncertainties in these changes to
325 unspciated assignments, and the uncertainties in SOA yields.

326

327 **MSDS survey:** Summarized in Table 2, the greatest single-ring aromatic content was observed in
328 adhesives (8%) and sealants (9%), while the 30 surveyed paints did not have aromatic VOC content
329 in contrast to the SPECIATE source profiles (Figures 3(b) and S4). In comparison, the chemical
330 speciation of 42 major solvents categorized as 'solvents' (across C₆₋₁₃) in 2002 reports a
331 wide range of total aromatic content (0-100%) and an average of 41% (±46%) with the remainder
332 comprised of acyclic and cyclic alkanes (Censullo et al., 2002). Differences when compared to our
333 present-day laboratory and MSDS paint speciation show reductions in aromatic content due to
334 increasingly stringent regulations in the U.S. However, for other locations, it highlights the likely,

Commented [DG1]: This section also addresses reviewer #1's question from an above section with additional discussion that extends beyond the methods section and intro paragraph to this main section "L269-270: Please add some more information to clarify how the composition is calculated. The source categories are based on the CARB inventory, but where is the speciation coming from? Or are you explaining what you will be doing with the speciate database and then MSDSs etc that is explored in the following sections. This isn't clear. "

continued prevalence of single-ring aromatics in solvents, especially for developing regions. We also found that MSDSs did not report I/SVOCs content in the composition of pesticides ~~while our laboratory speciation found a~~. However, we observed 8%-95% IVOC content in the emittable fraction of the ~~analyzed~~ pesticides samples analyzed in our laboratory. For all of the product categories examined, and especially for pesticides, the amount of compositional information provided by the MSDSs is limited and hinders our ability to constrain the average aromatic or I/SVOC content since compounds outside the VOC range are often not disclosed due to proprietary claims or regulatory exemptions, and where ~~provide~~provided all compound concentrations are usually provided as a wide range. For example, under “fragrance exemptions” VOCs and IVOCs are often not disclosed and can even be labeled as “VOC-free”.

4.1.2 Emission rates

While motor vehicles are still major sources in developed regions, their total gas-phase organic compound emissions have been gradually declining with the continued implementation of stricter emissions standards (Gentner et al., 2017), increasing the relative importance of other sources (Figures 1, 2a). At a global scale, the MACCity and ACCMIP emissions inventories estimate global solvent-related VOC emissions of 15 Tg yr⁻¹ in 2000 and year-over-year increases since 1960 (speciated by aromatics, C₆₊ alkanes, ketones, alcohols, and other VOCs), and emissions of aromatics from solvents are expected to outweigh those from transportation in 2020 (7.5 vs. 6.7 Tg yr⁻¹) (GEIA, 2017). The 2014 U.S. National Emissions Inventory (NEI) data reports that VOC emissions from solvent-related sources are just 25% less (300 tons day⁻¹) than those from on-road mobile sources nationally, while they exceed on-road emissions by 25% (21 tons day⁻¹) in California (U.S. Environmental Protection Agency, 2011). Figure S1 shows California’s statewide

358 bi-decadal emissions estimates from CARB where product/process-related VOCs will reach 450
359 tons day⁻¹ in 2020, exceeding motor vehicles by 116 tons day⁻¹. A similar picture may emerge in
360 developing nations, as the control of motor vehicle emissions is greatly accelerated by the
361 advancements and knowledge of developed nations; such that emission standards in major
362 emerging economies employ either U.S. or E.U. policies, and are generally only one generation
363 behind (Kodjak, 2015). International studies over the past 2 decades show highly varying
364 contributions (~5-45%) to the total anthropogenic VOC emissions from just “solvent” use at both
365 regional and national scales (summarized in SI) (van den Born et al., 1991; Caserini et al., 2004;
366 Chen et al., 2009; Deguillaume et al., 2008; Lu et al., 2007; Markakis et al., 2009; Menut, 2003;
367 Nielsen et al., 2008; Piccot et al., 1992; Song et al., 2007; U.S. Environmental Protection Agency
368 and Office of Air Quality Planning and Standards, 1991).

369
370 Emissions inventory data for Los Angeles demonstrate that anthropogenic emissions have
371 consistently decreased over the last four decades (Figure 2a), which is consistent with ambient
372 observations (1960-2010) (Warneke et al., 2012). As the contribution of on-road motor vehicles
373 to total anthropogenic emissions has declined, product/process-related sources have become a
374 major contributor of VOCs. Based on the CARB emissions inventory, contributions of
375 product/process-related sources are the largest single contributor of VOC emissions in the basin
376 and state (Figures 2a & S1), and are growing with population (i.e. increased in-basin usage). While
377 consumption volume is low compared to combustion fuels, emission factors are higher given that
378 most of the volatile components are emitted whereas fuels are burnt at $\geq 99\%$ efficiency. Yet, no
379 region can be fully representative of product/process-related emissions on a larger scale, and
380 regional/national specifics will influence the magnitude and composition of emissions. For

381 example, California's extensive regulations will modify the composition of products sold in CA,
382 and likely elsewhere in the U.S.

383

384 **4.1.3 Emissions of intermediate- and semi-volatile organic compounds (IVOCs and SVOCs)**

385 Despite representing only a small to moderate amount of emissions, IVOCs and SVOCs from
386 motor vehicles are key precursors to urban SOA (Gentner et al., 2012; Robinson et al., 2007).
387 Similarly, we conclude that consumer products and commercial/industrial processes are also large
388 sources of unspciated IVOCs and SVOCs, some of which are included in the CARB inventory,
389 while there is evidence that some other source pathways are not.

390

391 Many common products/materials (e.g. pesticides, fragrances, foams, plastics) have IVOCs or
392 SVOCs that partition to reach equilibrium and evaporate over long timescales (Figure 4)
393 (Batterman et al., 2009; Clausen et al., 2004; Mitro et al., 2016; Weschler and Nazaroff, 2010).
394 Our chemical analysis revealed IVOCs or SVOCs in 10 of the 17 products (Tables 1 & S1).
395 Including the MSDS survey, we found composition ranging from 0% to 95% I/SVOCs, with
396 I/SVOCs present in 23% of cleaners, 20% of adhesives, 24% of paints, and 46% of sealants (Table
397 2). Aliphatic or aromatic I/SVOCs are frequently used in some types of pesticides and are
398 sometimes replaced with biogenic oils (e.g. neem oil, fish oil), such that the bulk of the pesticides
399 in our MSDS survey were comprised of < 1 - 10% active compounds and a balance of undisclosed,
400 "non-hazardous" ingredients.

401

402 The SPECIATE profiles and CARB emissions inventory include some estimates of unspciated
403 “low-vapor pressure VOCs (LVP-VOCs)” in consumer products that are defined as larger than 12
404 carbon atoms (or equivalent volatility), which is roughly consistent with the beginning of the IVOC
405 range (California Air Resources Board, 2015b). While poorly constrained, non-aromatic I/SVOCs
406 included in the inventory constitute ~3% (6 tons day⁻¹) of total SoCAB emissions estimates in the
407 CARB/SPECIATE case study, 90% of which is classified as unspciated “LVP-VOCs”. Consumer
408 product pesticides, general purpose cleaners, and targeted cleaners (e.g. laundry) are the largest
409 sources of I/SVOC contributions emitting 3.6, 1.0 and 0.4 tons day⁻¹ of I/SVOCs, respectively.
410 Consumer product pesticide emissions had the highest fraction of I/SVOCs (29%).

411

412 While CARB’s consumer products inventory includes I/SVOCs due to manufacturer reporting
413 requirements, I/SVOCs are exempt from limits on VOC content except for multipurpose solvents
414 and paint thinners, and there are known limitations in their coverage, especially of oxygenated
415 species (California Air Resources Board, 2000, 2015b). We conclude there are other anthropogenic
416 sources of IVOCs and SVOCs that have not been considered due to their long emission timescales
417 (i.e. days-years; Figure 4, Table S9) (de Gouw et al., 2011; Weschler and Nazaroff, 2008). We
418 present evidence for two such examples in this paper: I/SVOC-containing pesticides and asphalt-
419 related products, materials, and processes, but other examples include the volatilization of I/SVOC
420 solvents, solutes, or solids (e.g. coatings, flame retardants) and materials that may degrade to form
421 compounds with volatilities of C₁₃₋₂₆ n-alkanes (e.g. construction materials/coatings, including
422 materials with petroleum distillates/residues, mineral oil, coal tar, or similar).

423

Example 1: Emissions of IVOCs and SVOCs from pesticides: We have several pieces of evidence that demonstrate pesticides (including herbicides, insecticides, and fungicides) contain I/SVOCs, but they also highlight the fact they are poorly documented and regulated. We analyzed the chemical composition of three pesticides available as consumer products and they were comprised of 8%, 20% and 95%, IVOCs in the C₁₄₋₁₇ range (Table 1, Figure S3), with trace levels of larger compounds. Our MSDS survey (Table 2) was inconclusive for pesticides since the majority of consumer pesticides are not disclosed in MSDSs due to claims of proprietary mixtures, non-regulated components outside of the VOC range, and/or because they use naturally-derived oils (e.g. neem oil). Studies have shown such naturally-derived oils are comprised of aliphatic and aromatic I/SVOC-range compounds (Isman, 2000; Kumar and Parmar, 19996). Our emissions inventory analysis shows that non-aromatic I/SVOCs are the largest inventoried source of I/SVOCs (Figure 3), but the lack of data in MSDSs and VOC exemptions from regulations suggest this is a lower estimate of actual emissions. To examine commercial/industrial products, we also analyzed the composition of three petroleum-based pesticides used in agriculture (Figure S2), which are comprised of C₁₆-C₂₆ cyclic and acyclic alkanes mostly in the SVOC range that are applied as thin films via a sprayed water emulsion. These can partition to the gas phase and be re-emitted on the timescales in Figure 4 and may impact urban (or downwind) areas in agricultural regions, with 18-29 Gg applied yr⁻¹ in California. California-wide emissions from applied agricultural pesticides were nearly one-fifth of that from consumer products in 2015. However, the high I/SVOCs fraction in their composition and potential to emit over long timescales makes them an important consideration for air quality.

Example 2: Emissions of IVOCs and SVOCs from asphalt-related products, materials, and

processes: We propose asphalt-related products/processes as important sources of IVOCs and SVOCs whose emissions are currently underestimated in inventories and require better quantification. Asphalt-containing materials are used in road paving and repair (and similar applications for roofing or other surfaces), and are comprised of petroleum-derived organic compounds; predominantly non-distillable (i.e. non-volatile) asphalts sometimes with smaller amounts of VOCs, IVOCs, and/or SVOCs. They are used as sealers, coatings, and binders; mixed with aggregates to pave roads; and applied using either high application temperatures, water emulsions, and/or solvent.

The three paving/roofing-related products we analyzed contained aliphatic and aromatic VOCs and IVOCs up to C₁₈ present as solvents, with minor SVOC content (Figure S3). Similarly, there were no I/SVOCs declared in the asphalt-containing products in our MSDS survey. Non-solvent emissions during the hot storage, application, or resurfacing of these asphalts are caused by the degradation (i.e. fragmentation) of larger organic compounds from asphalts to form smaller compounds (C₇-C₃₀), which include cyclic and acyclic alkanes, single-ring aromatics, PAHs (2-, 3-, and 4-ring), and sulfur- or nitrogen-containing species (i.e. benzo- and dibenzo- thiophenes and furans), all of which were not present in the asphalt prior to heating (Cavallari et al., 2012b; Gasthauer et al., 2008; Kitto et al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007; The Asphalt Institute & European Bitumen Association, 2015). The total mass and composition of emissions is dependent on production methods, asphalt grade, and increases in magnitude and maximum molecular weight with storage/application temperatures (i.e. more SVOCs at high temperatures), which ranged 100-240 °C, or higher for roofing asphalts (Cavallari

et al., 2012c; Gasthauer et al., 2008; Kitto et al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007; The Asphalt Institute & European Bitumen Association, 2015).

Emissions of aromatic and aliphatic VOCs, IVOCs, and SVOCs from heated asphalt mixtures (“hot mix”) during application have also been documented in occupational health studies on “asphalt fumes” (Cavallari et al., 2012a, 2012b; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007). Yet, current emissions inventories do not include emissions of VOCs, IVOCs, and SVOCs from the degradation of larger compounds during and after the application of asphalt mixes. Estimation methods focus solely on the evaporation of VOC solvents from “cutback asphalt”, which is a mixture of asphalt and solvent, included as an area source in the “solvent evaporation” category in Californian, U.S., and E.U. inventories (California Air Resources Board and Sonoma Technology Inc., 2003; European Environment Agency, 2017; San Joaquin Valley Air Pollution Control District, 2008; U.S. Environmental Protection Agency, 2014; U.S. EPA, 1995). Road paving solvents are prohibited in non-attainment areas in California (Table S4), so emissions in the SoCAB case study are minor (1 ton day⁻¹) and mostly smaller than C₁₀ in the SPECIATE source profiles (Table S5) (Cox et al., 2013).

Emission factors of degradation byproducts don’t exist, so we approximate lower limits on emission factors only for the period immediately during application using limited published data (see section S.3 in SI), but longer timescale experiments are necessary. Calculated lower limits range from 100-2000 mg kg⁻¹ of asphalt (not including aggregate) with a strong dependence on application/storage temperature. This is on the same order as motor vehicle emission factors and

491 is greater than CARB's current emission factor for hot-mix asphalt (District, 2012; Gentner et al.,
492 2017; San Joaquin Valley Air Pollution Control District, 2008). Yet, California's asphalt
493 consumption of 1,540,000 tons liquid asphalt year⁻¹ (Table S4) represents statewide I/SVOC
494 (+VOC) emissions of 0.5 – 8 tons day⁻¹ during application alone (The Asphalt Institute, 2015).
495 This does not overwhelm current solvent-VOC emissions from paving/roofing (33 tons day⁻¹) but
496 emphasizes the need for further research since the poorly-constrained emissions largely include
497 I/SVOCs emitted over long timescales which are known to have high SOA yields.

498
499 Asphalt-related emissions exemplify the stated need for lifecycle-focused approaches, with
500 potential emissions across storage, transport, application, curing, weathering (e.g. degradation due
501 to climate or UV radiation), and resurfacing. Their magnitude and composition will vary with
502 production/handling methods, geologic source, and application type and location (esp. climate).
503 Emission pathways (from Section 2) include (1) volatilization of application solvents and (3) the
504 production and release of degradation byproducts while (2) does not apply due to negligible off-
505 gassing from extremely low volatile un-degraded asphalt constituents. The emission of asphalt
506 degradation products may peak during construction-related activities primarily due to asphalt's
507 exposure to high temperatures during its storage, application, or resurfacing. Still, seasonal highs
508 in surface temperature (summer pavement maximums are 47-67 °C and up to 70 °C for roofs
509 (Parker et al., 1997; Pomerantz et al., 2000)), will likely affect the rate of internal transport and
510 diffusion out of the "cured" asphalt layer resulting in emissions extended over its lifetime. (Note:
511 paving solvents are currently assumed to be emitted over several months (California Air Resources
512 Board and Sonoma Technology Inc., 2003)).

513

4.2 Potential SOA and ozone formation of product/process-related emissions compared to other major sources

Products and processes emit a diverse array of organic compounds (Figure 3). Some are of relatively-low direct concern for human health (e.g. terpenoids, ~~siloxanes etc.~~), while others present issues as primary emissions, especially in indoor or concentrated workplace environments (e.g. aromatics, ethers, PAHs). Yet, most are reactive and will oxidize in outdoor or indoor environments to form oxidized VOCs with unknown, but large potential health effects (Pöschl and Shiraiwa, 2015). This section focuses on their impacts on air quality via SOA or ozone formation. However, the health or environmental effects of the primary emissions should be especially considered in developing regions where primary VOC emissions are larger, or for specific compound classes near sensitive natural environments. A comparison of the SOA yields and ozone formation potentials for major source categories (Figure 5) demonstrates that product/process-related emissions have SOA yields and ozone formation potentials that are on par with other major urban sources such as motor vehicles and are strongly dependent on composition. ~~Gas-phase cooking emissions from food represent an additional uncertain source in urban air quality along with cooking POA which has been more studied.~~

SOA formation potential: The potential SOA from on-road gasoline vehicles in greater Los Angeles region has decreased by ~65% between the years 1990 and 2015. By 2020, further reduction by 25% is expected relative to the 2015 value of 3.3 tons day⁻¹. These numbers for diesel vehicles are 75% and 1.3% ~~tons day⁻¹~~, respectively. If the emissions inventory is accurate, then the 2015 potential ~~ppproducts/process-related~~-SOA in the SoCAB basin is nearly equal to the SOA formation potential of on-road gasoline and diesel vehicles, and is estimated to surpass them by

2020 with an increasing share of SoCAB's total anthropogenic emissions (Figure 2b). While single-ring aromatics and PAHs constitute only 11% of the total product/process-related emissions, they are responsible for ~80% of the potential SOA from those sources in the region (4% PAHs and 76% single-ring aromatics, largely toluene and xylenes). Existing emission inventories badly underestimate emissions of I/SVOCs and their contributions to SOA. They are shown to be responsible for 0.18% of ~~pp~~products/process-related-SOA, but this excludes 87% of the total I/SVOCs emissions which are labeled as unspecified "LVP-VOCs" in the inventory and thus have no assigned SOA yield in calculations. Additionally, our calculations do not include "missing" emissions or their potential SOA. Anthropogenic terpenoids, including lemon oil, pine oil, orange oil, orange terpenes, D-limonene and α -pinene, are responsible for ~8% of the total product/process-related SOA.

It is important for future work to consider modifying factors for SOA (and ozone) formation chemistry. Relative VOC to NO_x ratios have been shown to affect the chemistry and production rates of SOA and ozone (Hallquist et al., 2009; Sillman, 1999; Zhao et al., 2017). For many cities outside Los Angeles, urban air quality develops within a backdrop of biogenic emissions of VOCs and IVOCs, which is critical to keep in mind as we pursue stricter emission targets for reactive organics. In some cases, such as in the Southeast U.S., biogenic emissions dominate over anthropogenic gas-phase organics, and emissions of SO₂ and NO_x are large drivers of biogenic SOA formation (Xu et al., 2015). Similarly, gas-phase cooking emissions from food represent an additional uncertain source in urban air quality along with cooking POA which has been more studied (Bruns et al., 2017; Hayes et al., 2013; Klein et al., 2016).

Ozone formation potential: Over the past several decades, potential ozone has been dominated by emissions from gasoline motor vehicles. Yet, in 2015 potential ozone from gasoline vehicles was only 30% greater than product/process-related sources, and by 2020 product/process-related emissions will surpass on-road motor vehicle contributions in the basin (Figure 2c). Potential ozone from on-road diesel vehicles in 2015 is only ~5% of that from product/process-related sources. Contributions to potential ozone from product/process-related sources are 33% alcohols, 29% aromatics, and 12% alkanes (not including unspciated “LVP-VOCs”) (Table S2). A recent ozone formation sensitivity analysis of solvent-related emissions speciation with 22 lumped species demonstrated that using input source profiles that are more detailed in terms of contributing compound classes would improve ozone model performance (von Schneidemesser et al., 2016).

Field Code Changed

Contributions from off-road combustion-related sources ~~to potential SOA and ozone~~: Off-road mobile sources are also significant sources of reactive precursors to SOA and ozone across a very diverse mix of ~~vehicles, boats, equipment, and other engines, and sometimes operate on specialized fuels (e.g. aviation gasoline, jet fuel, jet naphtha, fuel oil) sources and fuels~~ (Cox et al., 2013; Gordon et al., 2013; May et al., 2014; Zhao et al., 2016). These sources have received greater regulatory attention in the past 20 years, and control policies (e.g. CARB, EPA) will add additional variance to the magnitude and composition of ~~gas phase organic compounds they emit~~missions (California Air Resources Board, 2017; Miller and Facanha, 2014; U.S. EPA, 2017). Given the limited information on their diverse source profiles, and thus their SOA and ozone yields (Gordon et al., 2013; McDonald et al., 2015), we constrain their uncertain contributions independent from on-road sources. Their fraction of ~~anthropogenic~~-VOC emissions in the SoCAB leveled off ~~after 2005~~ and started to decrease after 2005 (Figure 2a), and in 2015 the ratio of on- to off-road

emissions in the ARB inventory ~~almanac~~ is 1.5:1 and 1:2 for gasoline- and diesel-powered engines, respectively, when including equipment, recreational vehicles, boats, trains, and aircraft that use either gasoline or diesel (Cox et al., 2013). ~~So, given~~ Given similar ozone and SOA yields to on-road gasoline or diesel vehicles, off-road emissions could approximately increase potential SOA and ozone ~~contributions~~ from gasoline-~~related sources by 67%~~ and diesel-related sources by 67% and 200% for the year ~~%, respectively, in 2015 in the~~ (SoCAB, ~~but are~~). ~~Yet, this is~~ subject to the uncertainty from the wide range of engines, fuels, and ~~emission~~ emission controls affecting the composition of emissions. These results are generally consistent with the relative source contributions of on- and off-road sources to total OA reported by McDonald et al. (2015) for 2010 ~~in the SoCAB~~. The current outsize impact of off-road sources, despite using a relatively small amount of fuels, is due to the fact that emission factors are much higher ~~for off-road sources~~, such as 2+ orders of magnitude higher for gasoline off-road compared to on-road sources (Gordon et al., 2013; McDonald et al., 2015; Zhao et al., 2016). The inclusion of off-road sources does not affect our conclusion ~~in this section~~ that non-combustion sources and motor vehicles (on- and off-road) contribute similar amounts of potential SOA and ozone in our case study megacity around the 2015-2020 period, especially with a rapidly growing role for non-combustion sources as combustion emissions are further controlled. In all, this highlights the importance of continued assessment and regulation of off-road combustion-related sources as part of a holistic air quality management plan ~~along with non-combustion sources and on-road vehicles~~.

~~**Modifying factors for SOA and ozone formation chemistry:** Relative VOC to NO_x ratios have been shown to affect the chemistry and production rates of SOA and ozone. For many cities outside Los Angeles, urban air quality develops within a backdrop of biogenic emissions of VOCs~~

~~and IVOCs, which is critical to keep in mind as we pursue stricter emission targets for reactive organics. In some cases, such as in the Southeast U.S., biogenic emissions dominate over anthropogenic gas phase organics, and emissions of SO₂ and NO_x are large drivers of biogenic SOA formation.~~

4.3 Empirical ambient evidence for IVOC and SVOC emissions, and their SOA contributions

Ambient measurements (2010) of I/SVOCs in our case study city are consistent with our findings; they demonstrate that IVOCs are important contributors to SOA in the region and other urban areas, but major uncertainties persist regarding the sources of primary IVOC emissions (Hayes et al., 2015; Ma et al., 2016; Zhao et al., 2014). Recent model results estimate that 70-86% of urban SOA in Pasadena come from the oxidation of primary I/SVOC emissions (Ma et al., 2016). Zhao et al. (2014) state that unidentified non-vehicular sources contribute ‘substantially’ to these emissions but no clear fraction is yet established. A major fraction of SOA cannot be explained without the inclusion of IVOCs or other unspeci-ated organics (Gentner et al., 2012, 2017; Hayes et al., 2015; Jathar et al., 2014; Zhao et al., 2014). Other results indicate that fossil-related sources contribute approximately half of OA and a majority of fresh, urban SOA in Los Angeles (Gentner et al., 2017; Hayes et al., 2013; Zotter et al., 2014). Yet, bottom-up estimates and top-down assessments of the SOA produced from gasoline and diesel vehicles (on- and off-road) cannot explain all of the observed fossil SOA in LA in 2010, which supports our conclusions and suggests the presence other major sources of fossil-derived SOA precursors (Ensberg et al., 2014; Gentner et al., 2017; Ma et al., 2016; McDonald et al., 2015; Zhao et al., 2014).

4.4 Isotopic carbon content and interpreting ambient isotopic data

The isotopic carbon content (i.e. ^{14}C vs. ^{13}C vs. ^{12}C) of organic aerosol has been used directly, and in tandem with source apportionment of bulk aerosol data from an aerosol mass spectrometer, to infer fossil vs. non-fossil origin of carbonaceous aerosols and their SOA precursors at several locations (Ceburnis et al., 2011; Hayes et al., 2013; Zotter et al., 2014). We tested the potential effect of ~~their~~products/process-related emissions on the interpretation of isotopic measurements of ambient SOA, and our isotopic analysis of 12 products demonstrates that their VOC and I/SVOC emissions and thus SOA will be depleted in Carbon-14 (Table 1). 8 of the 12 contained 97% or more fossil carbon, while the remaining 4 contained 57-81% fossil carbon. Asphalt-related sealants, solvents (e.g. paint thinner, naphtha), and pesticides were found to have the highest fossil carbon content. This is consistent with the fact that petrochemical feedstocks derived from petroleum and other fossil fuels are used in the products, materials, and processes discussed throughout this work.

Studies in greater Los Angeles (Pasadena) report that fossil-fuel driven emissions appear to contribute 68%-74% of the observed afternoon increase in SOA formed from urban sources with the remaining ~25% coming from non-fossil sources, principally regional biogenic and local cooking sources (Hayes et al., 2013, 2015; Zotter et al., 2014). The observed fossil SOA and its potential precursor sources have been analyzed across several studies with differing conclusions (Bahreini et al., 2012; Gentner et al., 2017; Hayes et al., 2013; Zotter et al., 2014). The studies agree that on-road diesel vehicles are a relatively minor contributor to fossil SOA (Bahreini et al., 2012; Hayes et al., 2013, 2015; Zotter et al., 2014), and along with other papers, have ~~highlighted~~proposed the importance of other unconstrained anthropogenic sources (e.g. off-road

[sources](#)). (Ensberg et al., 2014; McDonald et al., 2015; Zhao et al., 2014). Other evidence suggests a fossil source other than on-road diesel since concentrations of IVOCs show only minor weekday vs. weekend variation with changes in diesel traffic (Zhao et al., 2014). Based on our results and the evidence in the literature, we conclude that fossil-derived urban SOA precursors are emitted from product/process-related sources, and are responsible for some of the fossil SOA observed in the SoCAB. The isotopic signature of products, materials, and processes is due to their petrochemical feedstocks, and this has led to their misattribution to combustion-related sources in the past.

5. Conclusions and future research needs

Using multiple bottom-up approaches, we demonstrate the growing importance of non-combustion emissions of gas-phase organic compounds from anthropogenic sources. Yet, our understanding has been inhibited due to the chemical diversity of emissions across a myriad of source types, their fossil isotopic signatures, and in many cases their prolonged emission timescales, which occur over full lifecycles and a broader range of emissions pathways than is typically considered. Emission timescales can extend over months or longer (Figure 4) in the case of [sources of I/SVOCs, emissions via transport through](#) thick layers of materials/coatings ~~sources of I/SVOCs,~~ or the formation and emission of degradation by-products. The implications of these prolonged timescales are a legacy of unreleased potential emissions built up or “banked” in the products and materials spread across urban areas.

672 It is critical to emphasize that these results do not justify deviating attention from, or relaxing
673 emission standards for, combustion-related sources since they are still prominent factors in urban
674 air quality in the developed and developing world, and remain dominant contributors of carbon
675 dioxide. For the foreseeable future in many locations, they remain principal contributors of reactive
676 organic precursors and other criteria pollutants, especially in near-source hotspots, such as
677 roadways. Rather, we conclude that in order to support the coming decades of policy, modern air
678 quality research needs to holistically consider the full portfolio of anthropogenic (and biogenic)
679 sources that impact urban air quality. We highlight key research needs to support this objective.

680

681 ***A broad perspective on non-traditional sources of reactive carbon:*** This analysis highlights the
682 importance and further consideration of VOC, IVOC, and SVOC sources that have not received
683 sufficient attention to effectively support policy. With the successful control of “low-hanging fruit”
684 where single source types dominated emissions, the control of a broader array of disparate sources
685 becomes necessary. It is likely that there are other non-vehicular sources of reactive gas-phase
686 organics in urban areas (e.g. food-derived cooking emissions), which need to be better included in
687 inventories and models. Monitoring, studies, and inventories need to comprehensively include
688 functionalized compound classes rather than just traditional classes.

689

690 ***Validation of existing emissions inventories:*** Our results based on exiting emissions inventories
691 are subject to uncertainties in their methodology. Given the results, further research is needed to
692 review, evaluate, and validate both the emissions factors and source profiles in emissions
693 inventories. A lifecycle approach should include lifetime emissions over the three pathways stated

in Section 2 which include solvent evaporation, solute volatilization and emissions via formation of degradation by-products. Such emissions will be seasonally-dependent on factors such as air velocity, relative humidity, and temperature (Wolkoff, 1998). A large survey of products and materials with attention to national/regional differences is warranted to ensure that source profiles accurately represent product/process-related emissions-, including “green”-intentioned options for comparison. Single-ring aromatics are a key example; the current CARB inventory using the SPECIATE source profiles reports large emissions of single-ring aromatics, 72% of which comes from paints. Yet, this is in contrast to our MSDS ~~survey that~~ analysis of prevalent products, which observed little aromatic content in ~~current day~~ the surveyed U.S. paints (Table 2).

Inclusion of IVOCs and SVOCs in emissions inventories: There is a clear need for more detailed emissions inventories of I/SVOCs from all sources. Further research should support the evaluation of the I/SVOC exemption in strategic air quality management plans, and composition reporting requirements in MSDSs and similar databases. More detailed chemical speciation with newer methods is required to accurately determine the SOA (and ozone) formation potential of unresolved I/SVOC mixtures. ~~Emerging measurement methods will enable a more robust update of the currently unresolved I/SVOCs in existing source profiles, which sometimes stem from outdated survey data~~. In the process, attention should be paid to heterogeneity in products containing petroleum distillates and similar components, which are very broad and can result in an equally broad range of emissions.

715 The off-gassing of I/SVOCs (and VOCs) from materials was not experimentally tested in this study
716 and is not included in inventories. ~~Yet, recent~~Recent studies have shown plastics, foams, and
717 building materials off-gas I/SVOCs and VOCs, ~~such as (e.g.~~ aromatics, aliphatics, halocarbons,
718 terpenes, organophosphates, and oxygenated species~~)~~ (Brodzik et al., 2014; Faber et al., 2013;
719 Kemmlein et al., 2003; Toftum et al., 2008; Wensing et al., 2005). Similarly, detailed lab and field
720 characterization of asphalt-related emissions over long timescales is needed to constrain emission
721 factors, source profiles, and the effect of modified asphalts ~~(e.g. Superpave).~~
722

723 Understanding the application method and environment is key to determining air emissions of
724 I/SVOCs since some products may be used with water and disposed of into wastewater, described
725 as *down-the-drain factors*. For example, a recent ozone modeling study modeled the fate of 23
726 oxygenated I/SVOCs (e.g. glycols, glycol ethers, esters, alcohols) present in cleaning and personal
727 products that are used with water ~~found~~. They concluded that most of the compounds that go down-
728 the-drain do not volatilize and are biodegraded at wastewater treatment plants, while >90% of
729 compounds that volatilize outdoors will react with OH and contribute to ozone formation (Shin et
730 al., 2015).

731
732 ***Quantifying outdoor transport of indoor emissions:*** Similar to *down-the-drain* factors, we
733 highlight the need for a similar factor to determine fractions of indoor emissions that are
734 permanently lost to indoor sinks via chemical or physical deposition (*fraction lost to indoor sinks*).
735 Similarly, models will need factors to account for the increase in characteristic emission timescales
736 when products/materials used on indoor surfaces remain indoors longer due to generally lower

vertical transport coefficients and subsequent re-partitioning to other surfaces prior to transport outdoors.

Inter-location variability in the developed and developing world: While we present annual trends for our case study megacity Los Angeles, a similar situation is evolving elsewhere in developed urban areas where emissions of anthropogenic organics are key drivers of SOA and ozone formation, especially in megacities. We expect substantial heterogeneity between locations in the composition of emissions from products, materials, and processes due to national/regional regulations governing formulation, as well as climate, application specifics, and consumer preferences and options. In particular for this work, California's advanced regulatory program may have led to the phase out of some components in U.S. products. So globally the composition and magnitude of product/process-related emissions may contain a much greater fraction of reactive species, such as single-ring aromatics. Top-down ambient studies and bottom-up studies for other locations are needed to confirm the importance of product/process-related emissions, and air quality modeling studies should support these efforts. The situation in urban areas of developing regions and emerging economies is uncertain since they have motor vehicle emissions standards/technologies that may be much more advanced than the rest of their air quality management plans (Kodjak, 2015). In such locations, non-combustion sources may be important sources, but the combustion of fossil fuels or biomass for home heating/cooking and agricultural waste disposal will play a larger role than in developed regions. In all locations, the magnitude of regional biogenic emissions and SO₂/NO_x emissions will affect the impact of anthropogenic organics on SOA.

Understudied oxidation pathways and products: The oxidation pathways and products for many of the functionalized compound classes discussed in this work (e.g. Figure 3) are largely understudied, with a lack of experimental or theoretical studies to constrain the generation of SOA, ozone, and oxidized gases. Such oxidation products/pathways are also particularly important in indoor environments, so research on their precursor emissions and subsequent oxidation is also important for indoor air chemistry.

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Supporting Information. Please see the supplemental material for additional details on relevant regulations, methods, Tables S1-S9, and Figures S1-S5.

Appendix A: Current and historical regulations and policy on non-combustion products and processes

783 Emissions from consumer products and industrial processes received some attention in pre-2000
784 outdoor air quality research and policy, especially in the cases of toxic components, highly-reactive
785 volatile solvents that fueled rapid ozone production (e.g. alkenes), and stratospheric ozone-
786 depleting chemicals. Emissions of a select few hazardous air pollutants (HAPs) (e.g. benzene and
787 vinyl chloride) were first broadly regulated under the U.S. Clean Air Act (1970). Subsequent
788 amendments through 1990 required the U.S. Environmental Protection Agency (EPA) to regulate
789 key sources of precursors to ozone production and emissions of 189 newly-designated HAPs (now
790 “air toxics”), some of which were used in products and processes (National Research Council,
791 2004). Less well-known, the 1990 amendments also required the EPA to identify priorities and
792 guidelines to mitigate emissions from consumer and commercial products (National Research
793 Council, 2004). Several categories of paints and solvents were subsequently identified based on
794 results from paint drying and chamber experiment studies conducted during early 1970s through
795 late 1990s (Chang et al., 1997; Clausen et al., 1990, 1991, 1993; Hansen, 1974; Sparks et al., 1999;
796 Sullivan, 1975), and regulations were established for half of the product categories, but actions
797 were ultimately halted as these sources were not viewed as central to ozone or other criteria
798 pollutant mitigation at the time (National Research Council, 2004). Despite large uncertainties
799 about emissions and ambient contributions from products/processes, this strategy reflects the
800 magnitude and impact of motor vehicle emissions in 1990 (Figures 1-2); the keen focus on ozone
801 production; and the lack of knowledge on SOA formation chemistry and I/SVOCs.

802

803 U.S. state or air basin-level regulations vary with region and attainment status. California’s air
804 quality policy and regulations have been the most inclusive and detailed with respect to emissions
805 from consumer products and some industrial processes. The California Air Resources Board

806 (CARB) started the Consumer Products Regulatory Program in 1991, and a similar Coatings
807 Program, to address outdoor and indoor air quality problems associated with their emissions by
808 placing product type-specific limits on VOC content, and total reactivity limits (i.e. ozone
809 potential) specifically for aerosol coatings (California Air Resources Board, 2015b). CARB is also
810 required to maintain statewide and county/basin-level emissions inventories for 72 source
811 categories, available 1975 to 2020. Within these categories, the product/process-related sources
812 include consumer products, architectural coatings, pesticides, cleaning and surface coatings, and
813 asphalt paving/roofing. Chemically-speciated emissions profiles for sources within these
814 categories are in the US EPA's SPECIATE repository, some of which are used by the European
815 Union (EU) (Pernigotti et al., 2016). In the 2015 California code of regulations report, CARB has
816 further updated regulations focusing on VOC emissions from antiperspirants and deodorants,
817 hairsprays, other consumer products (both aerosol and non-aerosol), and aerosol coating products
818 (California Air Resources Board, 2015a).

819
820 The Economic Commission of Europe employs market-based mechanisms to reduce regional
821 emissions of VOCs. The National Emissions Ceilings directive sets country-specific reduction
822 targets on organic gas emissions ranging from 10% up to 60% from 2010 to 2020 (European
823 Environment Agency, 2010). The EU has two relevant policy directives: the 'VOC Solvents
824 Emissions Directive' to limit industrial VOC emissions resulting from processes such as printing,
825 surface cleaning, vehicular coating, and dry cleaning; and the 'Paints Directive' to reduce VOC
826 content in paints and varnishes (European Commission, 2014). In China, in addition to restrictions
827 on criteria pollutants including both PM₁₀ and PM_{2.5}, the Chinese Air Pollution Control Action
828 Plan 2013 also limits VOC emissions from paints, adhesives and petrochemical industry, and

promotes the use of low-volatility water-based paints (Ministry of Environmental Protection, 2012).

Appendix B: Calculating characteristic timescales for emissions from surface layers

The timescale for emission is defined by the simple relation:

$$\tau_{emission} = \frac{M_{applied}}{R_{emission}} \quad (1)$$

The mass applied ($M_{applied}$) and rate of emission ($R_{emission}$) are defined as in Weschler and Nazaroff (2008), as a function of emission velocity (v_e) refers to the airborne mass transfer from the surface of the applied layer to the free-stream air, which is the rate-limiting step compared to diffusion within the thin layer (Weschler and Nazaroff, 2008). The gas-phase saturation concentration immediately above the surface (C_{sat}) is determined by molecular structure, C_o is the concentration of a compound in the layer, A is the exposed surface, and d is the depth of the layer.

$$R_{emission} = v_e A C_{sat} \quad (2)$$

$$M_{applied} = C_o A d \quad (3)$$

Plugging in Eqns. 2-3 into Eqn. 1, and substituting in the partitioning coefficient between octanol and air (K_{OA}), one gets Eqns. 4 and 6: Octanol is chosen as the proxy for the mixed organic layer that is applied. K_{OA} is available for a wide range of species and is consistent with previous modeling of SVOC partitioning from surfaces (Weschler and Nazaroff, 2008).

$$\tau_{emission} = \frac{C_o A d}{v_e A C_{sat}} = \frac{C_o d}{v_e C_{sat}} \quad (4)$$

$$\text{since: } K_{OA} = \frac{C_o}{C_{sat}} \quad (5)$$

Field Code Changed

$$\tau_{emission} = \frac{K_{OAd}}{v_e} \quad (6)$$

v_e is a function of aerodynamic and boundary layer resistances (r_a and r_b), and can vary dramatically across indoor and outdoor environments. Given the diversity of compositions, applications, and environments that products and materials will be applied, constraining v_e is uncertain and highly variable. Calculations in Table S9 and Figure 4 are shown for a range of v_e from 10 m hr⁻¹ to 50 m hr⁻¹, which covers a mix of moderately stable to neutral meteorological conditions. Higher values regularly occur for transport from in/out of some plant canopies. Urban values are strongly dependent on the urban landscape and regional meteorology; vertical transport velocities in built up urban areas like Paris range 5-20 m hr⁻¹ (Cherin et al., 2015). Indoor values can decrease to < 5 m hr⁻¹ due to lower friction velocities, which is still fast enough to for the emissions and chemistry that drive indoor air quality (Weschler and Nazaroff, 2008). 10 - 50 m hr⁻¹ was chosen as a daytime range, but we acknowledge that slower vertical velocities and thus longer persistence on surfaces (i.e. lifetimes) may exist for some locations, especially indoors. Equations 7-11 to calculate v_e , r_a , and r_b are reproduced below for location/condition-specific analyses.

$$v_e = \frac{1}{r_a + r_b} \quad (7)$$

$$r_a = \frac{1}{\kappa u^*} \ln \left(\frac{z_r}{z_0} \right) \quad (8)$$

$$r_b = \frac{5Sc^{2/3}}{u^*} \quad (9)$$

$$Sc = \nu / \mathcal{D} \quad (10)$$

$$u^* = \kappa U_r \left[\ln \left(\frac{z_r}{z_0} \right) \right]^{-1} \quad (11)$$

869 where u^* : friction velocity, z_0 : roughness length, z_r : reference height, U_r : reference velocity, κ :
870 von Karman constant, Sc : Schmidt number, ν : kinematic viscosity, \mathcal{D} : gas diffusivity. Depending
871 on the relative impact of r_a vs. r_b , decreases in diffusivity with larger molecules may affect
872 transport.

873

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1363

1364 **Figure captions**

1365

1366 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle fleets. Trends in motor vehicle
1367 population, fuel use, and VOC emissions for model years conforming to pre-LEV, LEV-I, -II, and -III emissions
1368 standards in California's South Coast air basin for 1990-2020 from CARB's EMFAC database (California Air
1369 Resources Board, 2014).

1370

1371 **Figure 2.** Emissions and potential air quality impacts of product/process-related sources and motor vehicles over
1372 time in the South Coast air basin (i.e. Los Angeles). (a) Total VOC emissions (excluding ethane). (b) Potential SOA
1373 from product/process-related emissions compared to on-road motor vehicles, with uncertainties based on the ranges
1374 of compound class-specific SOA yields, and (c) Ozone formation potential via compound-specific maximum ozone
1375 incremental reactivity values (SAPRC, no uncertainty given). Product/process-related results are produced using
1376 CARB emissions data and EPA source profiles; see methods for further detail. Note: None of the panels include
1377 VOCs, IVOCs, or SVOCs, from products/processes that we identify in Sections 2 and 4 as missing from emissions
1378 inventories, but SOA and ozone formation from motor vehicle emissions does include I/SVOCs. Maximum potential
1379 SOA from gasoline vehicles in 1990 was ~41 tons day⁻¹ (off graph). Pre-1990 potential SOA and ozone for motor
1380 vehicles is excluded due to the lack of fleet-resolved data.

1381

1382 **Figure 3.** Average emissions from product/process-related sources for 2005-2020 based on data from CARB
1383 inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b)
1384 major product/process-related source categories in the inventory (values in Table S3). "Miscellaneous" contains
1385 70% mineral spirits (C₇₋₁₂ hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified
1386 in Section 2 and 4.

1387

1388 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
1389 glycol (b), and acetone (c). Based on applied layers of 0.01-1 mm and vertical transport coefficients for
1390 neutral/stable outdoor conditions (10-50 m hr⁻¹), with longer timescales in indoor environments or thicker layers

Field Code Changed

1391 (e.g. asphalt, building materials). The left axis is reproduced on the right in days. Timescales are limited by gas-
1392 phase transport from the surface, but absorption into polymeric or porous substrates could extend timescales
1393 (Weschler and Nazaroff, 2008). Volatility range boundaries for VOC-IVOC and IVOC-SVOC occur between C₁₂-
1394 C₁₃ and C₁₉-C₂₀, respectively, for n-alkanes or compounds with equivalent volatilities. See Appendix B for
1395 calculations and Table S9.

1396

1397 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
1398 represent average yields of the product/process-related source categories in Figure 3, and do not include "missing"
1399 emissions. The red markers show yields of sampled consumer products calculated from speciation obtained after
1400 GC-MS analysis. All are shown as a function of mass emitted, not product composition.

1401 **FIGURES & TABLES**

1402 **Table 1.** Composition and emittable fraction of twelve commercially available consumer products, and results of
 1403 their carbon isotope analysis. Please see Table S1 for detailed mass distribution profiles and Table S7 for detailed
 1404 carbon isotopic analysis results.

Product name	Percent Fossil Origin	IVOC Content	Aromatic Content	Emittable Fraction	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)
Naphtha cleaner	>99%	-	-	100%	-29.0 ± 0.1	-999.4 ± 0.5
Non-polar solvent	>99%	-	93%	100%	-27.4 ± 0.1	-998.5 ± 0.5
Fogging Insecticide [†]	>99%	95%	-	95%	-26.7 ± 0.1	-999.1 ± 0.5
Semi-gloss furniture coating	56%	-	6%	30%	-29.9 ± 0.1	-565.4 ± 0.7
Multipurpose solvent ^a	>99%	-	-	100%	-30.6 ± 0.1	-997.8 ± 0.7
Furniture coating [†]	65%	-	1%	40%	-30.2 ± 0.1	-650.0 ± 0.6
Roof paint ^b	>98%	-	-	-	-22.7 ± 0.1	-985.0 ± 0.5
Sealant ^{c, §}	97%	0.77%	21%	25%	-27.4 ± 0.1	-969.9 ± 0.5
Paint thinner [†]	>99%	2%	3%	100%	-29.1 ± 0.1	-999.7 ± 0.5
Asphalt coating ^{d, §}	>99%	4%	3%	25%	-27.6 ± 0.1	-994.8 ± 0.6
Detergent ^e	81%	3.5%	4%	25%	-27.9 ± 0.1	-807.8 ± 0.6
General purpose cleaner ^{f, †}	81%	1%	-	3%	-27.7 ± 0.1	-811.1 ± 1.3
Multipurpose lubricant	-	39%	-	97%	-	-
Aerosol Coating Product ^{g, †}	-	-	12%	17%	-	-
Flashing cement ^{h, §}	-	0.2%	3.5%	27%	-	-
Crawling Insecticide 1 ^{i, †}	-	20%	-	20%	-	-
Crawling Insecticide 2 ^{j, †}	-	8%	-	8%	-	-

^aCrude oil and plant-derived ethanol were used for multiple reference blanks (Table S8).

[†]Asphalt-related products.

[†]Applied as aerosols.

[‡]Largely consists of acetone (80%) with the rest as cyclotetrasiloxanes and aryl halides.

^bWater based product. Other components include titanium dioxide, silica and aluminum hydroxide.

^cLargely consists of petroleum asphalt, clays and cellulose.

^dLargely consists of petroleum asphalt.

^e46% of the emittable fraction (EF) consists of mostly esters, 70% of which have less than 12 carbon atoms. Terpenes including limonene, eucalyptol and α -terpineol form 37% of the EF.

^fWater-based product. Also has terpenes including camphene, d3-carene, α -pinene, linalool and δ -limonene. 60% of terpene fraction is δ -limonene.

^gContains 35% acetone and 25% hydrocarbon propellants not included in the EF. *25% of EF is 75% acetate + 25% ketones.

^hLargely consists of petroleum asphalt, kaolin, cellulose and aluminum magnesium silicate.

ⁱContains 15% hydrocarbon propellants not included in the EF.

^jContains 25% hydrocarbon propellants not include in the EF.

Note: All fractions given in the footnotes refer to the emittable fraction of a product. Emittable fraction is confirmed with MSDS where possible.

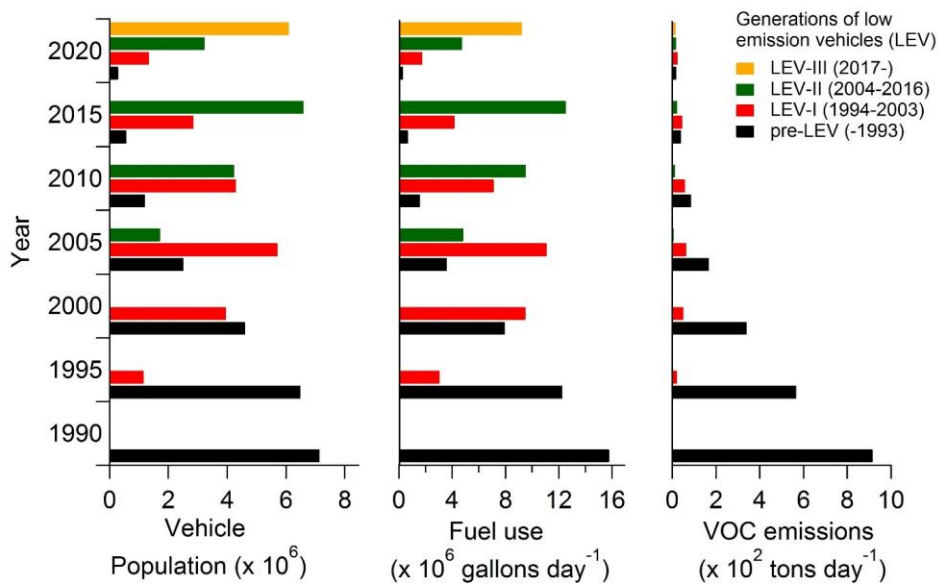
1406 **Table 2.** Analysis of material safety data sheets commercially available with products.

1407

Product Category	Fraction of products with aromatic content	MSDS Aromatics %wt. Mean (range)	Fraction of products with I/SVOC content	MSDS I/SVOCs %wt. Mean (range)
Paints*	0%	0	24% ^a	2.5 (1-5)
Adhesives	27%	8 (1-30)	7% ^b	20 (10-30)
Cleaning Products	8%	5 (1-10)	23% ^c	3.5 (1-7)
Sealants	53%	9 (1-30)	7% ^d	4 (1-10)
Pesticides**	NS	NS	NS	NS

^aConsists of benzoates. Log K_{OA}=7.75 which puts them in the IVOCs range.
^bDoes not include potential emissions from petroleum asphalt which on average constitutes 36% in 13% of the surveyed products. 50% products contain 38% limestone on average.
^cIncludes C9-C14 ethoxylated alcohols.
^dDoes not include potential emissions from petroleum asphalt which on average constitutes 40% in 33% of the surveyed products. 60% products contain 36% limestone on average.
*20% of MSDSs include 'non-hazardous' or 'proprietary' component in composition ranging from 60%-100%.
**MSDSs do not present sufficient information on the composition of pesticides (NS: not sufficient).

1408



1409
1410

1411 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle
1412 population, fuel use, and VOC emissions for model years conforming to pre-LEV, LEV-I, -II, and -III emissions
1413 standards in California's South Coast air basin for 1990-2020 from CARB's EMFAC database (California Air
1414 Resources Board, 2014).

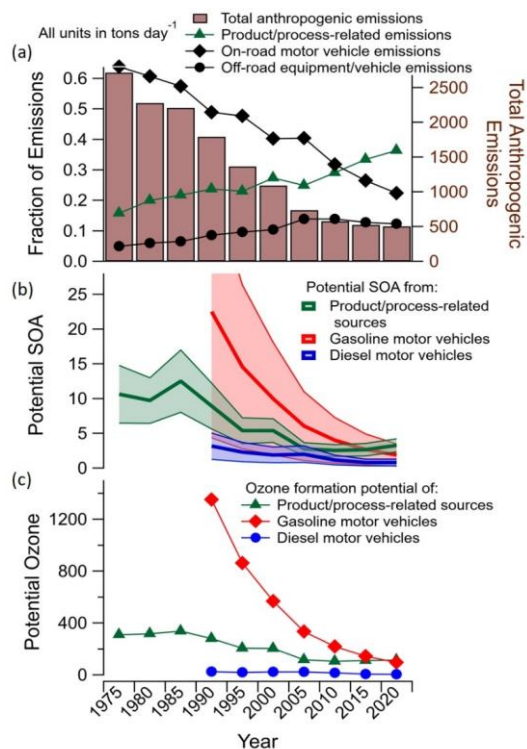
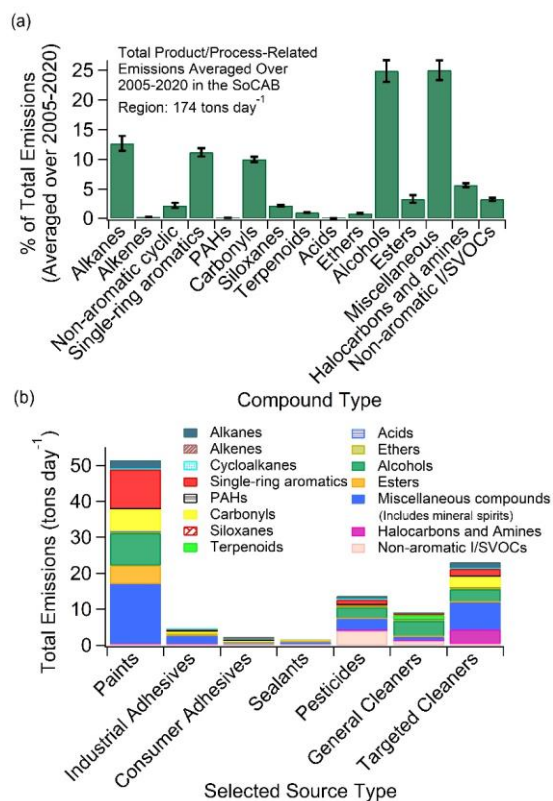
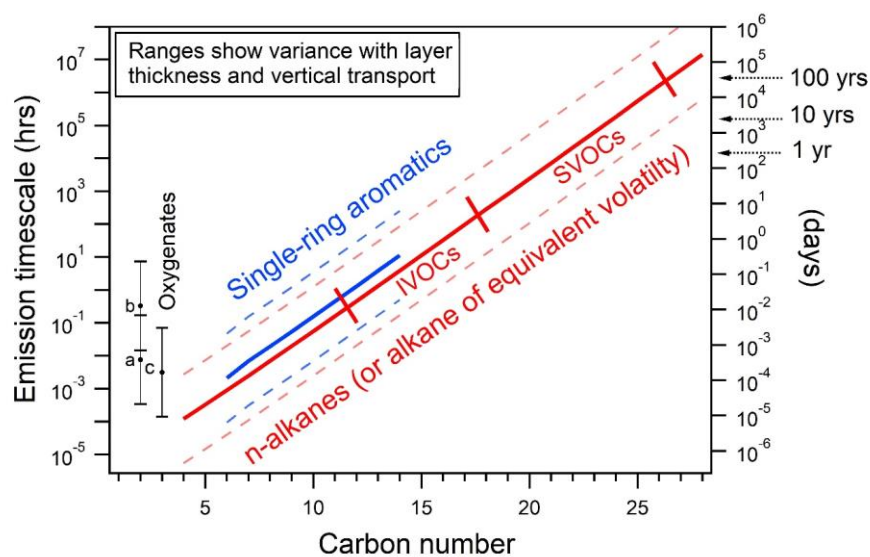


Figure 2. Emissions and potential air quality impacts of product/process-related sources and motor vehicles over time in the South Coast air basin (i.e. Los Angeles). (a) Total VOC emissions (excluding ethane). (b) Potential SOA from product/process-related emissions compared to on-road motor vehicles, with uncertainties based on the ranges of compound class-specific SOA yields, and (c) Ozone formation potential via compound-specific maximum ozone incremental reactivity values (SAPRC, no uncertainty given). Product/process-related results are produced using CARB emissions data and EPA source profiles; see methods for further detail. Note: None of the panels include VOCs, IVOCs, or SVOCs, from products/processes that we identify in Sections 2 and 4 as missing from emissions inventories, but SOA and ozone formation from motor vehicle emissions does include I/SVOCs. Maximum potential SOA from gasoline vehicles in 1990 was ~41 tons day⁻¹ (off graph). Pre-1990 potential SOA and ozone for motor vehicles is excluded due to the lack of fleet-resolved data.

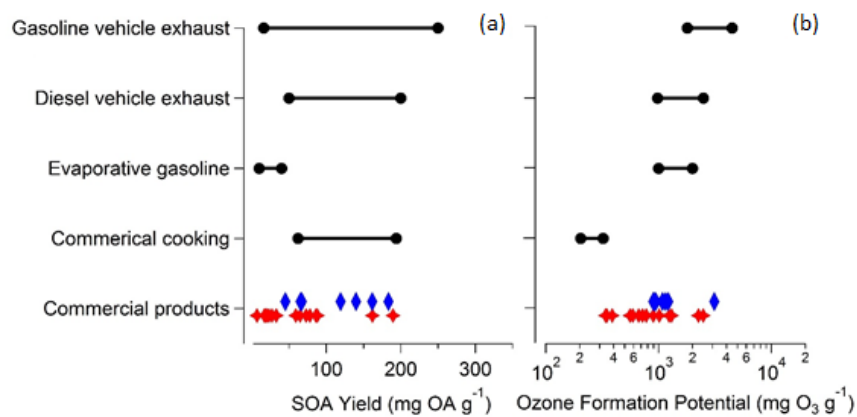


1427
1428
1429 **Figure 3.** Average emissions from product/process-related sources for 2005-2020 based on data from CARB
1430 inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b)
1431 major product/process-related source categories in the inventory (values in Table S3). “Miscellaneous” contains
1432 70% mineral spirits (C₇₋₁₂ hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified
1433 in Section 2 and 4.



1434
 1435 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
 1436 glycol (b), and acetone (c). Based on applied layers of 0.01-1 mm and vertical transport coefficients for
 1437 neutral/stable outdoor conditions ($10\text{-}50\text{ m hr}^{-1}$), with longer timescales in indoor environments or thicker layers
 1438 (e.g. asphalt, building materials). The left axis is reproduced on the right in days. Timescales are limited by gas-
 1439 phase transport from the surface, but absorption into polymeric or porous substrates could extend timescales
 1440 (Weschler and Nazaroff, 2008). Volatility range boundaries for VOC-IVOC and IVOC-SVOC occur between C_{12} -
 1441 C_{13} and C_{19} - C_{20} , respectively, for n-alkanes or compounds with equivalent volatilities. See Appendix B for
 1442 calculations and Table S9.

1443



1444
 1445
 1446 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
 1447 represent average yields of the product/process-related source categories in Figure 3, and do not include "missing"
 1448 emissions. The red markers show yields of sampled consumer products calculated from speciation obtained after
 1449 GC-MS analysis. All are shown as a function of mass emitted, not product composition.

Considering the future of anthropogenic gas-phase organic compound emissions and the increasing influence of non-combustion sources on urban air quality

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Abstract

Decades of policy in developed regions has successfully reduced total anthropogenic emissions of gas-phase organic compounds, especially volatile organic compounds (VOCs), with an intentional, sustained focus on motor vehicles and other combustion-related sources. We examine potential secondary organic aerosol (SOA) and ozone formation in our case study megacity (Los Angeles), and demonstrate that non-combustion-related sources now contribute a major fraction of SOA and ozone precursors. Thus, they warrant greater attention beyond indoor environments to resolve large uncertainties in their emissions, oxidation chemistry, and outdoor air quality impacts in cities worldwide. We constrain the magnitude and chemical composition of emissions via several bottom-up approaches using: chemical analyses of products, emissions inventory assessments,

theoretical calculations of emission timescales, and a survey of consumer product material safety datasheets. We demonstrate that the chemical composition of emissions from consumer products, and commercial/industrial products, processes, and materials is diverse across and within product/material-types with a wide range of SOA and ozone formation potentials that rivals other prominent sources, such as motor vehicles. With emission timescales from minutes to years, emission rates and source profiles need to be included, updated, and/or validated in emissions inventories, with expected regional/national variability. In particular, intermediate-volatility and semivolatile organic compounds (IVOCs and SVOCs) are key precursors to SOA but are excluded or poorly represented in emissions inventories, and exempt from emissions targets. We present an expanded framework for classifying VOC, IVOC, and SVOC emissions from this diverse array of sources that emphasizes a lifecycle approach over longer timescales and three emission pathways that extend beyond the short-term evaporation of VOCs: (1) solvent evaporation, (2) solute off-gassing, and (3) volatilization of degradation by-products. Furthermore, we find that ambient SOA formed from these non-combustion-related emissions could be misattributed to fossil fuel combustion due to the isotopic signature of their petroleum-based feedstocks.

1. Introduction

Anthropogenic emissions of gas-phase organic compounds, including volatile organic compounds (VOCs), are of direct concern as toxic or carcinogenic air pollutants in indoor and outdoor environments (Cohen et al., 2005; Nazaroff and Weschler, 2004; Weschler and Nazaroff, 2008). Often they are more important for air quality as reactive precursors to the formation of outdoor tropospheric ozone and secondary organic aerosol (SOA) as well as indoor SOA, and thus play a central role in the elevated mortality and morbidity rates caused by fine mode particulate matter

(i.e. PM_{2.5}) and ozone in both developed and developing regions (Destailats et al., 2006; Jerrett et al., 2009; Lim et al., 2012; Nazaroff and Weschler, 2004; Pope and Dockery, 2006; Sarwar et al., 2004; Singer et al., 2006; Weschler, 2011). In urban and downwind areas globally, 20-70% of PM_{2.5} is organic aerosol (OA), with SOA comprising 58% of OA in urban areas and 82% downwind on average (Zhang et al., 2007).

Yields of SOA and ozone are strongly dependent on precursor molecular size, volatility, structure, and moieties/functionalities as well as environmental conditions (Gentner et al., 2012; Kroll and Seinfeld, 2008). SOA models have struggled to reproduce observations due to incomplete knowledge on SOA precursors and their sources (Hallquist et al., 2009; Kroll and Seinfeld, 2008). Gas-phase organic compound measurements and emissions data have historically focused on VOCs (i.e. C₂-C₁₂ alkanes and VOCs with equivalent volatilities), but research has demonstrated the prevalence and importance of larger, intermediate-volatility and semivolatile organic compounds (IVOCs and SVOCs, respectively) (Gentner et al., 2012; Kroll and Seinfeld, 2008; Robinson et al., 2007; Zhao et al., 2014). With greater SOA mass yields, IVOCs and SVOCs are key for modeling and mitigating SOA but are less-studied due to measurement difficulties (Gentner et al., 2012; Goldstein and Galbally, 2007). While their emissions from motor vehicles have received more attention, emissions from most other sources are poorly constrained.

In recent history, motor vehicles, power plants, and residential combustion have been dominant drivers of detrimental air quality (e.g., VOCs, nitrogen oxides (NO_x), PM_{2.5}, ozone, sulfur dioxide (SO₂)), with automobiles dominating anthropogenic VOC emissions in major cities (Gentner et

67 al., 2017; Hao et al., 2007; McDonald et al., 2013, 2015; Warneke et al., 2012; Zhou et al., 2003).
68 Yet due to the success of combustion-related emissions control policies over the past 60 years,
69 motor vehicle VOC (and other pollutant) emission factors have decreased by orders of magnitude
70 in route to today's diesel and gasoline vehicles, albeit with some recent on-road diesel NO_x
71 compliance issues (Hao et al., 2007; Kirchstetter et al., 1999; U.S. Environmental Protection
72 Agency, 2011; Warneke et al., 2012). In no other place has this been more studied than Los
73 Angeles, CA, where PM_{2.5} and ozone still exceed standards, and OA comprises 41% of PM₁, 66%
74 of which is SOA (Hayes et al., 2013; Warneke et al., 2012). Recent work has suggested that motor
75 vehicle emissions in L.A. cannot fully explain observations of reactive precursors and SOA
76 (Ensberg et al., 2014; Hayes et al., 2015; McDonald et al., 2015; Zhao et al., 2014). In L.A. and
77 beyond, the role of motor vehicles in degrading air quality will decline further with the newest on-
78 road emissions standards (e.g. Tier 3/LEV-III, Euro 6/VI) (Figure 1), use of electric-powered
79 vehicles, and stricter regulations on emissions from off-road vehicles and other engines (Gentner
80 et al., 2017; Giannouli et al., 2011; Gordon et al., 2013; Platt et al., 2014; Tessum et al., 2014;
81 Weiss et al., 2011).

82
83 The main objective of this paper is to evaluate anthropogenic sources of gas-phase organic
84 compounds that are gaining prominence as motor vehicles and other combustion-related sources
85 become cleaner in the developed and developing world. We demonstrate that consumer products
86 and commercial/industrial products, processes, and materials are important, widely-distributed
87 area sources of VOCs, IVOCs, and SVOCs with variable source profiles making their emissions
88 and impacts difficult to observe and constrain. Our demonstration builds on relevant indoor air
89 quality studies observing emissions and substantial SOA formation from ozone-initiated reactions

with emissions from consumer products, building materials, and cleaning products (Chang et al., 2011; Destailats et al., 2006; Gold et al., 1978; Lewis, 2001; Mitro et al., 2016; Nazaroff and Weschler, 2004; Singer et al., 2006; Weschler, 2011; Weschler and Nazaroff, 2008; Wilke et al., 2004) as well as secondary emissions from oxidative degeneration of indoor paints, varnishes, and materials (Knudsen et al., 1999; Poppendieck et al., 2007a, 2007b; Salthammer and Fuhrmann, 2007).

We outline a holistic framework to assist future research in the field, and then use a multi-dataset approach to constrain emissions, which includes the most detailed emissions inventory available, laboratory analysis of product composition, a survey of consumer product material safety datasheets (MSDSs), and calculations of emissions timescales from theory. Specifically, we (a) evaluate the pathways, chemical composition, and magnitude of VOC, IVOC, and SVOC emissions from prominent types of products, processes, and materials; (b) calculate their potential to form SOA and ozone; and (c) compare their emissions and potential air quality impacts to other prominent sources. We identify major knowledge gaps in emission rates and composition, including I/SVOCs, which we explore via two examples: pesticides and asphalt-related emissions.

2. Defining a comprehensive framework for non-combustion-related emissions

There is a wide body of work on some aspects of emissions from products, processes, and materials in indoor or outdoor environments, but outdoor-focused emissions inventories are mostly focused on evaporative emissions of VOC solvents at the time of application. Here we outline the current

111 scope of emissions and expand on it to define a comprehensive framework to guide future studies
112 on this class of emissions.

113
114 **Volatility:** Emissions of gas-phase organic compounds can and should be differentiated by their
115 volatility: VOCs, IVOCs, and SVOCs, or further by their n-alkane-equivalent volatility (Murphy
116 et al., 2014). One can broadly refer to compounds emitted from products and processes ~~via the~~
117 ~~prefix ‘pp’ (e.g. pp VOCs), which is consistent with existing conventions for volatility basis set~~
118 ~~and aerosol abbreviations (e.g. pp SOA) as products/process-related emissions.~~

119
120 **Source categories:** While the spectrum of sources is very diverse, they are often grouped by use.
121 The following source categories are consistent with the most detailed VOC inventories for
122 consumer products, but are extended (shown with * below) to include incorporate a broader array
123 of materials/ and products, including those used in commercial and industrial processes ~~(shown~~
124 ~~with * below):~~

- 125 • Paints (indoor and outdoor),
- 126 • Industrial adhesives,
- 127 • Consumer adhesives,
- 128 • Sealants,
- 129 • Pesticides (consumer and agricultural),
- 130 • General cleaning products,
- 131 • Targeted cleaners,
- 132 • Personal products (e.g. beauty and hygiene),

- Building materials; (e.g. carpeting, flooring, insulation, wood, gypsum).*
- Furnishings (e.g. furniture).*
- Paving and roofing asphalt-related materials,
- Solid consumer products and packaging (e.g. plastics, elastomers), and their additives (e.g. plasticizers, flame retardants).*

However, the volatility range of most of the existing categories needs to be expanded to completely include I/SVOCs. We discuss I/SVOC-containing pesticides and asphalt-related products/processes as examples that are not sufficiently included in inventories (see Section 4.1.3). Notes: we exclude gasoline evaporation (from vehicles or fuel stations) and fossil fuel extraction/processing given their close connection to combustion. We briefly discuss food-derived cooking emissions for comparison, which are important and classified separately.

Emission pathways: We propose three general pathways of product/process-related emissions:

- 1) Evaporation of solvent from a product while it is in storage or being used, or during a process;
- 2) Volatilization of solute or active compounds from an applied coating or a solid product/material (i.e. off-gassing); and
- 3) Volatilization of by-products from the degradation or transformation of solute or active compounds; or a solid product/material.

The first is the principal pathway previously considered and often occurs over faster timescales (minutes-days) from liquids like cleaners, paints, and other solvents. The second acts over longer timescales (weeks-years), is not always included in inventories, and is a key potential source of

155 I/SVOCs (see Section 4.1.3). SVOC pesticides, flame retardants, and off-gassing plastics are
156 examples studied for indoor air quality (Batterman et al., 2009; Brodzik et al., 2014; Clausen et
157 al., 2004; Faber et al., 2013; Kemmlein et al., 2003; Lewis, 2001; Wensing et al., 2005; Weschler
158 and Nazaroff, 2008). The third is due to the generation of degradation by-products via thermal or
159 photochemical stress, exposure to oxidants (i.e. OH, O₃, or NO₃), or other reactive chemical
160 processes. These may have longer emission timescales depending on oxidant concentrations and
161 the chemical and physical properties of degradation precursors. These emissions are largely
162 understudied with the exception of asphalt-related emissions and ozonation of indoor materials
163 (Poppendieck et al., 2007a, 2007b; Salthammer and Fuhrmann, 2007; Toftum et al., 2008).

164
165 ***A lifecycle approach:*** Emissions during application or immediate use are most commonly studied,
166 but many emissions related to products, process, and materials occur over much longer timescales.
167 In the future, emissions studies need to include all three potential emissions pathways across full
168 lifecycles of:

- 169 • Storage,
- 170 • Transport,
- 171 • Application,
- 172 • ~~Curing~~²²₁₂,
- 173 • Active use,
- 174 • Weathering,
- 175 • Restoration,
- 176 • Removal and disposal.

There are too many facets within the lifecycles of each source category to discuss in this paper, but our case study on asphalt-related emissions in Section 4.1.3 demonstrates several aspects. Production methods are not explicitly included here since they may fall under the purview of industrial point sources, but with small, distributed production it may be appropriate to consider the industrial/commercial processes as area-wide sources. Chemical transformations of materials or products across their lifetimes need to be considered since they can influence the chemical composition and rates of emissions. For example, transformations can occur during a wide variety of in-use conditions or during the storage of unused or partially-used products/materials, both of which can be over long storage periods in a variety of environmental conditions. Application methodology is a determining factor in both emission rates and composition during application. For example, common methods include direct liquid application or aerosolization, either via a pressurized can or a nozzle and air compressor, and aerosolization provides a direct emission pathway for all components and subsequent evaporation of aerosol droplets. Finally, emissions in latter parts of lifecycles (i.e. weathering/deterioration, restoration, removal, and disposal) all need to be considered.

3. Methods

We constrain the magnitude, chemical composition, and potential air quality impacts of emissions using multiple bottom-up approaches: chemical characterization of a selection of consumer products via gas chromatography-mass spectrometry and carbon isotope mass spectrometry; a detailed assessment of emissions inventories and estimation of source contributions to potential SOA and ozone formation; theoretical calculations of emission timescales; and a survey of reported chemical composition in consumer product MSDSs. Details

on the emission timescale calculations can be found in Appendix B, and other additional methods details [pertaining to potential SOA and ozone estimation](#) can be found in the supporting information.

Chemical speciation of consumer products: We selected 17 consumer products across a range of product types with attention to those with unresolved alkane or aromatic mixtures (Table 1). Top-selling products from major commercial providers were selected in order to make a realistic assessment of products that are currently in significant public use, and to avoid biasing the analysis towards less-common products. With a limited sample size, only 1 water- and 1 ethanol-based product were included. Diluted samples were prepared at a concentration of 1000 ng μL^{-1} in either hexane or toluene depending upon the product composition provided in their MSDS. The chemical composition of the emittable fraction of products was determined using gas chromatography with electron ionization-mass spectrometry (GC-EI-MS) (Agilent 7890B/5977). A 1 μL syringe (Hamilton) was used to directly inject the sample onto the GC column through an inlet held at 320 °C. During each injection, the column was initially held at 40 °C for 2 minutes, then ramped at 10 °C min^{-1} to 325 °C and lastly held at 325 °C for 5 minutes. Mass spectra of background subtracted individual ion peaks were used to identify compounds in a sample using the NIST mass spectra library. Calibration curves were drawn for five different concentrations of authentic standards (AccuStandard) for diesel range alkanes ($\text{C}_{10}\text{-C}_{28}$), purgeable aromatics, and terpenes. The emittable organic fraction of raw products is defined by volatility and approximated as VOCs + IVOCs via our chemical analysis and reported MSDS data (Note: SVOCs excluded in this analysis to focus on compounds that are fully partitioned to the gas-phase at equilibrium under typical conditions). A carbon isotopic analysis was also carried out at the Keck Carbon Cycle Accelerator

223 Mass Spectrometer (AMS) facility at U.C. Irvine to measure carbon-14 ($\text{‰ } \Delta^{14}\text{C}$) and carbon-13
224 ($\text{‰ } \text{d}^{13}\text{C}$) relative to carbon-12 via a 0.5MV Compact AMS (National Electrostatics Corp.), and
225 estimate the fossil carbon content in individual products.

226
227 ***Urban emissions inventories:*** We used the California Air Resources Board (CARB) Almanac
228 emissions inventory and the U.S. EPA SPECIATE 4.4 source profiles to generate the total and
229 compound-specific emissions for ~~our~~ California and Los Angeles (Cox et al., 2013; U.S.
230 Environmental Protection Agency, 2014). The CARB emissions inventory is the most detailed
231 available with respect to source categories and basin-level rates. Los Angeles is our case study
232 megacity given its historical role in air quality research and policy with a multi-decadal record of
233 emissions data, ambient measurements, field campaigns, and publications (Bishop and Stedman,
234 2008; Fortin et al., 2005; Neligan, 1962; Warneke et al., 2012). In addition, the U.S. National
235 Emissions Inventory and emission inventories from the Global Emissions Initiative (GEIA)
236 ~~inventories~~ were used for nationwide and worldwide comparison of VOC emissions respectively
237 from solvents and on-road motor vehicles.

238
239 We estimated total daily potential ozone and SOA formation in greater Los Angeles from the
240 product/process-related emissions included in the CARB inventory. Values were also calculated
241 for exhaust and evaporative emissions from on-road motor vehicles using CARB's Emission
242 Factors (EMFAC) model database and literature ozone and SOA yields for each source pathway
243 and low-emission vehicles (LEV) generation (Gentner et al., 2013, 2017; Zhao et al., 2017).
244 Potential ozone formation values are based on maximum ozone incremental reactivity (MOIR)

values from the SAPRC-07 inventory (Carter, 2007; Gentner et al., 2013). The very diverse range of compound classes used in products, materials, and processes remain largely understudied with respect to their SOA yields (e.g. esters, siloxanes). Hence, [specific](#) literature SOA yields were used wherever possible (Algrim and Ziemann, 2016; Chacon-Madrid et al., 2010; Chan et al., 2010; Gentner et al., 2012; Kwok and Atkinson, 1995; Ng et al., 2006; Pankow and Asher, 2008; Sadezky et al., 2006; Tsimpidi et al., 2010), and estimated for other unstudied compounds ~~(without existing~~ [SOA yields \(see Table S6 for details\)](#). SOA yields are estimated at 10 $\mu\text{g OA m}^{-3}$ in urban, “high- NO_x ” conditions (approx. >5 ppb). Ozone and SOA yields for each analyzed consumer product and product categories are compared to other key sources. The SOA yields provide a conservative, lower estimate of potential SOA without aqueous SOA despite studies showing that aqueous pathways to SOA increase SOA yields for small oxidized precursors or their oxidation by-products (Daumit et al., 2016; Jia and Xu, 2014). For the case study city Los Angeles (and Mexico City), aqueous SOA formation was relatively small during major field studies (Dzepina et al., 2009; Hayes et al., 2015; Washenfelder et al., 2011). However, future work in other cities should consider aqueous SOA given the magnitude of oxygenated fraction in product/process-related emissions (see section 4.1.2).

Survey of material safety datasheets (MSDS): We obtained the MSDS data by surveying a set of 88 MSDS entries from the websites of major home improvement stores focusing on their top-selling products. Five product categories were chosen including paints, adhesives, cleaning products, sealants and pesticides. Chemical composition information was extracted from the “composition/information on ingredients” section reported in the datasheets. MSDS entries for commercial products frequently report 30% to 60% of product composition as “proprietary

mixtures”, so this survey only identifies the presence and establishes general ranges for current product types (Table 2).

4. Results and Discussion

4.1 Composition and magnitude of product/process-related emissions

The organic composition of consumer products and commercial/industrial products, materials, and processes are very diverse, which leads to similar diversity in emissions, and further region/nation-specific heterogeneity can be expected. We calculate the magnitude and average chemical composition of emissions from source categories included in the CARB [emissions](#) inventory [and SPECIATE database](#) (Figure 2a) with the goal of assessing the distribution of emissions across organic compound classes and product types (i.e. source profiles) in a megacity with the most representative inventory available. We find that the consumer products and commercial/industrial processes that comprise product/process-related sources are large emitters of a diverse suite of VOCs, but view these results as a lower estimate given likely missing emissions, such as those discussed in [Section 2](#) and [4.1.3](#).

4.1.1 Chemical composition

Laboratory analysis of consumer products: Our results summarized in Table 1 demonstrate the prevalence of non-benzene, single-ring aromatics and C₆-C₁₂ alkane mixtures as solvents, and the presence of IVOCs and SVOCs in consumer products. The emittable fraction of products ranged [from](#) 3-100%, and the single-ring aromatic and IVOC content ranged [from](#) 3.5-93% and 0.77-95%,

respectively. Another key result of this analysis is the frequency of many functionalized aliphatic or aromatic VOCs and IVOCs that are not traditionally measured in atmospheric monitoring (e.g. esters, acetates, siloxanes), which is echoed in the MSDS survey results. More detailed speciation results with a breakdown of alkanes and single-ring aromatic compounds can be found in Table S1. Validation of real-world product/process-related emission rates/timescales is necessary to advance the field but will require examining a wide range of products and geographic conditions.

Analysis of emissions inventories: Figure 3(a) shows the estimated overall composition of product/process-related emissions in Los Angeles averaged over the years 2005 to 2020 determined by combining SPECIATE source profiles with the CARB emissions inventory for the South Coast Air Basin (SoCAB). Alcohols and miscellaneous emissions together make up 50% of the total product/process-related emissions with ethanol as one of the largest individual species emitted, which is consistent with SoCAB ethanol observations that were substantially greater than what would be expected with gasoline vehicles as the only source (Gouw et al., 2012). 70% of the miscellaneous emissions are made up of mineral spirits whose composition varies greatly with application but are generally comprised of acyclic and cyclic C₇₋₁₂ alkanes with variable amounts of aromatic content. The remaining 30% includes unresolved asphalt mixtures, oxygenates, fragrances, and undefined petroleum distillates/oils/spirits, some of which also fall into the I/SVOC category discussed below. Single-ring aromatics are estimated at 13% of total emissions with a mix of 43% toluene, 38% C₈, 3% C₉, and 1.5% C₁₀ aromatics, and minor PAH emissions. Carbonyls represent 10% of total emissions, 36% of which is acetone. Anthropogenic terpenoid emissions in the inventory, while highly reactive, are small relative to other sources and biogenic contributions (see section S.4 in SI).

312

313 Figure 3(b) shows the distribution of organics in the SoCAB emissions inventory from major
314 product/process-related source categories. Paints emerged as the highest VOC emitter, ~21% of
315 which is single-ring aromatics (Figure 3(b) and S4). Targeted cleaners, pesticides, and
316 general cleaners were the next largest source types in the inventory. A more detailed breakdown
317 of emissions broken up by source category and compound class can be found in Table S3. We used
318 the EPA SPECIATE database for source profiles in this study, which had a national scope, some
319 inclusion of I/SVOCs, and greater chemical detail (i.e. number of species). We examined changes
320 in the chemical speciation of total emissions with CARB's 2010 adjustment to SPECIATE in the
321 final years of our study period. We found minor differences in the overall emissions (Figure S5)
322 that are largely attributable to changes in the assignment of unspciated (i.e. miscellaneous) VOC
323 and I/SVOC emissions that do not affect our conclusions. Differences in potential SOA and ozone
324 in the SoCAB for 2010-2020 (Section 4.2) are not clear due to uncertainties in these changes to
325 unspciated assignments, and the uncertainties in SOA yields.

326

327 **MSDS survey:** Summarized in Table 2, the greatest single-ring aromatic content was observed in
328 adhesives (8%) and sealants (9%), while the 30 surveyed paints did not have aromatic VOC content
329 in contrast to the SPECIATE source profiles (Figures 3(b) and S4). In comparison, the chemical
330 speciation of 42 major solvents categorized as 'solvents' (across C₆₋₁₃) in 2002 reports a
331 wide range of total aromatic content (0-100%) and an average of 41% (±46%) with the remainder
332 comprised of acyclic and cyclic alkanes (Censullo et al., 2002). Differences when compared to our
333 present-day laboratory and MSDS paint speciation show reductions in aromatic content due to
334 increasingly stringent regulations in the U.S. However, for other locations, it highlights the likely,

Commented [DG1]: This section also addresses reviewer #1's question from an above section with additional discussion that extends beyond the methods section and intro paragraph to this main section "L269-270: Please add some more information to clarify how the composition is calculated. The source categories are based on the CARB inventory, but where is the speciation coming from? Or are you explaining what you will be doing with the speciate database and then MSDSs etc that is explored in the following sections. This isn't clear. "

continued prevalence of single-ring aromatics in solvents, especially for developing regions. We also found that MSDSs did not report I/SVOCs content in the composition of pesticides ~~while our laboratory speciation found a~~. However, we observed 8%-95% IVOC content in the emittable fraction of the ~~analyzed~~ pesticides samples analyzed in our laboratory. For all of the product categories examined, and especially for pesticides, the amount of compositional information provided by the MSDSs is limited and hinders our ability to constrain the average aromatic or I/SVOC content since compounds outside the VOC range are often not disclosed due to proprietary claims or regulatory exemptions, and where ~~provide~~provided all compound concentrations are usually provided as a wide range. For example, under “fragrance exemptions” VOCs and IVOCs are often not disclosed and can even be labeled as “VOC-free”.

4.1.2 Emission rates

While motor vehicles are still major sources in developed regions, their total gas-phase organic compound emissions have been gradually declining with the continued implementation of stricter emissions standards (Gentner et al., 2017), increasing the relative importance of other sources (Figures 1, 2a). At a global scale, the MACCity and ACCMIP emissions inventories estimate global solvent-related VOC emissions of 15 Tg yr⁻¹ in 2000 and year-over-year increases since 1960 (speciated by aromatics, C₆₊ alkanes, ketones, alcohols, and other VOCs), and emissions of aromatics from solvents are expected to outweigh those from transportation in 2020 (7.5 vs. 6.7 Tg yr⁻¹) (GEIA, 2017). The 2014 U.S. National Emissions Inventory (NEI) data reports that VOC emissions from solvent-related sources are just 25% less (300 tons day⁻¹) than those from on-road mobile sources nationally, while they exceed on-road emissions by 25% (21 tons day⁻¹) in California (U.S. Environmental Protection Agency, 2011). Figure S1 shows California’s statewide

358 bi-decadal emissions estimates from CARB where product/process-related VOCs will reach 450
359 tons day⁻¹ in 2020, exceeding motor vehicles by 116 tons day⁻¹. A similar picture may emerge in
360 developing nations, as the control of motor vehicle emissions is greatly accelerated by the
361 advancements and knowledge of developed nations; such that emission standards in major
362 emerging economies employ either U.S. or E.U. policies, and are generally only one generation
363 behind (Kodjak, 2015). International studies over the past 2 decades show highly varying
364 contributions (~5-45%) to the total anthropogenic VOC emissions from just “solvent” use at both
365 regional and national scales (summarized in SI) (van den Born et al., 1991; Caserini et al., 2004;
366 Chen et al., 2009; Deguillaume et al., 2008; Lu et al., 2007; Markakis et al., 2009; Menut, 2003;
367 Nielsen et al., 2008; Piccot et al., 1992; Song et al., 2007; U.S. Environmental Protection Agency
368 and Office of Air Quality Planning and Standards, 1991).

369
370 Emissions inventory data for Los Angeles demonstrate that anthropogenic emissions have
371 consistently decreased over the last four decades (Figure 2a), which is consistent with ambient
372 observations (1960-2010) (Warneke et al., 2012). As the contribution of on-road motor vehicles
373 to total anthropogenic emissions has declined, product/process-related sources have become a
374 major contributor of VOCs. Based on the CARB emissions inventory, contributions of
375 product/process-related sources are the largest single contributor of VOC emissions in the basin
376 and state (Figures 2a & S1), and are growing with population (i.e. increased in-basin usage). While
377 consumption volume is low compared to combustion fuels, emission factors are higher given that
378 most of the volatile components are emitted whereas fuels are burnt at $\geq 99\%$ efficiency. Yet, no
379 region can be fully representative of product/process-related emissions on a larger scale, and
380 regional/national specifics will influence the magnitude and composition of emissions. For

example, California's extensive regulations will modify the composition of products sold in CA, and likely elsewhere in the U.S.

4.1.3 Emissions of intermediate- and semi-volatile organic compounds (IVOCs and SVOCs)

Despite representing only a small to moderate amount of emissions, IVOCs and SVOCs from motor vehicles are key precursors to urban SOA (Gentner et al., 2012; Robinson et al., 2007). Similarly, we conclude that consumer products and commercial/industrial processes are also large sources of unspciated IVOCs and SVOCs, some of which are included in the CARB inventory, while there is evidence that some other source pathways are not.

Many common products/materials (e.g. pesticides, fragrances, foams, plastics) have IVOCs or SVOCs that partition to reach equilibrium and evaporate over long timescales (Figure 4) (Batterman et al., 2009; Clausen et al., 2004; Mitro et al., 2016; Weschler and Nazaroff, 2010). Our chemical analysis revealed IVOCs or SVOCs in 10 of the 17 products (Tables 1 & S1). Including the MSDS survey, we found composition ranging from 0% to 95% I/SVOCs, with I/SVOCs present in 23% of cleaners, 20% of adhesives, 24% of paints, and 46% of sealants (Table 2). Aliphatic or aromatic I/SVOCs are frequently used in some types of pesticides and are sometimes replaced with biogenic oils (e.g. neem oil, fish oil), such that the bulk of the pesticides in our MSDS survey were comprised of < 1 - 10% active compounds and a balance of undisclosed, "non-hazardous" ingredients.

402 The SPECIATE profiles and CARB emissions inventory include some estimates of unspciated
403 “low-vapor pressure VOCs (LVP-VOCs)” in consumer products that are defined as larger than 12
404 carbon atoms (or equivalent volatility), which is roughly consistent with the beginning of the IVOC
405 range (California Air Resources Board, 2015b). While poorly constrained, non-aromatic I/SVOCs
406 included in the inventory constitute ~3% (6 tons day⁻¹) of total SoCAB emissions estimates in the
407 CARB/SPECIATE case study, 90% of which is classified as unspciated “LVP-VOCs”. Consumer
408 product pesticides, general purpose cleaners, and targeted cleaners (e.g. laundry) are the largest
409 sources of I/SVOC contributions emitting 3.6, 1.0 and 0.4 tons day⁻¹ of I/SVOCs, respectively.
410 Consumer product pesticide emissions had the highest fraction of I/SVOCs (29%).

411
412 While CARB’s consumer products inventory includes I/SVOCs due to manufacturer reporting
413 requirements, I/SVOCs are exempt from limits on VOC content except for multipurpose solvents
414 and paint thinners, and there are known limitations in their coverage, especially of oxygenated
415 species (California Air Resources Board, 2000, 2015b). We conclude there are other anthropogenic
416 sources of IVOCs and SVOCs that have not been considered due to their long emission timescales
417 (i.e. days-years; Figure 4, Table S9) (de Gouw et al., 2011; Weschler and Nazaroff, 2008). We
418 present evidence for two such examples in this paper: I/SVOC-containing pesticides and asphalt-
419 related products, materials, and processes, but other examples include the volatilization of I/SVOC
420 solvents, solutes, or solids (e.g. coatings, flame retardants) and materials that may degrade to form
421 compounds with volatilities of C₁₃₋₂₆ n-alkanes (e.g. construction materials/coatings, including
422 materials with petroleum distillates/residues, mineral oil, coal tar, or similar).

423

Example 1: Emissions of IVOCs and SVOCs from pesticides: We have several pieces of evidence that demonstrate pesticides (including herbicides, insecticides, and fungicides) contain I/SVOCs, but they also highlight the fact they are poorly documented and regulated. We analyzed the chemical composition of three pesticides available as consumer products and they were comprised of 8%, 20% and 95%, IVOCs in the C₁₄₋₁₇ range (Table 1, Figure S3), with trace levels of larger compounds. Our MSDS survey (Table 2) was inconclusive for pesticides since the majority of consumer pesticides are not disclosed in MSDSs due to claims of proprietary mixtures, non-regulated components outside of the VOC range, and/or because they use naturally-derived oils (e.g. neem oil). Studies have shown such naturally-derived oils are comprised of aliphatic and aromatic I/SVOC-range compounds (Isman, 2000; Kumar and Parmar, 19996). Our emissions inventory analysis shows that non-aromatic I/SVOCs are the largest inventoried source of I/SVOCs (Figure 3), but the lack of data in MSDSs and VOC exemptions from regulations suggest this is a lower estimate of actual emissions. To examine commercial/industrial products, we also analyzed the composition of three petroleum-based pesticides used in agriculture (Figure S2), which are comprised of C₁₆-C₂₆ cyclic and acyclic alkanes mostly in the SVOC range that are applied as thin films via a sprayed water emulsion. These can partition to the gas phase and be re-emitted on the timescales in Figure 4 and may impact urban (or downwind) areas in agricultural regions, with 18-29 Gg applied yr⁻¹ in California. California-wide emissions from applied agricultural pesticides were nearly one-fifth of that from consumer products in 2015. However, the high I/SVOCs fraction in their composition and potential to emit over long timescales makes them an important consideration for air quality.

Example 2: Emissions of IVOCs and SVOCs from asphalt-related products, materials, and

processes: We propose asphalt-related products/processes as important sources of IVOCs and SVOCs whose emissions are currently underestimated in inventories and require better quantification. Asphalt-containing materials are used in road paving and repair (and similar applications for roofing or other surfaces), and are comprised of petroleum-derived organic compounds; predominantly non-distillable (i.e. non-volatile) asphalts sometimes with smaller amounts of VOCs, IVOCs, and/or SVOCs. They are used as sealers, coatings, and binders; mixed with aggregates to pave roads; and applied using either high application temperatures, water emulsions, and/or solvent.

The three paving/roofing-related products we analyzed contained aliphatic and aromatic VOCs and IVOCs up to C₁₈ present as solvents, with minor SVOC content (Figure S3). Similarly, there were no I/SVOCs declared in the asphalt-containing products in our MSDS survey. Non-solvent emissions during the hot storage, application, or resurfacing of these asphalts are caused by the degradation (i.e. fragmentation) of larger organic compounds from asphalts to form smaller compounds (C₇-C₃₀), which include cyclic and acyclic alkanes, single-ring aromatics, PAHs (2-, 3-, and 4-ring), and sulfur- or nitrogen-containing species (i.e. benzo- and dibenzo- thiophenes and furans), all of which were not present in the asphalt prior to heating (Cavallari et al., 2012b; Gasthauer et al., 2008; Kitto et al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007; The Asphalt Institute & European Bitumen Association, 2015). The total mass and composition of emissions is dependent on production methods, asphalt grade, and increases in magnitude and maximum molecular weight with storage/application temperatures (i.e. more SVOCs at high temperatures), which ranged 100-240 °C, or higher for roofing asphalts (Cavallari

et al., 2012c; Gasthauer et al., 2008; Kitto et al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007; The Asphalt Institute & European Bitumen Association, 2015).

Emissions of aromatic and aliphatic VOCs, IVOCs, and SVOCs from heated asphalt mixtures (“hot mix”) during application have also been documented in occupational health studies on “asphalt fumes” (Cavallari et al., 2012a, 2012b; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007). Yet, current emissions inventories do not include emissions of VOCs, IVOCs, and SVOCs from the degradation of larger compounds during and after the application of asphalt mixes. Estimation methods focus solely on the evaporation of VOC solvents from “cutback asphalt”, which is a mixture of asphalt and solvent, included as an area source in the “solvent evaporation” category in Californian, U.S., and E.U. inventories (California Air Resources Board and Sonoma Technology Inc., 2003; European Environment Agency, 2017; San Joaquin Valley Air Pollution Control District, 2008; U.S. Environmental Protection Agency, 2014; U.S. EPA, 1995). Road paving solvents are prohibited in non-attainment areas in California (Table S4), so emissions in the SoCAB case study are minor (1 ton day⁻¹) and mostly smaller than C₁₀ in the SPECIATE source profiles (Table S5) (Cox et al., 2013).

Emission factors of degradation byproducts don’t exist, so we approximate lower limits on emission factors only for the period immediately during application using limited published data (see section S.3 in SI), but longer timescale experiments are necessary. Calculated lower limits range from 100-2000 mg kg⁻¹ of asphalt (not including aggregate) with a strong dependence on application/storage temperature. This is on the same order as motor vehicle emission factors and

491 is greater than CARB's current emission factor for hot-mix asphalt (District, 2012; Gentner et al.,
492 2017; San Joaquin Valley Air Pollution Control District, 2008). Yet, California's asphalt
493 consumption of 1,540,000 tons liquid asphalt year⁻¹ (Table S4) represents statewide I/SVOC
494 (+VOC) emissions of 0.5 – 8 tons day⁻¹ during application alone (The Asphalt Institute, 2015).
495 This does not overwhelm current solvent-VOC emissions from paving/roofing (33 tons day⁻¹) but
496 emphasizes the need for further research since the poorly-constrained emissions largely include
497 I/SVOCs emitted over long timescales which are known to have high SOA yields.

498
499 Asphalt-related emissions exemplify the stated need for lifecycle-focused approaches, with
500 potential emissions across storage, transport, application, curing, weathering (e.g. degradation due
501 to climate or UV radiation), and resurfacing. Their magnitude and composition will vary with
502 production/handling methods, geologic source, and application type and location (esp. climate).
503 Emission pathways (from Section 2) include (1) volatilization of application solvents and (3) the
504 production and release of degradation byproducts while (2) does not apply due to negligible off-
505 gassing from extremely low volatile un-degraded asphalt constituents. The emission of asphalt
506 degradation products may peak during construction-related activities primarily due to asphalt's
507 exposure to high temperatures during its storage, application, or resurfacing. Still, seasonal highs
508 in surface temperature (summer pavement maximums are 47-67 °C and up to 70 °C for roofs
509 (Parker et al., 1997; Pomerantz et al., 2000)), will likely affect the rate of internal transport and
510 diffusion out of the "cured" asphalt layer resulting in emissions extended over its lifetime. (Note:
511 paving solvents are currently assumed to be emitted over several months (California Air Resources
512 Board and Sonoma Technology Inc., 2003)).

513

4.2 Potential SOA and ozone formation of product/process-related emissions compared to other major sources

Products and processes emit a diverse array of organic compounds (Figure 3). Some are of relatively-low direct concern for human health (e.g. terpenoids, ~~siloxanes etc.~~), while others present issues as primary emissions, especially in indoor or concentrated workplace environments (e.g. aromatics, ethers, PAHs). Yet, most are reactive and will oxidize in outdoor or indoor environments to form oxidized VOCs with unknown, but large potential health effects (Pöschl and Shiraiwa, 2015). This section focuses on their impacts on air quality via SOA or ozone formation. However, the health or environmental effects of the primary emissions should be especially considered in developing regions where primary VOC emissions are larger, or for specific compound classes near sensitive natural environments. A comparison of the SOA yields and ozone formation potentials for major source categories (Figure 5) demonstrates that product/process-related emissions have SOA yields and ozone formation potentials that are on par with other major urban sources such as motor vehicles and are strongly dependent on composition. ~~Gas-phase cooking emissions from food represent an additional uncertain source in urban air quality along with cooking POA which has been more studied.~~

SOA formation potential: The potential SOA from on-road gasoline vehicles in greater Los Angeles region has decreased by ~65% between the years 1990 and 2015. By 2020, further reduction by 25% is expected relative to the 2015 value of 3.3 tons day⁻¹. These numbers for diesel vehicles are 75% and 1.3% ~~tons day⁻¹~~, respectively. If the emissions inventory is accurate, then the 2015 potential ~~ppproducts/process-related~~-SOA in the SoCAB basin is nearly equal to the SOA formation potential of on-road gasoline and diesel vehicles, and is estimated to surpass them by

2020 with an increasing share of SoCAB's total anthropogenic emissions (Figure 2b). While single-ring aromatics and PAHs constitute only 11% of the total product/process-related emissions, they are responsible for ~80% of the potential SOA from those sources in the region (4% PAHs and 76% single-ring aromatics, largely toluene and xylenes). Existing emission inventories badly underestimate emissions of I/SVOCs and their contributions to SOA. They are shown to be responsible for 0.18% of ~~pp~~products/process-related-SOA, but this excludes 87% of the total I/SVOCs emissions which are labeled as unspecified "LVP-VOCs" in the inventory and thus have no assigned SOA yield in calculations. Additionally, our calculations do not include "missing" emissions or their potential SOA. Anthropogenic terpenoids, including lemon oil, pine oil, orange oil, orange terpenes, D-limonene and α -pinene, are responsible for ~8% of the total product/process-related SOA.

It is important for future work to consider modifying factors for SOA (and ozone) formation chemistry. Relative VOC to NO_x ratios have been shown to affect the chemistry and production rates of SOA and ozone (Hallquist et al., 2009; Sillman, 1999; Zhao et al., 2017). For many cities outside Los Angeles, urban air quality develops within a backdrop of biogenic emissions of VOCs and IVOCs, which is critical to keep in mind as we pursue stricter emission targets for reactive organics. In some cases, such as in the Southeast U.S., biogenic emissions dominate over anthropogenic gas-phase organics, and emissions of SO₂ and NO_x are large drivers of biogenic SOA formation (Xu et al., 2015). Similarly, gas-phase cooking emissions from food represent an additional uncertain source in urban air quality along with cooking POA which has been more studied (Bruns et al., 2017; Hayes et al., 2013; Klein et al., 2016).

Ozone formation potential: Over the past several decades, potential ozone has been dominated by emissions from gasoline motor vehicles. Yet, in 2015 potential ozone from gasoline vehicles was only 30% greater than product/process-related sources, and by 2020 product/process-related emissions will surpass on-road motor vehicle contributions in the basin (Figure 2c). Potential ozone from on-road diesel vehicles in 2015 is only ~5% of that from product/process-related sources. Contributions to potential ozone from product/process-related sources are 33% alcohols, 29% aromatics, and 12% alkanes (not including unspciated “LVP-VOCs”) (Table S2). A recent ozone formation sensitivity analysis of solvent-related emissions speciation with 22 lumped species demonstrated that using input source profiles that are more detailed in terms of contributing compound classes would improve ozone model performance (von Schneidemesser et al., 2016).

Field Code Changed

Contributions from off-road combustion-related sources ~~to potential SOA and ozone~~: Off-road mobile sources are also significant sources of reactive precursors to SOA and ozone across a very diverse mix of ~~vehicles, boats, equipment, and other engines, and sometimes operate on specialized fuels (e.g. aviation gasoline, jet fuel, jet naphtha, fuel oil) sources and fuels~~ (Cox et al., 2013; Gordon et al., 2013; May et al., 2014; Zhao et al., 2016). These sources have received greater regulatory attention in the past 20 years, and control policies (e.g. CARB, EPA) will add additional variance to the magnitude and composition of ~~gas phase organic compounds they emit~~missions (California Air Resources Board, 2017; Miller and Facanha, 2014; U.S. EPA, 2017). Given the limited information on their diverse source profiles, and thus their SOA and ozone yields (Gordon et al., 2013; McDonald et al., 2015), we constrain their uncertain contributions independent from on-road sources. Their fraction of ~~anthropogenic~~-VOC emissions in the SoCAB leveled off ~~after 2005~~ and started to decrease after 2005 (Figure 2a), and in 2015 the ratio of on- to off-road

emissions in the ARB inventory ~~almanac~~ is 1.5:1 and 1:2 for gasoline- and diesel-powered engines, respectively, when including equipment, recreational vehicles, boats, trains, and aircraft that use either gasoline or diesel (Cox et al., 2013). ~~So, given~~ Given similar ozone and SOA yields to on-road gasoline or diesel vehicles, off-road emissions could approximately increase potential SOA and ozone ~~contributions~~ from gasoline-~~related sources by 67%~~ and diesel-related sources by 67% and 200% for the year ~~%, respectively, in 2015 in the~~ (SoCAB, ~~but are~~). ~~Yet, this is~~ subject to the uncertainty from the wide range of engines, fuels, and ~~emission~~ emission controls affecting the composition of emissions. These results are generally consistent with the relative source contributions of on- and off-road sources to total OA reported by McDonald et al. (2015) for 2010 ~~in the SoCAB~~. The current outsize impact of off-road sources, despite using a relatively small amount of fuels, is due to the fact that emission factors are much higher ~~for off-road sources~~, such as 2+ orders of magnitude higher for gasoline off-road compared to on-road sources (Gordon et al., 2013; McDonald et al., 2015; Zhao et al., 2016). The inclusion of off-road sources does not affect our conclusion ~~in this section~~ that non-combustion sources and motor vehicles (on- and off-road) contribute similar amounts of potential SOA and ozone in our case study megacity around the 2015-2020 period, especially with a rapidly growing role for non-combustion sources as combustion emissions are further controlled. In all, this highlights the importance of continued assessment and regulation of off-road combustion-related sources as part of a holistic air quality management plan ~~along with non-combustion sources and on-road vehicles~~.

~~**Modifying factors for SOA and ozone formation chemistry:** Relative VOC to NO_x ratios have been shown to affect the chemistry and production rates of SOA and ozone. For many cities outside Los Angeles, urban air quality develops within a backdrop of biogenic emissions of VOCs~~

~~and IVOCs, which is critical to keep in mind as we pursue stricter emission targets for reactive organics. In some cases, such as in the Southeast U.S., biogenic emissions dominate over anthropogenic gas phase organics, and emissions of SO₂ and NO_x are large drivers of biogenic SOA formation.~~

4.3 Empirical ambient evidence for IVOC and SVOC emissions, and their SOA contributions

Ambient measurements (2010) of I/SVOCs in our case study city are consistent with our findings; they demonstrate that IVOCs are important contributors to SOA in the region and other urban areas, but major uncertainties persist regarding the sources of primary IVOC emissions (Hayes et al., 2015; Ma et al., 2016; Zhao et al., 2014). Recent model results estimate that 70-86% of urban SOA in Pasadena come from the oxidation of primary I/SVOC emissions (Ma et al., 2016). Zhao et al. (2014) state that unidentified non-vehicular sources contribute ‘substantially’ to these emissions but no clear fraction is yet established. A major fraction of SOA cannot be explained without the inclusion of IVOCs or other unspeciatiated organics (Gentner et al., 2012, 2017; Hayes et al., 2015; Jathar et al., 2014; Zhao et al., 2014). Other results indicate that fossil-related sources contribute approximately half of OA and a majority of fresh, urban SOA in Los Angeles (Gentner et al., 2017; Hayes et al., 2013; Zotter et al., 2014). Yet, bottom-up estimates and top-down assessments of the SOA produced from gasoline and diesel vehicles (on- and off-road) cannot explain all of the observed fossil SOA in LA in 2010, which supports our conclusions and suggests the presence other major sources of fossil-derived SOA precursors (Ensberg et al., 2014; Gentner et al., 2017; Ma et al., 2016; McDonald et al., 2015; Zhao et al., 2014).

4.4 Isotopic carbon content and interpreting ambient isotopic data

The isotopic carbon content (i.e. ^{14}C vs. ^{13}C vs. ^{12}C) of organic aerosol has been used directly, and in tandem with source apportionment of bulk aerosol data from an aerosol mass spectrometer, to infer fossil vs. non-fossil origin of carbonaceous aerosols and their SOA precursors at several locations (Ceburnis et al., 2011; Hayes et al., 2013; Zotter et al., 2014). We tested the potential effect of ~~their~~products/process-related emissions on the interpretation of isotopic measurements of ambient SOA, and our isotopic analysis of 12 products demonstrates that their VOC and I/SVOC emissions and thus SOA will be depleted in Carbon-14 (Table 1). 8 of the 12 contained 97% or more fossil carbon, while the remaining 4 contained 57-81% fossil carbon. Asphalt-related sealants, solvents (e.g. paint thinner, naphtha), and pesticides were found to have the highest fossil carbon content. This is consistent with the fact that petrochemical feedstocks derived from petroleum and other fossil fuels are used in the products, materials, and processes discussed throughout this work.

Studies in greater Los Angeles (Pasadena) report that fossil-fuel driven emissions appear to contribute 68%-74% of the observed afternoon increase in SOA formed from urban sources with the remaining ~25% coming from non-fossil sources, principally regional biogenic and local cooking sources (Hayes et al., 2013, 2015; Zotter et al., 2014). The observed fossil SOA and its potential precursor sources have been analyzed across several studies with differing conclusions (Bahreini et al., 2012; Gentner et al., 2017; Hayes et al., 2013; Zotter et al., 2014). The studies agree that on-road diesel vehicles are a relatively minor contributor to fossil SOA (Bahreini et al., 2012; Hayes et al., 2013, 2015; Zotter et al., 2014), and along with other papers, have ~~highlighted~~proposed the importance of other unconstrained anthropogenic sources (e.g. off-road

[sources](#)). (Ensberg et al., 2014; McDonald et al., 2015; Zhao et al., 2014). Other evidence suggests a fossil source other than on-road diesel since concentrations of IVOCs show only minor weekday vs. weekend variation with changes in diesel traffic (Zhao et al., 2014). Based on our results and the evidence in the literature, we conclude that fossil-derived urban SOA precursors are emitted from product/process-related sources, and are responsible for some of the fossil SOA observed in the SoCAB. The isotopic signature of products, materials, and processes is due to their petrochemical feedstocks, and this has led to their misattribution to combustion-related sources in the past.

5. Conclusions and future research needs

Using multiple bottom-up approaches, we demonstrate the growing importance of non-combustion emissions of gas-phase organic compounds from anthropogenic sources. Yet, our understanding has been inhibited due to the chemical diversity of emissions across a myriad of source types, their fossil isotopic signatures, and in many cases their prolonged emission timescales, which occur over full lifecycles and a broader range of emissions pathways than is typically considered. Emission timescales can extend over months or longer (Figure 4) in the case of [sources of I/SVOCs, emissions via transport through](#) thick layers of materials/coatings ~~sources of I/SVOCs,~~ or the formation and emission of degradation by-products. The implications of these prolonged timescales are a legacy of unreleased potential emissions built up or “banked” in the products and materials spread across urban areas.

672 It is critical to emphasize that these results do not justify deviating attention from, or relaxing
673 emission standards for, combustion-related sources since they are still prominent factors in urban
674 air quality in the developed and developing world, and remain dominant contributors of carbon
675 dioxide. For the foreseeable future in many locations, they remain principal contributors of reactive
676 organic precursors and other criteria pollutants, especially in near-source hotspots, such as
677 roadways. Rather, we conclude that in order to support the coming decades of policy, modern air
678 quality research needs to holistically consider the full portfolio of anthropogenic (and biogenic)
679 sources that impact urban air quality. We highlight key research needs to support this objective.

680

681 ***A broad perspective on non-traditional sources of reactive carbon:*** This analysis highlights the
682 importance and further consideration of VOC, IVOC, and SVOC sources that have not received
683 sufficient attention to effectively support policy. With the successful control of “low-hanging fruit”
684 where single source types dominated emissions, the control of a broader array of disparate sources
685 becomes necessary. It is likely that there are other non-vehicular sources of reactive gas-phase
686 organics in urban areas (e.g. food-derived cooking emissions), which need to be better included in
687 inventories and models. Monitoring, studies, and inventories need to comprehensively include
688 functionalized compound classes rather than just traditional classes.

689

690 ***Validation of existing emissions inventories:*** Our results based on exiting emissions inventories
691 are subject to uncertainties in their methodology. Given the results, further research is needed to
692 review, evaluate, and validate both the emissions factors and source profiles in emissions
693 inventories. A lifecycle approach should include lifetime emissions over the three pathways stated

in Section 2 which include solvent evaporation, solute volatilization and emissions via formation of degradation by-products. Such emissions will be seasonally-dependent on factors such as air velocity, relative humidity, and temperature (Wolkoff, 1998). A large survey of products and materials with attention to national/regional differences is warranted to ensure that source profiles accurately represent product/process-related emissions-, including “green”-intentioned options for comparison. Single-ring aromatics are a key example; the current CARB inventory using the SPECIATE source profiles reports large emissions of single-ring aromatics, 72% of which comes from paints. Yet, this is in contrast to our MSDS ~~survey that~~ analysis of prevalent products, which observed little aromatic content in ~~current day~~ the surveyed U.S. paints (Table 2).

Inclusion of IVOCs and SVOCs in emissions inventories: There is a clear need for more detailed emissions inventories of I/SVOCs from all sources. Further research should support the evaluation of the I/SVOC exemption in strategic air quality management plans, and composition reporting requirements in MSDSs and similar databases. More detailed chemical speciation with newer methods is required to accurately determine the SOA (and ozone) formation potential of unresolved I/SVOC mixtures. ~~Emerging measurement methods will enable a more robust update of the currently unresolved I/SVOCs in existing source profiles, which sometimes stem from outdated survey data.~~ In the process, attention should be paid to heterogeneity in products containing petroleum distillates and similar components, which are very broad and can result in an equally broad range of emissions.

715 The off-gassing of I/SVOCs (and VOCs) from materials was not experimentally tested in this study
716 and is not included in inventories. ~~Yet, recent~~Recent studies have shown plastics, foams, and
717 building materials off-gas I/SVOCs and VOCs, ~~such as (e.g.~~ aromatics, aliphatics, halocarbons,
718 terpenes, organophosphates, and oxygenated species~~)~~ (Brodzik et al., 2014; Faber et al., 2013;
719 Kemmlein et al., 2003; Toftum et al., 2008; Wensing et al., 2005). Similarly, detailed lab and field
720 characterization of asphalt-related emissions over long timescales is needed to constrain emission
721 factors, source profiles, and the effect of modified asphalts ~~(e.g. Superpave).~~
722

723 Understanding the application method and environment is key to determining air emissions of
724 I/SVOCs since some products may be used with water and disposed of into wastewater, described
725 as *down-the-drain factors*. For example, a recent ozone modeling study modeled the fate of 23
726 oxygenated I/SVOCs (e.g. glycols, glycol ethers, esters, alcohols) present in cleaning and personal
727 products that are used with water ~~found~~. They concluded that most of the compounds that go down-
728 the-drain do not volatilize and are biodegraded at wastewater treatment plants, while >90% of
729 compounds that volatilize outdoors will react with OH and contribute to ozone formation (Shin et
730 al., 2015).

731
732 ***Quantifying outdoor transport of indoor emissions:*** Similar to *down-the-drain* factors, we
733 highlight the need for a similar factor to determine fractions of indoor emissions that are
734 permanently lost to indoor sinks via chemical or physical deposition (*fraction lost to indoor sinks*).
735 Similarly, models will need factors to account for the increase in characteristic emission timescales
736 when products/materials used on indoor surfaces remain indoors longer due to generally lower

vertical transport coefficients and subsequent re-partitioning to other surfaces prior to transport outdoors.

Inter-location variability in the developed and developing world: While we present annual trends for our case study megacity Los Angeles, a similar situation is evolving elsewhere in developed urban areas where emissions of anthropogenic organics are key drivers of SOA and ozone formation, especially in megacities. We expect substantial heterogeneity between locations in the composition of emissions from products, materials, and processes due to national/regional regulations governing formulation, as well as climate, application specifics, and consumer preferences and options. In particular for this work, California's advanced regulatory program may have led to the phase out of some components in U.S. products. So globally the composition and magnitude of product/process-related emissions may contain a much greater fraction of reactive species, such as single-ring aromatics. Top-down ambient studies and bottom-up studies for other locations are needed to confirm the importance of product/process-related emissions, and air quality modeling studies should support these efforts. The situation in urban areas of developing regions and emerging economies is uncertain since they have motor vehicle emissions standards/technologies that may be much more advanced than the rest of their air quality management plans (Kodjak, 2015). In such locations, non-combustion sources may be important sources, but the combustion of fossil fuels or biomass for home heating/cooking and agricultural waste disposal will play a larger role than in developed regions. In all locations, the magnitude of regional biogenic emissions and SO₂/NO_x emissions will affect the impact of anthropogenic organics on SOA.

Understudied oxidation pathways and products: The oxidation pathways and products for many of the functionalized compound classes discussed in this work (e.g. Figure 3) are largely understudied, with a lack of experimental or theoretical studies to constrain the generation of SOA, ozone, and oxidized gases. Such oxidation products/pathways are also particularly important in indoor environments, so research on their precursor emissions and subsequent oxidation is also important for indoor air chemistry.

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Supporting Information. Please see the supplemental material for additional details on relevant regulations, methods, Tables S1-S9, and Figures S1-S4S5.

Appendix A: Current and historical regulations and policy on non-combustion products and processes

783 Emissions from consumer products and industrial processes received some attention in pre-2000
784 outdoor air quality research and policy, especially in the cases of toxic components, highly-reactive
785 volatile solvents that fueled rapid ozone production (e.g. alkenes), and stratospheric ozone-
786 depleting chemicals. Emissions of a select few hazardous air pollutants (HAPs) (e.g. benzene and
787 vinyl chloride) were first broadly regulated under the U.S. Clean Air Act (1970). Subsequent
788 amendments through 1990 required the U.S. Environmental Protection Agency (EPA) to regulate
789 key sources of precursors to ozone production and emissions of 189 newly-designated HAPs (now
790 “air toxics”), some of which were used in products and processes (National Research Council,
791 2004). Less well-known, the 1990 amendments also required the EPA to identify priorities and
792 guidelines to mitigate emissions from consumer and commercial products (National Research
793 Council, 2004). Several categories of paints and solvents were subsequently identified based on
794 results from paint drying and chamber experiment studies conducted during early 1970s through
795 late 1990s (Chang et al., 1997; Clausen et al., 1990, 1991, 1993; Hansen, 1974; Sparks et al., 1999;
796 Sullivan, 1975), and regulations were established for half of the product categories, but actions
797 were ultimately halted as these sources were not viewed as central to ozone or other criteria
798 pollutant mitigation at the time (National Research Council, 2004). Despite large uncertainties
799 about emissions and ambient contributions from products/processes, this strategy reflects the
800 magnitude and impact of motor vehicle emissions in 1990 (Figures 1-2); the keen focus on ozone
801 production; and the lack of knowledge on SOA formation chemistry and I/SVOCs.

802

803 U.S. state or air basin-level regulations vary with region and attainment status. California’s air
804 quality policy and regulations have been the most inclusive and detailed with respect to emissions
805 from consumer products and some industrial processes. The California Air Resources Board

806 (CARB) started the Consumer Products Regulatory Program in 1991, and a similar Coatings
807 Program, to address outdoor and indoor air quality problems associated with their emissions by
808 placing product type-specific limits on VOC content, and total reactivity limits (i.e. ozone
809 potential) specifically for aerosol coatings (California Air Resources Board, 2015b). CARB is also
810 required to maintain statewide and county/basin-level emissions inventories for 72 source
811 categories, available 1975 to 2020. Within these categories, the product/process-related sources
812 include consumer products, architectural coatings, pesticides, cleaning and surface coatings, and
813 asphalt paving/roofing. Chemically-speciated emissions profiles for sources within these
814 categories are in the US EPA's SPECIATE repository, some of which are used by the European
815 Union (EU) (Pernigotti et al., 2016). In the 2015 California code of regulations report, CARB has
816 further updated regulations focusing on VOC emissions from antiperspirants and deodorants,
817 hairsprays, other consumer products (both aerosol and non-aerosol), and aerosol coating products
818 (California Air Resources Board, 2015a).

819
820 The Economic Commission of Europe employs market-based mechanisms to reduce regional
821 emissions of VOCs. The National Emissions Ceilings directive sets country-specific reduction
822 targets on organic gas emissions ranging from 10% up to 60% from 2010 to 2020 (European
823 Environment Agency, 2010). The EU has two relevant policy directives: the 'VOC Solvents
824 Emissions Directive' to limit industrial VOC emissions resulting from processes such as printing,
825 surface cleaning, vehicular coating, and dry cleaning; and the 'Paints Directive' to reduce VOC
826 content in paints and varnishes (European Commission, 2014). In China, in addition to restrictions
827 on criteria pollutants including both PM₁₀ and PM_{2.5}, the Chinese Air Pollution Control Action
828 Plan 2013 also limits VOC emissions from paints, adhesives and petrochemical industry, and

promotes the use of low-volatility water-based paints (Ministry of Environmental Protection, 2012).

Appendix B: Calculating characteristic timescales for emissions from surface layers

The timescale for emission is defined by the simple relation:

$$\tau_{emission} = \frac{M_{applied}}{R_{emission}} \quad (1)$$

The mass applied ($M_{applied}$) and rate of emission ($R_{emission}$) are defined as in Weschler and Nazaroff (2008), as a function of emission velocity (v_e) refers to the airborne mass transfer from the surface of the applied layer to the free-stream air, which is the rate-limiting step compared to diffusion within the thin layer (Weschler and Nazaroff, 2008). The gas-phase saturation concentration immediately above the surface (C_{sat}) is determined by molecular structure, C_o is the concentration of a compound in the layer, A is the exposed surface, and d is the depth of the layer.

$$R_{emission} = v_e A C_{sat} \quad (2)$$

$$M_{applied} = C_o A d \quad (3)$$

Plugging in Eqns. 2-3 into Eqn. 1, and substituting in the partitioning coefficient between octanol and air (K_{OA}), one gets Eqns. 4 and 6: Octanol is chosen as the proxy for the mixed organic layer that is applied. K_{OA} is available for a wide range of species and is consistent with previous modeling of SVOC partitioning from surfaces (Weschler and Nazaroff, 2008).

$$\tau_{emission} = \frac{C_o A d}{v_e A C_{sat}} = \frac{C_o d}{v_e C_{sat}} \quad (4)$$

$$\text{since: } K_{OA} = \frac{C_o}{C_{sat}} \quad (5)$$

Field Code Changed

$$\tau_{emission} = \frac{K_{OAd}}{v_e} \quad (6)$$

v_e is a function of aerodynamic and boundary layer resistances (r_a and r_b), and can vary dramatically across indoor and outdoor environments. Given the diversity of compositions, applications, and environments that products and materials will be applied, constraining v_e is uncertain and highly variable. Calculations in Table S9 and Figure 4 are shown for a range of v_e from 10 m hr⁻¹ to 50 m hr⁻¹, which covers a mix of moderately stable to neutral meteorological conditions. Higher values regularly occur for transport from in/out of some plant canopies. Urban values are strongly dependent on the urban landscape and regional meteorology; vertical transport velocities in built up urban areas like Paris range 5-20 m hr⁻¹ (Cherin et al., 2015). Indoor values can decrease to < 5 m hr⁻¹ due to lower friction velocities, which is still fast enough to for the emissions and chemistry that drive indoor air quality (Weschler and Nazaroff, 2008). 10 - 50 m hr⁻¹ was chosen as a daytime range, but we acknowledge that slower vertical velocities and thus longer persistence on surfaces (i.e. lifetimes) may exist for some locations, especially indoors. Equations 7-11 to calculate v_e , r_a , and r_b are reproduced below for location/condition-specific analyses.

$$v_e = \frac{1}{r_a + r_b} \quad (7)$$

$$r_a = \frac{1}{\kappa u^*} \ln \left(\frac{z_r}{z_0} \right) \quad (8)$$

$$r_b = \frac{5Sc^{2/3}}{u^*} \quad (9)$$

$$Sc = \nu / \mathcal{D} \quad (10)$$

$$u^* = \kappa U_r \left[\ln \left(\frac{z_r}{z_0} \right) \right]^{-1} \quad (11)$$

869 where u^* : friction velocity, z_0 : roughness length, z_r : reference height, U_r : reference velocity, κ :
870 von Karman constant, Sc : Schmidt number, ν : kinematic viscosity, \mathcal{D} : gas diffusivity. Depending
871 on the relative impact of r_a vs. r_b , decreases in diffusivity with larger molecules may affect
872 transport.

873

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1363

1364 **Figure captions**

1365

1366 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle fleets. Trends in motor vehicle
1367 population, fuel use, and VOC emissions for model years conforming to pre-LEV, LEV-I, -II, and -III emissions
1368 standards in California's South Coast air basin for 1990-2020 from CARB's EMFAC database (California Air
1369 Resources Board, 2014).

1370

1371 **Figure 2.** Emissions and potential air quality impacts of product/process-related sources and motor vehicles over
1372 time in the South Coast air basin (i.e. Los Angeles). (a) Total VOC emissions (excluding ethane). (b) Potential SOA
1373 from product/process-related emissions compared to on-road motor vehicles, with uncertainties based on the ranges
1374 of compound class-specific SOA yields, and (c) Ozone formation potential via compound-specific maximum ozone
1375 incremental reactivity values (SAPRC, no uncertainty given). Product/process-related results are produced using
1376 CARB emissions data and EPA source profiles; see methods for further detail. Note: None of the panels include
1377 VOCs, IVOCs, or SVOCs, from products/processes that we identify in Sections 2 and 4 as missing from emissions
1378 inventories, but SOA and ozone formation from motor vehicle emissions does include I/SVOCs. Maximum potential
1379 SOA from gasoline vehicles in 1990 was ~41 tons day⁻¹ (off graph). Pre-1990 potential SOA and ozone for motor
1380 vehicles is excluded due to the lack of fleet-resolved data.

1381

1382 **Figure 3.** Average emissions from product/process-related sources for 2005-2020 based on data from CARB
1383 inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b)
1384 major product/process-related source categories in the inventory (values in Table S3). "Miscellaneous" contains
1385 70% mineral spirits (C₇₋₁₂ hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified
1386 in Section 2 and 4.

1387

1388 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
1389 glycol (b), and acetone (c). Based on applied layers of 0.01-1 mm and vertical transport coefficients for
1390 neutral/stable outdoor conditions (10-50 m hr⁻¹), with longer timescales in indoor environments or thicker layers

Field Code Changed

1391 (e.g. asphalt, building materials). The left axis is reproduced on the right in days. Timescales are limited by gas-
1392 phase transport from the surface, but absorption into polymeric or porous substrates could extend timescales
1393 (Weschler and Nazaroff, 2008). Volatility range boundaries for VOC-IVOC and IVOC-SVOC occur between C₁₂-
1394 C₁₃ and C₁₉-C₂₀, respectively, for n-alkanes or compounds with equivalent volatilities. See Appendix B for
1395 calculations and Table S9.

1396

1397 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
1398 represent average yields of the product/process-related source categories in Figure 3, and do not include "missing"
1399 emissions. The red markers show yields of sampled consumer products calculated from speciation obtained after
1400 GC-MS analysis. All are shown as a function of mass emitted, not product composition.

FIGURES & TABLES

Table 1. Composition and emittable fraction of twelve commercially available consumer products, and results of their carbon isotope analysis. Please see Table S1 for detailed mass distribution profiles and Table S7 for detailed carbon isotopic analysis results.

Product name	Percent Fossil Origin	IVOC Content	Aromatic Content	Emittable Fraction	$\delta^{13}\text{C}$ (‰)	$\Delta^{14}\text{C}$ (‰)
Naphtha cleaner	>99%	-	-	100%	-29.0 ± 0.1	-999.4 ± 0.5
Non-polar solvent	>99%	-	93%	100%	-27.4 ± 0.1	-998.5 ± 0.5
Fogging Insecticide [†]	>99%	95%	-	95%	-26.7 ± 0.1	-999.1 ± 0.5
Semi-gloss furniture coating	56%	-	6%	30%	-29.9 ± 0.1	-565.4 ± 0.7
Multipurpose solvent ^a	>99%	-	-	100%	-30.6 ± 0.1	-997.8 ± 0.7
Furniture coating [†]	65%	-	1%	40%	-30.2 ± 0.1	-650.0 ± 0.6
Roof paint ^b	>98%	-	-	-	-22.7 ± 0.1	-985.0 ± 0.5
Sealant ^{c, §}	97%	0.77%	21%	25%	-27.4 ± 0.1	-969.9 ± 0.5
Paint thinner [†]	>99%	2%	3%	100%	-29.1 ± 0.1	-999.7 ± 0.5
Asphalt coating ^{d, §}	>99%	4%	3%	25%	-27.6 ± 0.1	-994.8 ± 0.6
Detergent ^e	81%	3.5%	4%	25%	-27.9 ± 0.1	-807.8 ± 0.6
General purpose cleaner ^{f, †}	81%	1%	-	3%	-27.7 ± 0.1	-811.1 ± 1.3
Multipurpose lubricant	-	39%	-	97%	-	-
Aerosol Coating Product ^{g, †}	-	-	12%	17%	-	-
Flashing cement ^{h, §}	-	0.2%	3.5%	27%	-	-
Crawling Insecticide 1 ^{i, †}	-	20%	-	20%	-	-
Crawling Insecticide 2 ^{j, †}	-	8%	-	8%	-	-

^aCrude oil and plant-derived ethanol were used for multiple reference blanks (Table S8).

[†]Asphalt-related products.

[†]Applied as aerosols.

[‡]Largely consists of acetone (80%) with the rest as cyclotetrasiloxanes and aryl halides.

^bWater based product. Other components include titanium dioxide, silica and aluminum hydroxide.

^cLargely consists of petroleum asphalt, clays and cellulose.

^dLargely consists of petroleum asphalt.

^e46% of the emittable fraction (EF) consists of mostly esters, 70% of which have less than 12 carbon atoms. Terpenes including limonene, eucalyptol and α -terpineol form 37% of the EF.

^fWater-based product. Also has terpenes including camphene, d3-carene, α -pinene, linalool and δ -limonene. 60% of terpene fraction is δ -limonene.

^gContains 35% acetone and 25% hydrocarbon propellants not included in the EF. *25% of EF is 75% acetate + 25% ketones.

^hLargely consists of petroleum asphalt, kaolin, cellulose and aluminum magnesium silicate.

ⁱContains 15% hydrocarbon propellants not included in the EF.

^jContains 25% hydrocarbon propellants not included in the EF.

Note: All fractions given in the footnotes refer to the emittable fraction of a product. Emittable fraction is confirmed with MSDS where possible.

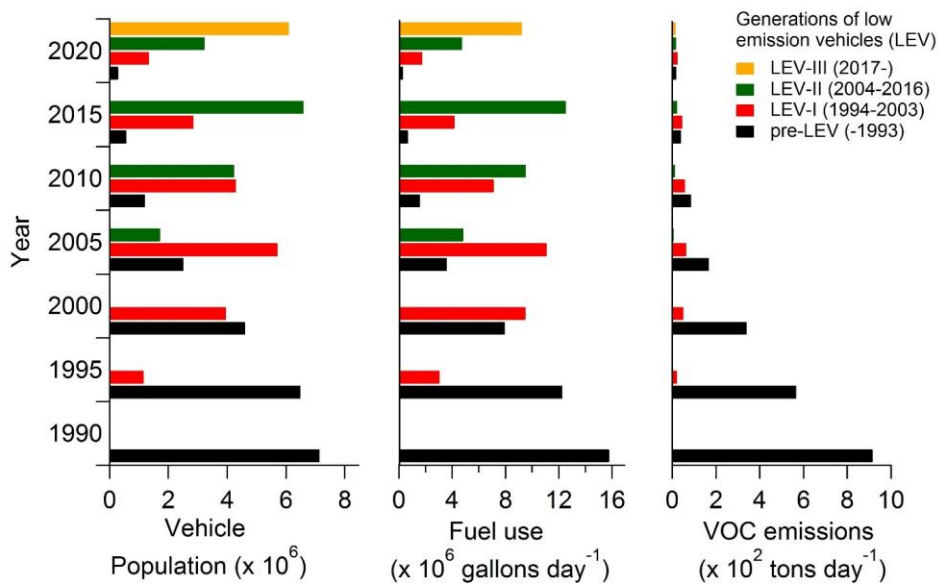
1406 **Table 2.** Analysis of material safety data sheets commercially available with products.

1407

Product Category	Fraction of products with aromatic content	MSDS Aromatics %wt. Mean (range)	Fraction of products with I/SVOC content	MSDS I/SVOCs %wt. Mean (range)
Paints*	0%	0	24% ^a	2.5 (1-5)
Adhesives	27%	8 (1-30)	7% ^b	20 (10-30)
Cleaning Products	8%	5 (1-10)	23% ^c	3.5 (1-7)
Sealants	53%	9 (1-30)	7% ^d	4 (1-10)
Pesticides**	NS	NS	NS	NS

^aConsists of benzoates. Log K_{OA}=7.75 which puts them in the IVOCs range.
^bDoes not include potential emissions from petroleum asphalt which on average constitutes 36% in 13% of the surveyed products. 50% products contain 38% limestone on average.
^cIncludes C9-C14 ethoxylated alcohols.
^dDoes not include potential emissions from petroleum asphalt which on average constitutes 40% in 33% of the surveyed products. 60% products contain 36% limestone on average.
*20% of MSDSs include 'non-hazardous' or 'proprietary' component in composition ranging from 60%-100%.
**MSDSs do not present sufficient information on the composition of pesticides (NS: not sufficient).

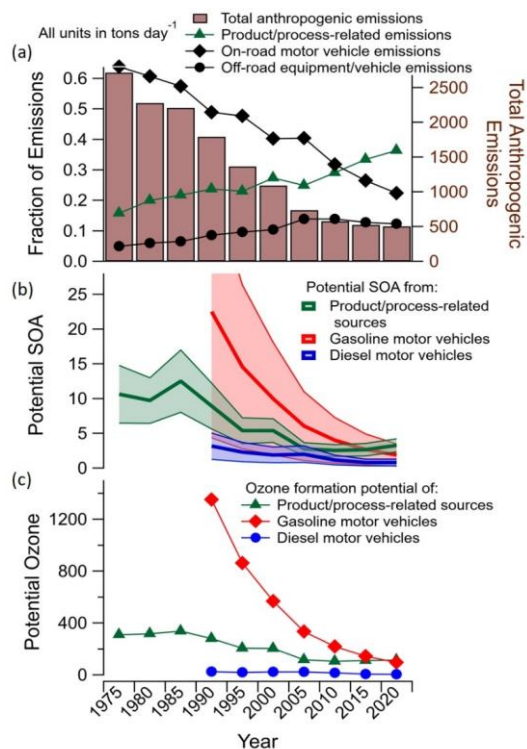
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1411 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle
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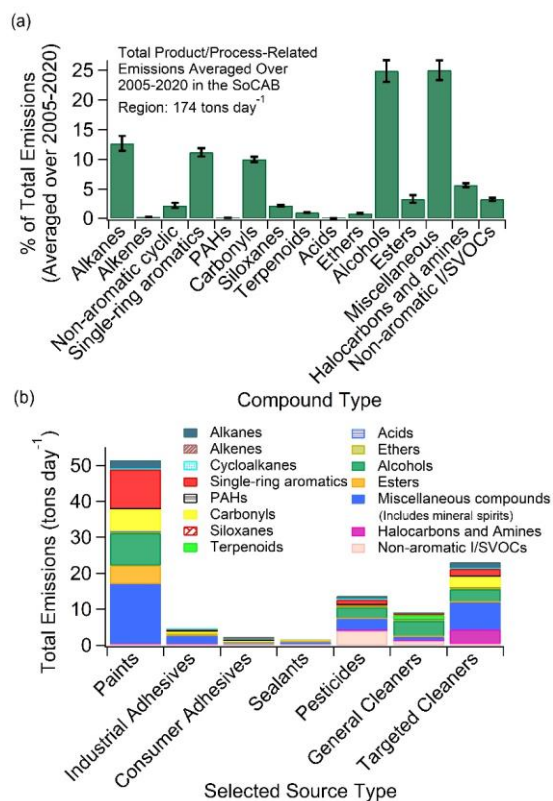
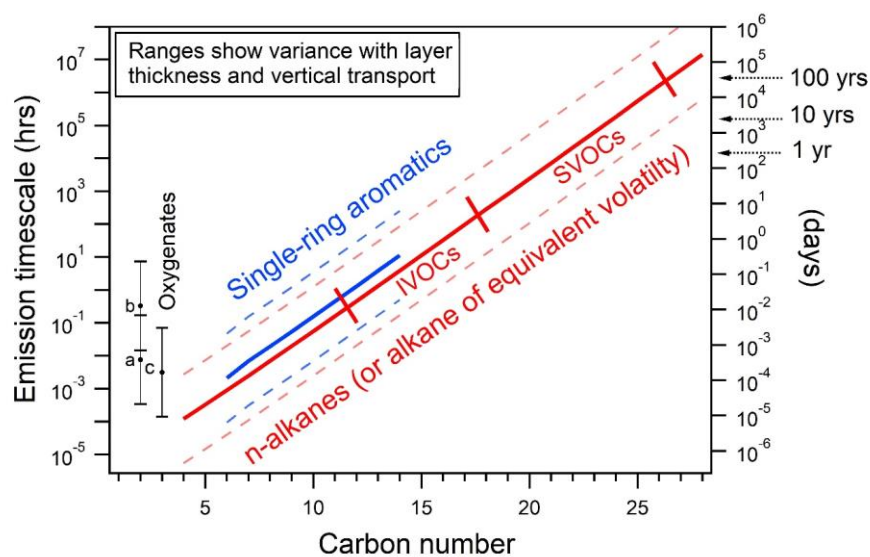
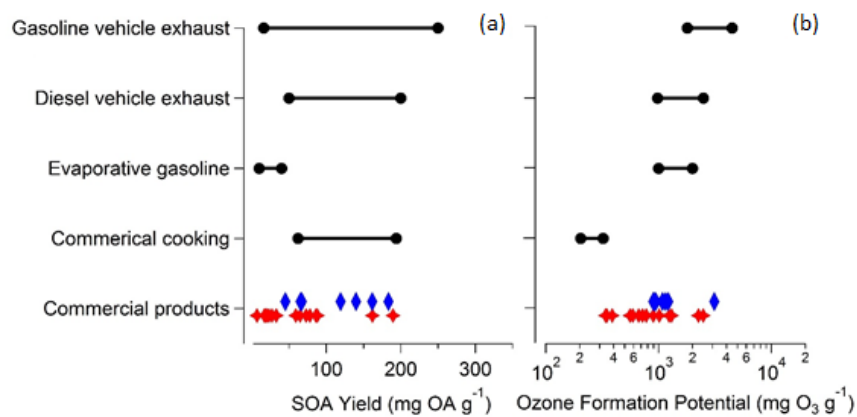


Figure 3. Average emissions from product/process-related sources for 2005-2020 based on data from CARB inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b) major product/process-related source categories in the inventory (values in Table S3). “Miscellaneous” contains 70% mineral spirits (C_{7-12} hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified in Section 2 and 4.



1434
 1435 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
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1444
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 1446 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
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