



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**

4 Peeyush Khare¹ and Drew R. Gentner^{1,2,*}

5 ¹Department of Chemical and Environmental Engineering, Yale University, New Haven CT-06511 USA

6 ²School of Forestry and Environmental Studies, Yale University, New Haven CT-06511 USA

7 * To whom correspondence may be addressed. Email: drew.gentner@yale.edu

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

11

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,



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

37

38 **1. Introduction**

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



45 (i.e. $PM_{2.5}$) and ozone in both developed and developing regions (Destailats et al., 2006; Jerrett et
46 al., 2009; Lim et al., 2012; Nazaroff and Weschler, 2004; Pope and Dockery, 2006; Sarwar et al.,
47 2004; Singer et al., 2006; Weschler, 2011). In urban and downwind areas globally, 20-70% of
48 $PM_{2.5}$ is organic aerosol (OA), with SOA comprising 58% of OA in urban areas and 82%
49 downwind on average (Zhang et al., 2007).

50

51 Yields of SOA and ozone are strongly dependent on precursor molecular size, volatility, structure,
52 and moieties/functionalities as well as environmental conditions (Gentner et al., 2012; Kroll and
53 Seinfeld, 2008). SOA models have struggled to reproduce observations due to incomplete
54 knowledge on SOA precursors and their sources (Hallquist et al., 2009; Kroll and Seinfeld, 2008).
55 Gas-phase organic compound measurements and emissions data have historically focused on
56 VOCs (i.e. C_2 - C_{12} alkanes and VOCs with equivalent volatilities), but research has demonstrated
57 the prevalence and importance of larger, intermediate-volatility and semivolatile organic
58 compounds (IVOCs and SVOCs, respectively) (Gentner et al., 2012; Kroll and Seinfeld, 2008;
59 Robinson et al., 2007; Zhao et al., 2014). With greater SOA mass yields, IVOCs and SVOCs are
60 key for modeling and mitigating SOA but are less-studied due to measurement difficulties
61 (Gentner et al., 2012; Goldstein and Galbally, 2007). While their emissions from motor vehicles
62 have received more attention, emissions from most other sources are poorly constrained.

63

64 In recent history, motor vehicles, power plants, and residential combustion have been dominant
65 drivers of detrimental air quality (e.g., VOCs, nitrogen oxides (NO_x), $PM_{2.5}$, ozone, sulfur dioxide
66 (SO_2)), 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



90 with emissions from consumer products, building materials, and cleaning products (Chang et al.,
91 2011; Destailats et al., 2006; Gold et al., 1978; Lewis, 2001; Mitro et al., 2016; Nazaroff and
92 Weschler, 2004; Singer et al., 2006; Weschler, 2011; Weschler and Nazaroff, 2008; Wilke et al.,
93 2004) as well as secondary emissions from oxidative degeneration of indoor paints, varnishes, and
94 materials (Knudsen et al., 1999; Poppendieck et al., 2007a, 2007b; Salthammer and Fuhrmann,
95 2007).

96

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

106

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

108 There is a wide body of work on some aspects of emissions from products, processes, and materials
109 in indoor or outdoor environments, but outdoor-focused emissions inventories are mostly focused
110 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) (Murphy et al., 2014).

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 to include materials/products used in commercial and
123 industrial processes (shown with * below):

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



- 133 • Paving and roofing asphalt-related materials,
134 • Solid consumer products and packaging (e.g. plastics, elastomers), and their additives (e.g.
135 plasticizers, flame retardants).*

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

142

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

- 144 1) Evaporation of solvent from a product or during a process;
145 2) Volatilization of solute or active compounds from an applied coating or a solid
146 product/material (i.e. off-gassing); and
147 3) Volatilization of by-products from the degradation or transformation of solute or active
148 compounds.

149 The first is the principal pathway previously considered and often occurs over faster timescales
150 (minutes-days) from liquids like cleaners, paints, and other solvents. The second acts over longer
151 timescales (weeks-years), is not always included in inventories, and is a key potential source of
152 I/SVOCs (see Section 4.1.3). SVOC pesticides, flame retardants, and off-gassing plastics are
153 examples studied for indoor air quality (Batterman et al., 2009; Brodzik et al., 2014; Clausen et
154 al., 2004; Faber et al., 2013; Kemmlein et al., 2003; Lewis, 2001; Wensing et al., 2005; Weschler



155 and Nazaroff, 2008). The third is due to the generation of degradation by-products via thermal or
156 photochemical stress, exposure to oxidants (i.e. OH, O₃, or NO₃), or other reactive chemical
157 processes. These emissions are largely understudied with the exception of asphalt-related
158 emissions and ozonation of indoor materials (Poppendieck et al., 2007a, 2007b; Salthammer and
159 Fuhrmann, 2007; Toftum et al., 2008).

160

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

- 165 • Storage,
- 166 • Transport,
- 167 • Application,
- 168 • “Curing”,
- 169 • Active use,
- 170 • Weathering,
- 171 • Restoration,
- 172 • Removal and disposal.

173 There are too many facets within the lifecycles of each source category to discuss in this paper,
174 but our case study on asphalt-related emissions in Section 4.1.3 demonstrates several aspects.
175 Production methods are not explicitly included here since they may fall under the purview of
176 industrial point sources, but with small, distributed production it may be appropriate to consider



177 the industrial/commercial processes as area-wide sources. Chemical transformations of materials
178 or products across their lifetimes need to be considered since they can influence the chemical
179 composition and rates of emissions. For example, transformations can occur during a wide variety
180 of in-use conditions or during the storage of unused or partially-used products/materials, both of
181 which can be over long storage periods in a variety of environmental conditions. Application
182 methodology is a determining factor in both emission rates and composition during application.
183 For example, common methods include direct liquid application or aerosolization, either via a
184 pressurized can or a nozzle and air compressor, and aerosolization provides a direct emission
185 pathway for all components and subsequent evaporation of aerosol droplets. Finally, emissions in
186 latter parts of lifecycles (i.e. weathering/deterioration, restoration, removal, and disposal) all need
187 to be considered.

188

189 **3. Methods**

190 We constrain the magnitude, chemical composition, and potential air quality impacts of
191 emissions using multiple bottom-up approaches: chemical characterization of a selection of
192 consumer products via gas chromatography-mass spectrometry and carbon isotope mass
193 spectrometry; a detailed assessment of emissions inventories and estimation of source
194 contributions to potential SOA and ozone formation; theoretical calculations of emission
195 timescales; and a survey of reported chemical composition in consumer product MSDSs. Details
196 on the emission timescale calculations can be found in Appendix B, and other additional
197 methods details can be found in the supporting information.

198



199 ***Chemical speciation of consumer products:*** We selected 17 consumer products across a range of
200 product types with attention to those with unresolved alkane or aromatic mixtures (Table 1). Top-
201 selling products from major commercial providers were selected in order to make a realistic
202 assessment of products that are currently in significant public use, and to avoid biasing the analysis
203 towards less-common products. With a limited sample size, only 1 water- and 1 ethanol-based
204 product were included. Diluted samples were prepared at a concentration of $1000 \text{ ng } \mu\text{L}^{-1}$ in either
205 hexane or toluene depending upon the product composition provided in their MSDS. The chemical
206 composition of the emittable fraction of products was determined using gas chromatography with
207 electron ionization-mass spectrometry (GC-EI-MS) (Agilent 7890B/5977). A $1 \mu\text{L}$ syringe
208 (Hamilton) was used to directly inject the sample onto the GC column through an inlet held at 320
209 $^{\circ}\text{C}$. During each injection, the column was initially held at $40 \text{ }^{\circ}\text{C}$ for 2 minutes, then ramped at 10
210 $^{\circ}\text{C min}^{-1}$ to $325 \text{ }^{\circ}\text{C}$ and lastly held at $325 \text{ }^{\circ}\text{C}$ for 5 minutes. Mass spectra of background subtracted
211 individual ion peaks were used to identify compounds in a sample using the NIST mass spectra
212 library. Calibration curves were drawn for five different concentrations of authentic standards
213 (AccuStandard) for diesel range alkanes ($\text{C}_{10}\text{-C}_{28}$), purgeable aromatics, and terpenes. The
214 emittable organic fraction of raw products is defined by volatility and approximated as VOCs +
215 IVOCs via our chemical analysis and reported MSDS data (Note: SVOCs excluded in this analysis
216 to focus on compounds that are fully partitioned to the gas-phase at equilibrium under typical
217 conditions). A carbon isotopic analysis was also carried out at the Keck Carbon Cycle Accelerator
218 Mass Spectrometer (AMS) facility at U.C. Irvine to measure carbon-14 ($\text{‰ } \Delta^{14}\text{C}$) and carbon-13
219 ($\text{‰ } \text{d}^{13}\text{C}$) relative to carbon-12 via a 0.5MV Compact AMS (National Electrostatics Corp.), and
220 estimate the fossil carbon content in individual products.

221



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

233

234 We estimated total daily potential ozone and SOA formation in greater Los Angeles from the
235 product/process-related emissions included in the CARB inventory. Values were also calculated
236 for exhaust and evaporative emissions from on-road motor vehicles using CARB's EMFAC model
237 database and literature ozone and SOA yields for each source pathway and LEV generation
238 (Gentner et al., 2013, 2017; Zhao et al., 2017). Potential ozone formation values are based on
239 maximum ozone incremental reactivity (MOIR) values from the SAPRC-07 inventory (Carter,
240 2007; Gentner et al., 2013). The very diverse range of compound classes used in products,
241 materials, and processes remain largely understudied with respect to their SOA yields (e.g. esters,
242 siloxanes). Hence, literature SOA yields were used wherever possible (Algrim and Ziemann, 2016;
243 Chacon-Madrid et al., 2010; Chan et al., 2010; Gentner et al., 2012; Kwok and Atkinson, 1995;
244 Ng et al., 2006; Pankow and Asher, 2008; Sadezky et al., 2006; Tsimpidi et al., 2010), and



245 estimated for other unstudied compounds (Table S6). SOA yields are estimated at $10 \mu\text{g OA m}^{-3}$
246 in urban, “high-NO_x” conditions (approx. >5 ppb). Ozone and SOA yields for each analyzed
247 consumer product and product categories are compared to other key sources. The SOA yields
248 provide a conservative, lower estimate of potential SOA without aqueous SOA despite studies
249 showing that aqueous pathways to SOA increase SOA yields for small oxidized precursors or their
250 oxidation by-products (Daumit et al., 2016; Jia and Xu, 2014). For the case study city Los Angeles
251 (and Mexico City), aqueous SOA formation was relatively small during major field studies
252 (Dzepina et al., 2009; Hayes et al., 2015; Washenfelder et al., 2011). However, future work in
253 other cities should consider aqueous SOA given the magnitude of oxygenated fraction in
254 product/process-related emissions (see section 4.1.2).

255

256 *Survey of material safety datasheets (MSDS):* We obtained the MSDS data by surveying a set of
257 88 MSDS entries from the websites of major home improvement stores focusing on their top-
258 selling products. Five product categories were chosen including paints, adhesives, cleaning
259 products, sealants and pesticides. Chemical composition information was extracted from the
260 “composition/information on ingredients” section reported in the datasheets. MSDS entries for
261 commercial products frequently report 30% to 60% of product composition as “proprietary
262 mixtures”, so this survey only identifies the presence and establishes general ranges for current
263 product types (Table 2).

264

265 **4. Results and Discussion**

266 **4.1 Composition and magnitude of product/process-related emissions**



267 The organic composition of consumer products and commercial/industrial products, materials, and
268 processes are very diverse, which leads to similar diversity in emissions, and further region/nation-
269 specific heterogeneity can be expected. We calculate the magnitude and average chemical
270 composition of emissions from source categories included in the CARB inventory (Figure 2a) with
271 the goal of assessing the distribution of emissions across organic compound classes and product
272 types (i.e. source profiles) in a megacity with the most representative inventory available. We find
273 that the consumer products and commercial/industrial processes that comprise product/process-
274 related sources are large emitters of a diverse suite of VOCs, but view these results as a lower
275 estimate given likely missing emissions, such as those discussed in Section 4.

276

277 **4.1.1 Chemical composition**

278 *Laboratory analysis of consumer products:* Our results summarized in Table 1 demonstrate the
279 prevalence of non-benzene, single-ring aromatics and C₆-C₁₂ alkane mixtures as solvents, and the
280 presence of IVOCs and SVOCs in consumer products. The emittable fraction of products ranged
281 3-100%, and the single-ring aromatic and IVOC content ranged 3.5-93% and 0.77-95%,
282 respectively. Another key result of this analysis is the frequency of many functionalized aliphatic
283 or aromatic VOCs and IVOCs that are not traditionally measured in atmospheric monitoring (e.g.
284 esters, acetates, siloxanes), which is echoed in the MSDS survey results. More detailed speciation
285 results with a breakdown of alkanes and single-ring aromatic compounds can be found in Table
286 S1. Validation of real-world product/process-related emission rates/timescales is necessary to
287 advance the field but will require examining a wide range of products and geographic conditions.

288



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

305

306 Figure 3(b) shows the distribution of organics in the SoCAB emissions inventory from major
307 product/process-related source categories. Paints emerged as the highest VOC emitter, ~21% of
308 which is single-ring aromatics (Figure S4). Targeted cleaners, pesticides, and general cleaners
309 were the next largest source types in the inventory. A more detailed breakdown of emissions
310 broken up by source category and compound class can be found in Table S3.

311



312 **MSDS survey:** Summarized in Table 2, the greatest single-ring aromatic content was observed in
313 adhesives (8%) and sealants (9%), while the 30 surveyed paints did not have aromatic VOC content
314 in contrast to the SPECIATE source profiles. In comparison, the chemical speciation of 42 major
315 solvents (across C₆₋₁₃) in 2002 reports a wide range of total aromatic content (0-100%) and an
316 average of 41% ($\pm 46\%$) with the remainder comprised of acyclic and cyclic alkanes (Censullo et
317 al., 2002). Differences when compared to our present-day laboratory and MSDS paint speciation
318 show reductions in aromatic content due to increasingly stringent regulations. However, for other
319 locations, it highlights the likely, continued prevalence of single-ring aromatics in solvents,
320 especially for developing regions. We also found that MSDSs did not report I/SVOCs content in
321 the composition of pesticides while our laboratory speciation found a 8%-95% IVOC content in
322 the emittable fraction of the analyzed pesticides samples. For all of the product categories
323 examined, and especially for pesticides, the amount of compositional information provided by the
324 MSDSs is limited and hinders our ability to constrain the average aromatic or I/SVOC content
325 since compounds outside the VOC range are often not disclosed due to proprietary claims or
326 regulatory exemptions, and where provide all compound concentrations are usually provided as a
327 wide range. For example, under “fragrance exemptions” VOCs and IVOCs are often not disclosed
328 and can even be labeled as “VOC-free”.

329

330 **4.1.2 Emission rates**

331 While motor vehicles are still major sources in developed regions, their total gas-phase organic
332 compound emissions have been gradually declining with the continued implementation of stricter
333 emissions standards (Gentner et al., 2017), increasing the relative importance of other sources
334 (Figures 1, 2a). At a global scale, the MACCity and ACCMIP emissions inventories estimate



335 global solvent-related VOC emissions of 15 Tg yr⁻¹ in 2000 and year-over-year increases since
336 1960 (speciated by aromatics, C₆₊ alkanes, ketones, alcohols, and other VOCs), and emissions of
337 aromatics from solvents are expected to outweigh those from transportation in 2020 (7.5 vs. 6.7
338 Tg yr⁻¹) (GEIA, 2017). The 2014 U.S. National Emissions Inventory (NEI) data reports that VOC
339 emissions from solvent-related sources are just 25% less (300 tons day⁻¹) than those from on-road
340 mobile sources nationally, while they exceed on-road emissions by 25% (21 tons day⁻¹) in
341 California (U.S. Environmental Protection Agency, 2011). Figure S1 shows California's statewide
342 bi-decadal emissions estimates from CARB where product/process-related VOCs will reach 450
343 tons day⁻¹ in 2020, exceeding motor vehicles by 116 tons day⁻¹. A similar picture may emerge in
344 developing nations, as the control of motor vehicle emissions is greatly accelerated by the
345 advancements and knowledge of developed nations; such that emission standards in major
346 emerging economies employ either U.S. or E.U. policies, and are generally only one generation
347 behind (Kodjak, 2015). International studies over the past 2 decades show highly varying
348 contributions (~5-45%) to the total anthropogenic VOC emissions from just "solvent" use at both
349 regional and national scales (summarized in SI) (van den Born et al., 1991; Caserini et al., 2004;
350 Chen et al., 2009; Deguillaume et al., 2008; Lu et al., 2007; Markakis et al., 2009; Menut, 2003;
351 Nielsen et al., 2008; Piccot et al., 1992; Song et al., 2007; U.S. Environmental Protection Agency
352 and Office of Air Quality Planning and Standards, 1991).

353

354 Emissions inventory data for Los Angeles demonstrate that anthropogenic emissions have
355 consistently decreased over the last four decades (Figure 2a), which is consistent with ambient
356 observations (1960-2010) (Warneke et al., 2012). As the contribution of on-road motor vehicles
357 to total anthropogenic emissions has declined, product/process-related sources have become a



358 major contributor of VOCs. Based on the CARB emissions inventory, contributions of
359 product/process-related sources are the largest single contributor of VOC emissions in the basin
360 and state (Figures 2a & S1), and are growing with population (i.e. increased in-basin usage). While
361 consumption volume is low compared to combustion fuels, emission factors are higher given that
362 most of the volatile components are emitted whereas fuels are burnt at $\geq 99\%$ efficiency. Yet, no
363 region can be fully representative of product/process-related emissions on a larger scale, and
364 regional/national specifics will influence the magnitude and composition of emissions. For
365 example, California's extensive regulations will modify the composition of products sold in CA,
366 and likely elsewhere in the U.S.

367

368 **4.1.3 Emissions of intermediate- and semi-volatile organic compounds (IVOCs and SVOCs)**

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

374

375 Many common products/materials (e.g. pesticides, fragrances, foams, plastics) have IVOCs or
376 SVOCs that partition to reach equilibrium and evaporate over long timescales (Figure 4)
377 (Batterman et al., 2009; Clausen et al., 2004; Mitro et al., 2016; Weschler and Nazaroff, 2010).
378 Our chemical analysis revealed IVOCs or SVOCs in 10 of the 17 products (Tables 1 & S1).
379 Including the MSDS survey, we found composition ranging from 0% to 95% I/SVOCs, with



380 I/SVOCs present in 23% of cleaners, 20% of adhesives, 24% of paints, and 46% of sealants (Table
381 2). Aliphatic or aromatic I/SVOCs are frequently used in some types of pesticides and are
382 sometimes replaced with biogenic oils (e.g. neem oil, fish oil), such that the bulk of the pesticides
383 in our MSDS survey were comprised of < 1 - 10% active compounds and a balance of undisclosed,
384 "non-hazardous" ingredients.

385

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

395

396 While CARB's consumer products inventory includes I/SVOCs due to manufacturer reporting
397 requirements, I/SVOCs are exempt from limits on VOC content except for multipurpose solvents
398 and paint thinners, and there are known limitations in their coverage, especially of oxygenated
399 species (California Air Resources Board, 2000a, 2015b). We conclude there are other
400 anthropogenic sources of IVOCs and SVOCs that have not been considered due to their long
401 emission timescales (i.e. days-years; Figure 4, Table S9) (de Gouw et al., 2011; Weschler and



402 Nazaroff, 2008). We present evidence for two such examples in this paper: I/SVOC-containing
403 pesticides and asphalt-related products, materials, and processes, but other examples include the
404 volatilization of I/SVOC solvents, solutes, or solids (e.g. coatings, flame retardants) and materials
405 that may degrade to form compounds with volatilities of C₁₃₋₂₆ n-alkanes (e.g. construction
406 materials/coatings, including materials with petroleum distillates/residues, mineral oil, coal tar, or
407 similar).

408

409 ***Example 1: Emissions of IVOCs and SVOCs from pesticides:*** We have several pieces of evidence
410 that demonstrate pesticides (including herbicides, insecticides, and fungicides) contain I/SVOCs,
411 but they also highlight the fact they are poorly documented and regulated. We analyzed the
412 chemical composition of three pesticides available as consumer products and they were comprised
413 of 8%, 20% and 95%, IVOCs in the C₁₄₋₁₇ range (Table 1, Figure S3), with trace levels of larger
414 compounds. Our MSDS survey (Table 2) was inconclusive for pesticides since the majority of
415 consumer pesticides are not disclosed in MSDSs due to claims of proprietary mixtures, non-
416 regulated components outside of the VOC range, and/or because they use naturally-derived oils
417 (e.g. neem oil). Studies have shown such naturally-derived oils are comprised of aliphatic and
418 aromatic I/SVOC-range compounds (Isman, 2000; Kumar and Parmar, 19996). Our emissions
419 inventory analysis shows that non-aromatic I/SVOCs are the largest inventoried source of
420 I/SVOCs (Figure 3), but the lack of data in MSDSs and VOC exemptions from regulations suggest
421 this is a lower estimate of actual emissions. To examine commercial/industrial products, we also
422 analyzed the composition of three petroleum-based pesticides used in agriculture (Figure S2),
423 which are comprised of C_{16-C26} cyclic and acyclic alkanes mostly in the SVOC range that are
424 applied as thin films via a sprayed water emulsion. These can partition to the gas phase and be re-



425 emitted on the timescales in Figure 4 and may impact urban (or downwind) areas in agricultural
426 regions, with 18-29 Gg applied yr⁻¹ in California.

427

428 ***Example 2: Emissions of IVOCs and SVOCs from asphalt-related products, materials, and***
429 ***processes:*** We propose asphalt-related products/processes as important sources of IVOCs and
430 SVOCs whose emissions are currently underestimated in inventories and require better
431 quantification. Asphalt-containing materials are used in road paving and repair (and similar
432 applications for roofing or other surfaces), and are comprised of petroleum-derived organic
433 compounds; predominantly non-distillable (i.e. non-volatile) asphalts sometimes with smaller
434 amounts of VOCs, IVOCs, and/or SVOCs. They are used as sealers, coatings, and binders; mixed
435 with aggregates to pave roads; and applied using either high application temperatures, water
436 emulsions, and/or solvent.

437

438 The three paving/roofing-related products we analyzed contained aliphatic and aromatic VOCs
439 and IVOCs up to C₁₈ present as solvents, with minor SVOC content (Figure S3). Similarly, there
440 were no I/SVOCs declared in the asphalt-containing products in our MSDS survey. Non-solvent
441 emissions during the hot storage, application, or resurfacing of these asphalts are caused by the
442 degradation (i.e. fragmentation) of larger asphalts to form smaller compounds (C₇-C₃₀), which
443 include cyclic and acyclic alkanes, single-ring aromatics, PAHs (2-, 3-, and 4-ring), and sulfur- or
444 nitrogen-containing species (i.e. benzo- and dibenzo- thiophenes and furans), all of which were
445 not present in the asphalt prior to heating (Cavallari et al., 2012b; Gasthauer et al., 2008; Kitto et
446 al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007; The Asphalt



447 Institute & European Bitumen Association, 2015). The total mass and composition of emissions is
448 dependent on production methods, asphalt grade, and increases in magnitude and maximum
449 molecular weight with storage/application temperatures (i.e. more SVOCs at high temperatures),
450 which ranged 100-240 °C, or higher for roofing asphalts (Cavallari et al., 2012b; Gasthauer et al.,
451 2008; Kitto et al., 1997; Kriech et al., 2002; Lange et al., 2005; Lange and Stroup-Gardiner, 2007;
452 The Asphalt Institute & European Bitumen Association, 2015).

453

454 Emissions of aromatic and aliphatic VOCs, IVOCs, and SVOCs from heated asphalt mixtures
455 (“hot mix”) during application have also been documented in occupational health studies on
456 “asphalt fumes” (Cavallari et al., 2012a, 2012b; Kriech et al., 2002; Lange et al., 2005; Lange and
457 Stroup-Gardiner, 2007). Yet, current emissions inventories do not include emissions of VOCs,
458 IVOCs, and SVOCs from the degradation of larger compounds during and after the application of
459 asphalt mixes. Estimation methods focus solely on the evaporation of VOC solvents from “cutback
460 asphalt”, included as an area source in the “solvent evaporation” category in Californian, U.S., and
461 E.U. inventories (California Air Resources Board and Sonoma Technology Inc., 2003; San Joaquin
462 Valley Air Pollution Control District, 2008; U.S. Environmental Protection Agency, 2014; U.S.
463 EPA, 1995). Road paving solvents are prohibited in non-attainment areas in California (Table S4),
464 so emissions in the SoCAB case study are minor (1 ton day^{-1}) and mostly smaller than C_{10} in the
465 SPECIATE source profiles (Table S5) (Cox et al., 2013).

466

467 Emission factors of degradation byproducts don’t exist, so we approximate lower limits on
468 emission factors only for the period immediately during application using limited published data



469 (see section S.3 in SI), but longer timescale experiments are necessary. Calculated lower limits
470 range from 100-2000 mg kg⁻¹ of asphalt (not including aggregate) with a strong dependence on
471 application/storage temperature. This is on the same order as motor vehicle emission factors and
472 is greater than CARB's current emission factor for hot-mix asphalt (District, 2012; Gentner et al.,
473 2017; San Joaquin Valley Air Pollution Control District, 2008). Yet, California's asphalt
474 consumption of 1,540,000 tons liquid asphalt year⁻¹ (Table S4) represents statewide I/SVOC
475 (+VOC) emissions of 0.5 – 8 tons day⁻¹ during application alone (The Asphalt Institute, 2015).
476 This does not overwhelm current solvent-VOC emissions from paving/roofing (33 tons day⁻¹) but
477 emphasizes the need for further research since the poorly-constrained emissions largely include
478 I/SVOCs emitted over long timescales which are known to have high SOA yields.

479

480 Asphalt-related emissions exemplify the stated need for lifecycle-focused approaches, with
481 potential emissions across storage, transport, application, curing, weathering (e.g. degradation due
482 to climate or UV radiation), and resurfacing. Their magnitude and composition will vary with
483 production/handling methods, geologic source, and application type and location (esp. climate).
484 Emission pathways (from Section 2) include (1) volatilization of application solvents and (3) the
485 production and release of degradation byproducts while (2) does not apply due to negligible off-
486 gassing from extremely low volatile un-degraded asphalt constituents. The emission of asphalt
487 degradation products may peak during construction-related activities primarily due to asphalt's
488 exposure to high temperatures during its storage, application, or resurfacing. Still, seasonal highs
489 in surface temperature (summer pavement maximums are 47-67 °C and up to 70 °C for roofs
490 (Parker et al., 1997; Pomerantz et al., 2000)), will likely affect the rate of internal transport and
491 diffusion out of the "cured" asphalt layer resulting in emissions extended over its lifetime. (Note:



492 paving solvents are currently assumed to be emitted over several months (California Air Resources
493 Board and Sonoma Technology Inc., 2003)).

494

495 **4.2 Potential SOA and ozone formation of product/process-related emissions compared to**
496 **other major sources**

497 Products and processes emit a diverse array of organic compounds (Figure 3). Some are of low
498 direct concern for human health (terpenoids, siloxanes etc.), while others present issues as primary
499 emissions, especially in indoor or concentrated workplace environments (aromatics, ethers,
500 PAHs). Yet, most are reactive and will oxidize in outdoor or indoor environments to form oxidized
501 VOCs with unknown, but large potential health effects (Pöschl and Shiraiwa, 2015). This section
502 focuses on their impacts on air quality via SOA or ozone formation. However, the health or
503 environmental effects of the primary emissions should be especially considered in developing
504 regions where primary VOC emissions are larger, or for specific compound classes near sensitive
505 natural environments.

506

507 A comparison of the SOA yields and ozone formation potentials for major source categories
508 (Figure 5) demonstrates that product/process-related emissions have SOA yields and ozone
509 formation potentials that are on par with other major urban sources such as motor vehicles and are
510 strongly dependent on composition. Gas-phase cooking emissions from food represent an
511 additional uncertain source in urban air quality along with cooking POA which has been more
512 studied (Bruns et al., 2017; Hayes et al., 2013; Klein et al., 2016).

513



514 **SOA formation potential:** The potential SOA from on-road gasoline vehicles in greater Los
515 Angeles region has decreased by ~65% between the years 1990 and 2015. By 2020, further
516 reduction by 25% is expected relative to the 2015 value of 3.3 tons day⁻¹. These numbers for diesel
517 vehicles are 75% and 1.3%, respectively. If the emissions inventory is accurate, then the 2015
518 potential pp-SOA in the SoCAB basin is nearly equal to the SOA formation potential of on-road
519 gasoline and diesel vehicles, and is estimated to surpass them by 2020 with an increasing share of
520 SoCAB's total anthropogenic emissions (Figure 2b). While single-ring aromatics and PAHs
521 constitute only 11% of the total product/process-related emissions, they are responsible for ~80%
522 of the potential SOA from those sources in the region (4% PAHs and 76% single-ring aromatics,
523 largely toluene and xylenes). Existing emission inventories badly underestimate emissions of
524 I/SVOCs and their contributions to SOA. They are shown to be responsible for 0.18% of pp-SOA,
525 but this excludes 87% of the total I/SVOCs emissions which are labeled as unspciated "LVP-
526 VOCs" in the inventory and thus have no assigned SOA yield in calculations. Additionally, our
527 calculations do not include "missing" emissions or their potential SOA. Anthropogenic terpenoids,
528 including lemon oil, pine oil, orange oil, orange terpenes, D-limonene and α -pinene, are
529 responsible for ~8% of the total product/process-related SOA.

530

531 **Ozone formation potential:** Over the past several decades, potential ozone has been dominated by
532 emissions from gasoline motor vehicles. Yet, in 2015 potential ozone from gasoline vehicles was
533 only 30% greater than product/process-related sources, and by 2020 product/process-related
534 emissions will surpass on-road motor vehicle contributions in the basin (Figure 2c). Potential
535 ozone from on-road diesel vehicles is only ~5% of that from product/process-related sources.
536 Contributions to potential ozone from product/process-related sources are 33% alcohols, 29%



537 aromatics, and 12% alkanes (not including unspiciated “LVP-VOCs”) (Table S2). A recent ozone
538 formation sensitivity analysis of solvent-related emissions speciation with 22 lumped species
539 demonstrated that using input source profiles that are more detailed in terms of contributing
540 compound classes would improve ozone model performance (von Schneidemesser et al., 2016).

541

542 ***Contributions from off-road combustion-related sources to potential SOA and ozone:*** Off-road
543 mobile sources are also significant sources of reactive precursors to SOA and ozone across a very
544 diverse mix of vehicles, boats, equipment, and other engines, and sometimes operate on specialized
545 fuels (e.g. aviation gasoline, jet fuel, jet naphtha, fuel oil) (Cox et al., 2013; Gordon et al., 2013;
546 May et al., 2014; Zhao et al., 2016). These sources have received greater regulatory attention in
547 the past 20 years, and control policies (e.g. CARB, EPA) will add additional variance to the
548 magnitude and composition of gas-phase organic compounds they emit (California Air Resources
549 Board, 2017; Miller and Facanha, 2014; U.S. EPA, 2017). Given the limited information on their
550 diverse source profiles, and thus their SOA and ozone yields (Gordon et al., 2013; McDonald et
551 al., 2015), we constrain their uncertain contributions independent from on-road sources. Their
552 fraction of anthropogenic VOC emissions in the SoCAB leveled off after 2005 and started to
553 decrease (Figure 2a), and in 2015 the ratio of on- to off-road emissions in the ARB inventory
554 almanac is 1.5:1 and 1:2 for gasoline- and diesel-powered engines, respectively, when including
555 equipment, recreational vehicles, boats, trains, and aircraft that use either gasoline or diesel (Cox
556 et al., 2013). So, given similar ozone and SOA yields to on-road gasoline or diesel vehicles, off-
557 road emissions could approximately increase potential SOA and ozone contributions from
558 gasoline-related sources by 67% and diesel-related sources by 200% for the year 2015 in the
559 SoCAB, but are subject to the uncertainty from the wide range of engines, fuels, and emissions



560 controls affecting the composition of emissions. These results are generally consistent with the
561 relative source contributions of on- and off-road sources to total OA reported by McDonald et al.
562 (2015) for 2010 in the SoCAB. The current outside impact of off-road sources, despite using a
563 relatively small amount of fuels, is due to the fact that emission factors are much higher for off-
564 road sources, such as 2+ orders of magnitude higher for gasoline off-road compared to on-road
565 sources (Gordon et al., 2013; McDonald et al., 2015; Zhao et al., 2016).

566

567 The inclusion of off-road sources does not affect our conclusion in this section that non-
568 combustion sources and motor vehicles (on- and off-road) contribute similar amounts of potential
569 SOA and ozone in our case study megacity around the 2015-2020 period, with a rapidly growing
570 role for non-combustion sources as combustion emissions are further controlled. In all, this
571 highlights the importance of continued assessment and regulation of off-road combustion-related
572 sources as part of a holistic air quality management plan along with non-combustion sources and
573 on-road vehicles.

574

575 ***Modifying factors for SOA and ozone formation chemistry:*** Relative VOC to NO_x ratios have
576 been shown to affect the chemistry and production rates of SOA and ozone (Hallquist et al., 2009;
577 Sillman, 1999; Zhao et al., 2017). For many cities outside Los Angeles, urban air quality develops
578 within a backdrop of biogenic emissions of VOCs and IVOCs, which is critical to keep in mind as
579 we pursue stricter emission targets for reactive organics. In some cases, such as in the Southeast
580 U.S., biogenic emissions dominate over anthropogenic gas-phase organics, and emissions of SO₂
581 and NO_x are large drivers of biogenic SOA formation (Xu et al., 2015).



582

583 4.3 Empirical ambient evidence for IVOC and SVOC emissions, and their SOA contributions

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

599

600 4.4 Isotopic carbon content and interpreting ambient isotopic data

601 The isotopic carbon content (i.e. ^{14}C vs. ^{13}C vs. ^{12}C) of organic aerosol has been used directly, and
602 in tandem with source apportionment of bulk aerosol data from an aerosol mass spectrometer, to
603 infer fossil vs. non-fossil origin of carbonaceous aerosols and their SOA precursors at several



604 locations (Ceburnis et al., 2011; Hayes et al., 2013; Zotter et al., 2014). We tested the potential
605 effect of their emissions on the interpretation of isotopic measurements of ambient SOA, and our
606 isotopic analysis of 12 products demonstrates that their VOC and I/SVOC emissions and thus SOA
607 will be depleted in Carbon-14 (Table 1). 8 of the 12 contained 97% or more fossil carbon, while
608 the remaining 4 contained 57-81% fossil carbon. Asphalt-related sealants, solvents (e.g. paint
609 thinner, naphtha), and pesticides were found to have the highest fossil carbon content. This is
610 consistent with the fact that petrochemical feedstocks derived from petroleum and other fossil fuels
611 are used in the products, materials, and processes discussed throughout this work.

612

613 Studies in greater Los Angeles (Pasadena) report that fossil-fuel driven emissions appear to
614 contribute 68%-74% of the observed afternoon increase in SOA formed from urban sources with
615 the remaining ~25% coming from non-fossil sources, principally regional biogenic and local
616 cooking sources (Hayes et al., 2013, 2015; Zotter et al., 2014). The observed fossil SOA and its
617 potential precursor sources have been analyzed across several studies with differing conclusions
618 (Bahreini et al., 2012; Gentner et al., 2017; Hayes et al., 2013; Zotter et al., 2014). The studies
619 agree that on-road diesel vehicles are a relatively minor contributor to fossil SOA (Bahreini et al.,
620 2012; Hayes et al., 2013, 2015; Zotter et al., 2014), and along with other papers, have highlighted
621 the importance of other anthropogenic sources (Ensberg et al., 2014; McDonald et al., 2015; Zhao
622 et al., 2014). Other evidence suggests a fossil source other than on-road diesel since concentrations
623 of IVOCs show only minor weekday vs. weekend variation with changes in diesel traffic (Zhao et
624 al., 2014). Based on our results and the evidence in the literature, we conclude that fossil-derived
625 urban SOA precursors are emitted from product/process-related sources, and are responsible for
626 some of the fossil SOA observed in the SoCAB. The isotopic signature of products, materials, and



627 processes is due to their petrochemical feedstocks, and this has led to their misattribution to
628 combustion-related sources in the past.

629

630 **5. Conclusions and future research needs**

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

640

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



649

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

658

659 ***Validation of existing emissions inventories:*** Our results based on existing emissions inventories
660 are subject to uncertainties in their methodology. Given the results, further research is needed to
661 review, evaluate, and validate both the emissions factors and source profiles in emissions
662 inventories. A lifecycle approach should include lifetime emissions over the three pathways in
663 Section 2. Such emissions will be seasonally-dependent on factors such as air velocity, relative
664 humidity, and temperature (Wolkoff, 1998). A large survey of products and materials with
665 attention to national/regional differences is warranted to ensure that source profiles accurately
666 represent product/process-related emissions. Single-ring aromatics are a key example; the current
667 CARB inventory using the SPECIATE source profiles reports large emissions of single-ring
668 aromatics, 72% of which comes from paints. Yet, this is in contrast to our MSDS survey that
669 observed little aromatic content in current-day U.S. paints (Table 2).

670



671 ***Inclusion of IVOCs and SVOCs in emissions inventories:*** There is a clear need for more detailed
672 emissions inventories of I/SVOCs from all sources. Further research should support the evaluation
673 of the I/SVOC exemption in strategic air quality management plans and composition reporting
674 requirements in MSDSs and similar databases. More detailed speciation is required to accurately
675 determine the SOA (and ozone) formation potential of unresolved I/SVOC mixtures. Emerging
676 measurement methods will enable a more robust update of the currently unresolved I/SVOCs in
677 existing source profiles, which sometimes stem from outdated survey data (California Air
678 Resources Board, 2000b). In the process, attention should be paid to heterogeneity in products
679 containing petroleum distillates and similar components, which are very broad and can result in an
680 equally broad range of emissions.

681

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

689

690 Understanding the application method and environment is key to determining air emissions of
691 I/SVOCs since some products may be used with water and disposed of into wastewater, described
692 as *down-the-drain factors*. For example, a recent ozone modeling study modeled the fate of 23



693 oxygenated I/SVOCs (e.g. glycols, glycol ethers, esters, alcohols) present in cleaning and personal
694 products that are used with water found that most of the compounds that go down-the-drain do not
695 volatilize and are biodegraded at wastewater treatment plants, while >90% of compounds that
696 volatilize outdoors will react with OH and contribute to ozone formation (Shin et al., 2015).

697

698 ***Quantifying outdoor transport of indoor emissions:*** Similar to *down-the-drain* factors, we
699 highlight the need for a similar factor to determine fractions of indoor emissions that are
700 permanently lost to indoor sinks via chemical or physical deposition (*fraction lost to indoor sinks*).
701 Similarly, models will need factors to account for the increase in characteristic emission timescales
702 when products/materials used on indoor surfaces remain indoors longer due to generally lower
703 vertical transport coefficients and subsequent re-partitioning other surfaces prior to transport
704 outdoors.

705

706 ***Inter-location variability in the developed and developing world:*** While we present annual trends
707 for our case study megacity Los Angeles, a similar situation is evolving elsewhere in developed
708 urban areas where emissions of anthropogenic organics are key drivers of SOA and ozone
709 formation, especially in megacities. We expect substantial heterogeneity between locations in the
710 composition of emissions from products, materials, and processes due to national/regional
711 regulations governing formulation, as well as climate, application specifics, and consumer
712 preferences and options. In particular for this work, California's advanced regulatory program may
713 have led to the phase out of some components in U.S. products. So globally the composition and
714 magnitude of product/process-related emissions may contain a much greater fraction of reactive



715 species, such as single-ring aromatics. Top-down ambient studies and bottom-up studies for other
716 locations are needed to confirm the importance of product/process-related emissions, and air
717 quality modeling studies should support these efforts. The situation in urban areas of developing
718 regions and emerging economies is uncertain since they have motor vehicle emissions
719 standards/technologies that may be much more advanced than the rest of their air quality
720 management plans (Kodjak, 2015). In such locations, non-combustion sources may be important
721 sources, but the combustion of fossil fuels or biomass for home heating/cooking and agricultural
722 waste disposal will play a larger role than in developed regions. In all locations, the magnitude of
723 regional biogenic emissions and SO₂/NO_x emissions will affect the impact of anthropogenic
724 organics on SOA.

725

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

732

733 **Acknowledgements**

734 For the agricultural pesticide analysis, we would like to thank John Karlik (U. California
735 Cooperative Extension Kern County) for samples, Allen Goldstein (UC Berkeley) for access to
736 analytical instrumentation, and Emily Barnes (Yale) for help with preliminary analysis. We also



737 thank Jonathan Williams (Max Planck Institute for Chemistry) for his feedback on the
738 manuscript and the reviewers whose comments helped improve this manuscript.

739

740 **Supporting Information.** Please see the supplemental material for additional details on relevant
741 regulations, methods, Tables S1-S9, and Figures S1-S4.

742

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

745

746 Emissions from consumer products and industrial processes received some attention in pre-2000
747 outdoor air quality research and policy, especially in the cases of toxic components, highly-reactive
748 volatile solvents that fueled rapid ozone production (e.g. alkenes), and stratospheric ozone-
749 depleting chemicals. Emissions of a select few hazardous air pollutants (HAPs) (e.g. benzene and
750 vinyl chloride) were first broadly regulated under the U.S. Clean Air Act (1970). Subsequent
751 amendments through 1990 required the U.S. Environmental Protection Agency (EPA) to regulate
752 key sources of precursors to ozone production and emissions of 189 newly-designated HAPs (now
753 “air toxics”), some of which were used in products and processes (National Research Council,
754 2004). Less well-known, the 1990 amendments also required the EPA to identify priorities and
755 guidelines to mitigate emissions from consumer and commercial products (National Research
756 Council, 2004). Several categories of paints and solvents were subsequently identified based on
757 results from paint drying and chamber experiment studies conducted during early 1970s through
758 late 1990s (Chang et al., 1997; Clausen et al., 1993, 1990, 1991; Hansen, 1974; Sparks et al., 1999;
759 Sullivan, 1975), and regulations were established for half of the product categories, but actions
760 were ultimately halted as these sources were not viewed as central to ozone or other criteria



761 pollutant mitigation at the time (National Research Council, 2004). Despite large uncertainties
762 about emissions and ambient contributions from products/processes, this strategy reflects the
763 magnitude and impact of motor vehicle emissions in 1990 (Figures 1-2); the keen focus on ozone
764 production; and the lack of knowledge on SOA formation chemistry and I/SVOCs.

765

766 U.S. state or air basin-level regulations vary with region and attainment status. California's air
767 quality policy and regulations have been the most inclusive and detailed with respect to emissions
768 from consumer products and some industrial processes. The California Air Resources Board
769 (CARB) started the Consumer Products Regulatory Program in 1991, and a similar Coatings
770 Program, to address outdoor and indoor air quality problems associated with their emissions by
771 placing product type-specific limits on VOC content, and total reactivity limits (i.e. ozone
772 potential) specifically for aerosol coatings (California Air Resources Board, 2015b). CARB is also
773 required to maintain statewide and county/basin-level emissions inventories for 72 source
774 categories, available 1975 to 2020. Within these categories, the product/process-related sources
775 include consumer products, architectural coatings, pesticides, cleaning and surface coatings, and
776 asphalt paving/roofing. Chemically-speciated emissions profiles for sources within these
777 categories are in the US EPA's SPECIATE repository, some of which are used by the European
778 Union (EU) (Pernigotti et al., 2016). In the 2015 California code of regulations report, CARB has
779 further updated regulations focusing on VOC emissions from antiperspirants and deodorants,
780 hairsprays, other consumer products (both aerosol and non-aerosol), and aerosol coating products
781 (California Air Resources Board, 2015a).

782



783 The Economic Commission of Europe employs market-based mechanisms to reduce regional
784 emissions of VOCs. The National Emissions Ceilings directive sets country-specific reduction
785 targets on organic gas emissions ranging from 10% up to 60% from 2010 to 2020 (European
786 Environment Agency, 2010). The EU has two relevant policy directives: the ‘VOC Solvents
787 Emissions Directive’ to limit industrial VOC emissions resulting from processes such as printing,
788 surface cleaning, vehicular coating, and dry cleaning; and the ‘Paints Directive’ to reduce VOC
789 content in paints and varnishes (European Commission, 2014). In China, in addition to restrictions
790 on criteria pollutants including both PM₁₀ and PM_{2.5}, the Chinese Air Pollution Control Action
791 Plan 2013 also limits VOC emissions from paints, adhesives and petrochemical industry, and
792 promotes the use of low-volatility water-based paints (Ministry of Environmental Protection,
793 2012).

794

795 **Appendix B: Calculating characteristic timescales for emissions from surface layers**

796 The timescale for emission is defined by the simple relation:

$$797 \quad \tau_{emission} = \frac{M_{applied}}{R_{emission}} \quad (1)$$

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

$$804 \quad R_{emission} = v_e A C_{sat} \quad (2)$$



$$805 \quad M_{\text{applied}} = C_{OA} d \quad (3)$$

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

$$810 \quad \tau_{\text{emission}} = \frac{C_{OA} d}{v_e A C_{\text{sat}}} = \frac{C_{OA} d}{v_e C_{\text{sat}}} \quad (4)$$

$$811 \quad \text{since: } K_{OA} = \frac{C_o}{C_{\text{sat}}} \quad (5)$$

$$812 \quad \tau_{\text{emission}} = \frac{K_{OA} d}{v_e} \quad (6)$$

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



825 Equations 7-11 to calculate v_e , r_a , and r_b are reproduced below for location/condition-specific
826 analyses.

$$827 \quad v_e = \frac{1}{r_a + r_b} \quad (7)$$

$$828 \quad r_a = \frac{1}{\kappa u^*} \ln \left(\frac{z_r}{z_0} \right) \quad (8)$$

$$829 \quad r_b = \frac{5Sc^{2/3}}{u^*} \quad (9)$$

$$830 \quad Sc = \nu / \mathcal{D} \quad (10)$$

$$831 \quad u^* = \kappa U_r \left[\ln \left(\frac{z_r}{z_0} \right) \right]^{-1} \quad (11)$$

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

836



837 **References**

- 838 Algrim, L. B. and Ziemann, P. J.: Effect of the Keto Group on Yields and Composition of
839 Organic Aerosol Formed from OH Radical-Initiated Reactions of Ketones in the Presence of NO
840 x , *J. Phys. Chem. A*, *acs.jpca.6b05839*, doi:10.1021/acs.jpca.6b05839, 2016.
- 841 Bahreini, R., Middlebrook, a. M., de Gouw, J. A., Warneke, C., Trainer, M., Brock, C. a., Stark,
842 H., Brown, S. S., Dube, W. P., Gilman, J. B., Hall, K., Holloway, J. S., Kuster, W. C., Perring,
843 a. E., Prevot, a. S. H., Schwarz, J. P., Spackman, J. R., Szidat, S., Wagner, N. L., Weber, R. J.,
844 Zotter, P. and Parrish, D. D.: Gasoline emissions dominate over diesel in formation of secondary
845 organic aerosol mass, *Geophys. Res. Lett.*, *39*(6), L06805, doi:10.1029/2011GL050718, 2012.
- 846 Batterman, S. A., Chernyak, S., Jia, C., Godwin, C. and Charles, S.: Concentrations and
847 emissions of polybrominated diphenyl ethers from U.S. houses and garages., *Environ. Sci.*
848 *Technol.*, *43*(8), 2693–2700, doi:10.1021/ES8029957, 2009.
- 849 Bishop, G. A. and Stedman, D. H.: A Decade of On-road Emissions Measurements, *Environ.*
850 *Sci. Technol.*, *42*(5), 1651–1656, doi:10.1021/es702413b, 2008.
- 851 van den Born, G. J., Bouwman, A. F., Oliver, J. G. J. and Swart, R. J.: The Emission of
852 Greenhouse Gases in the Netherlands (Report no. 222901003), The Netherlands., 1991.
- 853 Brodzik, K., Faber, J., Łomankiewicz, D. and Gołda-Kopek, A.: In-vehicle VOCs composition of
854 unconditioned, newly produced cars, *J. Environ. Sci.*, *26*(5), 1052–1061, doi:10.1016/S1001-
855 0742(13)60459-3, 2014.
- 856 Bruns, E. A., Slowik, J. G., El Haddad, I., Kilic, D., Klein, F., Dommen, J., Temime-Roussel, B.,
857 Marchand, N., Baltensperger, U. and Prévôt, A. S. H.: Characterization of gas-phase organics



858 using proton transfer reaction time-of-flight mass spectrometry: fresh and aged residential wood
859 combustion emissions, Atmos. Chem. Phys., 17(1), 705–720, doi:10.5194/acp-17-705-2017,
860 2017.

861 California Air Resources Board: Aerosol Coatings & Consumer Products Methodology, Sect. 6.1
862 Consum. Prod. [online] Available from:
863 <https://www.arb.ca.gov/ei/areasrc/arbsolevapaercoatcp.htm> (Accessed 24 February 2017a), 2000.

864 California Air Resources Board: Section 6.1 Consumer Products (Revised April 2000)., 2000b.

865 California Air Resources Board: EMFAC2014 Web Database (v1.0.7), 2014.

866 California Air Resources Board: Consumer Products Program Regulations, [online] Available
867 from: <https://www.arb.ca.gov/consprod/regs/regs.htm> (Accessed 23 February 2017a), 2015.

868 California Air Resources Board: Regulation for reducing emissions from consumer products,
869 Sacramento., 2015b.

870 California Air Resources Board: Off-Road Mobile Sources Emission Reduction Program,
871 [online] Available from: <https://www.arb.ca.gov/msprog/offroad/offroad.htm> (Accessed 15
872 August 2017), 2017.

873 California Air Resources Board and Sonoma Technology Inc.: Attachment C: Asphalt Paving
874 and Roofing, CCOS II Emiss. Invent. Proj. [online] Available from:
875 <https://www.arb.ca.gov/ei/areasrc/ccosmethods.htm> (Accessed 23 February 2017), 2003.

876 Carter, W. P. L.: SAPRC Atmospheric Chemical Mechanisms and VOC Reactivity Scales,
877 [online] Available from: <http://www.engr.ucr.edu/~carter/SAPRC/>, 2007.

878 Caserini, S., Fraccaroli, A., Monguzzi, A. M., Moretti, M., Giudici, A. and Gurrieri, G. L.: A



879 detailed Emission Inventory for air quality planning at local scale: the Lombardy (Italy)
880 experience, in 13th International Emission Inventory Conference “Working for Clean Air in
881 Clearwater,” Clearwater, FL., 2004.

882 Cavallari, J. M., Osborn, L. V., Snawder, J. E., Kriech, A. J., Olsen, L. D., Herrick, R. F. and
883 Mcclean, M. D.: Predictors of Airborne Exposures to Polycyclic Aromatic Compounds and Total
884 Organic Matter among Hot-Mix Asphalt Paving Workers and Influence of Work Conditions and
885 Practices, *Ann. Occup. Hyg.*, 56(2), 138–147, doi:10.1093/annhyg/mer088, 2012a.

886 Cavallari, J. M., Zwack, L. M., Lange, C. R., Herrick, R. F. and Mcclean, M. D.: Temperature-
887 Dependent Emission Concentrations of Polycyclic Aromatic Hydrocarbons in Paving and Built-
888 Up Roofing Asphalts, *Ann. Occup. Hyg.*, 56(2), 148–160, doi:10.1093/annhyg/mer107, 2012b.

889 Ceburnis, D., Garbaras, A., Szidat, S., Rinaldi, M., Fahrni, S., Perron, N., Wacker, L., Leinert,
890 S., Remeikis, V., Facchini, M. C., Prevot, A. S. H., Jennings, S. G., Ramonet, M. and O’Dowd,
891 C. D.: Quantification of the carbonaceous matter origin in submicron marine aerosol by ¹³C and
892 ¹⁴C isotope analysis, *Atmos. Chem. Phys.*, 11(16), 8593–8606, doi:10.5194/acp-11-8593-2011,
893 2011.

894 Censullo, A. C., Jones, D. R. and Wills, M. T.: Investigation of Low Reactivity Solvents- Final
895 Report to California Air Resources Board Research Division on Contract No. 98-310., 2002.

896 Chacon-Madrid, H. J., Presto, A. A., Donahue, N. M., Lipari, F., Dasch, J. M., Scruggs, W. F.,
897 Schauer, J. J., Kleeman, M. J., Cass, G. R., Simoneit, B. R. T., Schauer, J. J., Kleeman, M. J.,
898 Cass, G. R., Simoneit, B. R. T., Cicciolia, P., Brancaleonia, E., Frattonia, M., Cecinatoa, A.,
899 Brachettia, A., Carlier, P., Hannachi, H., Mouvier, G., Calogirou, A., Larsen, B. R., Kotzias, D.,
900 Vandenberk, S., Peeters, J., Cleary, P. A., Wooldridge, P. J., Millet, D. B., McKay, M.,



- 901 Goldstein, A. H., Cohen, R. C., Laden, F., Schwartz, J., Speizer, F. E., Dockery, D. W., Kroll, J.
902 H., Ng, N. L., Murphy, S. M., Flagan, R. C., Seinfeld, J. H., Donahue, N. M., Hartz, K. E. H.,
903 Chuong, B., Presto, A. A., Stanier, C. O., Rosenhørn, T., Robinson, A. L., Pandis, S. N.,
904 Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Kroll, J. H., Smith, J. D., Che, D. L.,
905 Kessler, S. H., Worsnop, D. R., Wilson, K. R., Atkinson, R., Arey, J., Tyndall, G. S., Cox, R. A.,
906 Granier, C., Lesclaux, R., Moortgat, G. K., Pilling, M. J., Ravishankara, A. R., Wallington, T. J.,
907 Arey, J., Aschmann, S. M., Kwok, E. S. C., Atkinson, R., Cassanelli, P., Fox, D. J., Cox, R. A.,
908 Lim, Y. B., Ziemann, P. J., Lim, Y. B., Ziemann, P. J., Kwok, E. S. C., Arey, J., Atkinson, R.,
909 Atkinson, R., Kwok, E. S. C., Atkinson, R., Jenkin, M. E., Shallcross, D. E., Harvey, J. N.,
910 Roger, A., Hildebrandt, L., Donahue, N. M., Pandis, S. N., Odum, J. R., Hoffmann, T., Bowman,
911 F., Collins, D., Flagan, R. C., Seinfeld, J. H., et al.: Functionalization vs. fragmentation: n-
912 aldehyde oxidation mechanisms and secondary organic aerosol formation, *Phys. Chem. Chem.*
913 *Phys.*, 12(42), 13975, doi:10.1039/c0cp00200c, 2010.
- 914 Chan, A. W. H., Chan, M. N., Surratt, J. D., Chhabra, P. S., Loza, C. L., Crouse, J. D., Yee, L.
915 D., Flagan, R. C., Wennberg, P. O. and Seinfeld, J. H.: Role of aldehyde chemistry and NO_x
916 concentrations in secondary organic aerosol formation, *Atmos. Chem. Phys.*, 10(15), 7169–7188,
917 doi:10.5194/acp-10-7169-2010, 2010.
- 918 Chang, J. C. S., Tichenor, B. A., Guo, Z. and Krebs, K. A.: Substrate Effects on VOC Emissions
919 from a Latex Paint, *Indoor Air*, 7(4), 241–247, doi:10.1111/j.1600-0668.1997.00003.x, 1997.
- 920 Chang, Y.-M., Hu, W.-H., Fang, W.-B., Chen, S.-S., Chang, C.-T. and Ching, H.-W.: A study on
921 dynamic volatile organic compound emission characterization of water-based paints., *J. Air*
922 *Waste Manag. Assoc.*, 61(1), 35–45, 2011.



- 923 Chen, C. H., Li, L., Huang, C., Chen, Z., Wang, H. L., Huang, H. Y. and Al., E.: Study on the air
924 pollution control countermeasures for 2010 EXPO, Shanghai Acad. Environ. Sci., 2009.
- 925 Cherin, N., Roustan, Y., Musson-Genon, L. and Seigneur, C.: Modelling atmospheric dry
926 deposition in urban areas using an urban canopy approach, *Geosci. Model Dev*, 8, 893–910,
927 doi:10.5194/gmd-8-893-2015, 2015.
- 928 Clausen, P., Laursen, B., Wolkoff, P., Rasmusen, E. and Nielsen, P.: Emission of Volatile
929 Organic Compounds from a Vinyl Floor Covering, in *Modeling of Indoor Air Quality and*
930 *Exposure*, edited by N. L. Nagda, pp. 3-3–11, ASTM International, Philadelphia., 1993.
- 931 Clausen, P. A., Wolkoff, P. and Nielsen, P. A.: Long-term environmental chamber tests of
932 waterborne paints (in Danish) (SBI report 207), Horsholm, Denmark., 1990.
- 933 Clausen, P. A., Wolkoff, P., Hoist, E. and Nielsen, P. A.: Long-term Emission of Volatile
934 Organic Compounds from Waterborne Paints - Methods of Comparison, *Indoor Air*, 1(4), 562–
935 576, doi:10.1111/j.1600-0668.1991.00019.x, 1991.
- 936 Clausen, P. A., Hansen, V., Gunnarsen, L., Afshari, A. and Wolkoff, P.: Emission of di-2-
937 ethylhexyl phthalate from PVC flooring into air and uptake in dust: emission and sorption
938 experiments in FLEC and CLIMPAQ., *Environ. Sci. Technol.*, 38(9), 2531–2537, 2004.
- 939 Cohen, A. J., Ross Anderson, H., Ostro, B., Pandey, K. D., Krzyzanowski, M., Künzli, N.,
940 Gutschmidt, K., Pope, A., Romieu, I., Samet, J. M. and others: The global burden of disease due
941 to outdoor air pollution, *J. Toxicol. Environ. Heal. Part A*, 68(13–14), 1301–1307, 2005.
- 942 Cox, P., Delao, A. and Komorniczak, A.: *The California Almanac of Emissions and Air Quality -*
943 *2013 Edition*, [online] Available from:



- 944 <https://www.arb.ca.gov/aqd/almanac/almanac13/almanac13.htm> (Accessed 1 August 2016),
945 2013.
- 946 Daumit, K. E., Carrasquillo, A. J., Sugrue, R. A. and Kroll, J. H.: Effects of Condensed-Phase
947 Oxidants on Secondary Organic Aerosol Formation, *J. Phys. Chem. A*, 120(9), 1386–1394,
948 doi:10.1021/acs.jpca.5b06160, 2016.
- 949 Deguillaume, L., Beekmann, M. and Derognat, C.: Uncertainty evaluation of ozone production
950 and its sensitivity to emission changes over the Ile-de-France region during summer periods, *J.*
951 *Geophys. Res.*, 113(D2), D02304, doi:10.1029/2007JD009081, 2008.
- 952 Destailats, H., Lunden, M. M., Singer, B. C., Coleman, B. K., Hodgson, A. T., Weschler, C. J.
953 and Nazaroff, W. W.: Indoor Secondary Pollutants from Household Product Emissions in the
954 Presence of Ozone: A Bench-Scale Chamber Study, *Environ. Sci. Technol.*, 40, 4421–4428,
955 doi:10.1021/ES052198Z, 2006.
- 956 District, I. C. A. P. C.: 2012 Area Source Emissions Inventory Methodology 540 – Asphalt
957 Paving., 2012.
- 958 Dzepina, K., Volkamer, R. M., Madronich, S., Tulet, P., Ulbrich, I. M., Zhang, Q., Cappa, C. D.,
959 Ziemann, P. J. and Jimenez, J. L.: Evaluation of recently-proposed secondary organic aerosol
960 models for a case study in Mexico City, *Atmos. Chem. Phys.*, 9(15), 5681–5709,
961 doi:10.5194/acp-9-5681-2009, 2009.
- 962 Ensberg, J. J., Hayes, P. L., Jimenez, J. L., Gilman, J. B., Kuster, W. C., de Gouw, J. A.,
963 Holloway, J. S., Gordon, T. D., Jathar, S., Robinson, A. L. and Seinfeld, J. H.: Emission factor
964 ratios, SOA mass yields, and the impact of vehicular emissions on SOA formation, *Atmos.*
965 *Chem. Phys.*, 14(5), 2383–2397, doi:10.5194/acp-14-2383-2014, 2014.



- 966 European Commission: VOC Solvents Emissions Directive - Environment - European
967 Commission, [online] Available from:
968 <http://ec.europa.eu/environment/archives/air/stationary/solvents/legislation.htm> (Accessed 17
969 February 2017), 2014.
- 970 European Environment Agency: Emissions of ozone precursors, Copenhagen., 2010.
- 971 Faber, J., Brodzik, K., Gołda-Kopek, A. and Łomankiewicz, D.: Benzene, toluene and xylenes
972 levels in new and used vehicles of the same model, *J. Environ. Sci.*, 25(11), 2324–2330,
973 doi:10.1016/S1001-0742(12)60333-7, 2013.
- 974 Fortin, T. J., Howard, B. J., Parrish, D. D., Goldan, P. D., Kuster, W. C., Atlas, E. L. and Harley,
975 R. A.: Temporal changes in U.S. benzene emissions inferred from atmospheric measurements.,
976 *Environ. Sci. Technol.*, 39(6), 1403–1408, doi:10.1021/ES049316N, 2005.
- 977 Gasthauer, E., Mazé, M., Marchand, J. P. and Amouroux, J.: Characterization of asphalt fume
978 composition by GC/MS and effect of temperature, *Fuel*, 87(7), 1428–1434,
979 doi:10.1016/j.fuel.2007.06.025, 2008.
- 980 GEIA: Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) - The
981 Global Emissions Initiative (GEIA) Database, [online] Available from:
982 http://eccad.sedoo.fr/eccad_extract_interface/JSF/page_login.jsf (Accessed 24 February 2017),
983 2017.
- 984 Gentner, D. R., Isaacman, G., Worton, D. R., Chan, A. W. H., Dallmann, T. R., Davis, L., Liu,
985 S., Day, D. A., Russell, L. M., Wilson, K. R., Weber, R., Guha, A., Harley, R. A. and Goldstein,
986 A. H.: Elucidating secondary organic aerosol from diesel and gasoline vehicles through detailed
987 characterization of organic carbon emissions, *Proc. Natl. Acad. Sci. U. S. A.*, 109(45), 18318–



- 988 18323, doi:10.1073/pnas.1212272109, 2012.
- 989 Gentner, D. R., Worton, D. R., Isaacman, G., Davis, L. C., Dallmann, T. R., Wood, E. C.,
990 Herndon, S. C., Goldstein, A. H. and Harley, R. A.: Chemical composition of gas-phase organic
991 carbon emissions from motor vehicles and implications for ozone production, Environ. Sci.
992 Technol., 47(20), 11837–11848, doi:10.1021/es401470e, 2013.
- 993 Gentner, D. R., Jathar, S. H., Gordon, T. D., Bahreini, R., Day, D. A., El Haddad, I., Hayes, P.
994 L., Pieber, S. M., Platt, S. M., de Gouw, J., Goldstein, A. H., Harley, R. A., Jimenez, J. L.,
995 Prévôt, A. S. H. and Robinson, A. L.: Review of Urban Secondary Organic Aerosol Formation
996 from Gasoline and Diesel Motor Vehicle Emissions, Environ. Sci. Technol., 51(3), 1074–1093,
997 doi:10.1021/acs.est.6b04509, 2017.
- 998 Giannouli, M., Kalognomou, E.-A., Mellios, G., Moussiopoulos, N., Samaras, Z. and Fiala, J.:
999 Impact of European emission control strategies on urban and local air quality, Atmos. Environ.,
1000 45(27), 4753–4762, doi:10.1016/j.atmosenv.2010.03.016, 2011.
- 1001 Gold, M. D., Blum, A. and Ames, B. N.: Another flame retardant, tris-(1,3-dichloro-2-propyl)-
1002 phosphate, and its expected metabolites are mutagens., Science (80-.), 200(4343), 785–787,
1003 1978.
- 1004 Goldstein, A. H. and Galbally, I. E.: Known and Unexplored Organic Constituents in the Earth's
1005 Atmosphere, Environ. Sci. Technol., 41(5), 1514–1521, doi:10.1021/es072476p, 2007.
- 1006 Gordon, T. D., Tkacik, D. S., Presto, A. A., Zhang, M., Jathar, S. H., Nguyen, N. T., Massetti, J.,
1007 Truong, T., Cicero-Fernandez, P., Maddox, C., Rieger, P., Chattopadhyay, S., Maldonado, H.,
1008 Maricq, M. M. and Robinson, A. L.: Primary gas- and particle-phase emissions and secondary
1009 organic aerosol production from gasoline and diesel off-road engines, Environ. Sci. Technol.,



- 1010 47(24), 14137–14146, doi:10.1021/es403556e, 2013.
- 1011 de Gouw, J. A., Middlebrook, A. M., Warneke, C., Ahmadov, R., Atlas, E. L., Bahreini, R.,
1012 Blake, D. R., Brock, C. A., Brioude, J., Fahey, D. W., Fehsenfeld, F. C., Holloway, J. S., Le
1013 Henaff, M., Lueb, R. A., McKeen, S. A., Meagher, J. F., Murphy, D. M., Paris, C., Parrish, D.
1014 D., Perring, A. E., Pollack, I. B., Ravishankara, A. R., Robinson, A. L., Ryerson, T. B., Schwarz,
1015 J. P., Spackman, J. R., Srinivasan, A. and Watts, L. A.: Organic aerosol formation downwind
1016 from the Deepwater Horizon oil spill., *Science*, 331(6022), 1295–9,
1017 doi:10.1126/science.1200320, 2011.
- 1018 de Gouw, J. A., Gilman, J. B., Borbon, A., Warneke, C., Kuster, W. C., Goldan, P. D., Holloway,
1019 J. S., Peischl, J., Ryerson, T. B., Parrish, D. D., Gentner, D. R., Goldstein, A. H. and Harley, R.
1020 A.: Increasing atmospheric burden of ethanol in the United States, *Geophys. Res. Lett.*, 39(15),
1021 L15803, doi:10.1029/2012GL052109, 2012.
- 1022 Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen,
1023 J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T.,
1024 Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W.,
1025 McFiggans, G., Mentel, T. F., Monod, A., Prevot, A. S. H., Seinfeld, J. H., Surratt, J. D.,
1026 Szmigielski, R. and Wildt, J.: The formation, properties and impact of secondary organic aerosol:
1027 current and emerging issues, *Atmos. Chem. Phys.*, 9, 5155–5236, 2009.
- 1028 Hansen, C. M.: The Air Drying of Latex Coatings, *Ind. Eng. Chem. Prod. Res. Dev.*, 13(2), 150–
1029 152, doi:10.1021/i360050a015, 1974.
- 1030 Hao, J., Wang, L., Shen, M., Li, L. and Hu, J.: Air quality impacts of power plant emissions in
1031 Beijing, *Environ. Pollut.*, 147, 401–408, doi:10.1016/j.envpol.2006.06.013, 2007.



- 1032 Hayes, P. L., Ortega, A. M., Cubison, M. J., Froyd, K. D., Zhao, Y., Cliff, S. S., Hu, W. W.,
1033 Toohey, D. W., Flynn, J. H., Lefer, B. L., Grossberg, N., Alvarez, S., Rappenglück, B., Taylor, J.
1034 W., Allan, J. D., Holloway, J. S., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Massoli, P.,
1035 Zhang, X., Liu, J., Weber, R. J., Corrigan, A. L., Russell, L. M., Isaacman, G., Worton, D. R.,
1036 Kreisberg, N. M., Goldstein, A. H., Thalman, R., Waxman, E. M., Volkamer, R., Lin, Y. H.,
1037 Surratt, J. D., Kleindienst, T. E., Offenberg, J. H., Dusanter, S., Griffith, S., Stevens, P. S.,
1038 Brioude, J., Angevine, W. M. and Jimenez, J. L.: Organic aerosol composition and sources in
1039 Pasadena, California, during the 2010 CalNex campaign, *J. Geophys. Res. Atmos.*, 118, 9233–
1040 9257, doi:10.1002/jgrd.50530, 2013.
- 1041 Hayes, P. L., Carlton, A. G., Baker, K. R., Ahmadov, R., Washenfelder, R. A., Alvarez, S.,
1042 Rappenglück, B., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Zotter, P., Prévôt, A. S. H.,
1043 Szidat, S., Kleindienst, T. E., Offenberg, J. H., Ma, P. K. and Jimenez, J. L.: Modeling the
1044 formation and aging of secondary organic aerosols in Los Angeles during CalNex 2010, *Atmos.*
1045 *Chem. Phys.*, 15(10), 5773–5801, doi:10.5194/acp-15-5773-2015, 2015.
- 1046 Isman, M. B.: Plant essential oils for pest and disease management, *Crop Prot.*, 19(8–10), 603–
1047 608, doi:10.1016/S0261-2194(00)00079-X, 2000.
- 1048 Jathar, S. H., Gordon, T. D., Hennigan, C. J., Pye, H. O. T., Pouliot, G., Adams, P. J., Donahue,
1049 N. M. and Robinson, A. L.: Unspeciated organic emissions from combustion sources and their
1050 influence on the secondary organic aerosol budget in the United States., *Proc. Natl. Acad. Sci. U.*
1051 *S. A.*, 111(29), 10473–10478, doi:10.1073/pnas.1323740111, 2014.
- 1052 Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E. and
1053 Thun, M.: Long-term ozone exposure and mortality, *N. Engl. J. Med.*, 360(11), 1085–1095,



- 1054 2009.
- 1055 Jia, L. and Xu, Y.: Effects of Relative Humidity on Ozone and Secondary Organic Aerosol
1056 Formation from the Photooxidation of Benzene and Ethylbenzene, *Aerosol Sci. Technol.*, 48(1),
1057 1–12, doi:10.1080/02786826.2013.847269, 2014.
- 1058 Kemmlein, S., Hahn, O. and Jann, O.: Emissions of organophosphate and brominated flame
1059 retardants from selected consumer products and building materials, *Atmos. Environ.*, 37(39),
1060 5485–5493, doi:10.1016/j.atmosenv.2003.09.025, 2003.
- 1061 Kirchstetter, T. W., Singer, B. C., Harley, R. A., Kendall, G. R. and Traverse, M.: Impact of
1062 California Reformulated Gasoline on Motor Vehicle Emissions. 1. Mass Emission Rates,
1063 *Environ. Sci. Technol.*, 33(2), 318–328, doi:10.1021/es9803714, 1999.
- 1064 Kitto, A. M., Pirbazari, M., Badriyha, B. N., Ravindran, V., Tyner, R. and Synolakis, C. E.:
1065 Emissions of Volatile and Semi-Volatile Organic Compounds and Particulate Matter from Hot
1066 Asphalts, *Environ. Technol.*, 18(2), 121–138, doi:10.1080/09593331808616520, 1997.
- 1067 Klein, F., Platt, S. M., Farren, N. J., Detournay, A., Bruns, E. A., Bozzetti, C., Daellenbach, K.
1068 R., Kilic, D., Kumar, N. K., Pieber, S. M., Slowik, J. G., Temime-Roussel, B., Marchand, N.,
1069 Hamilton, J. F., Baltensperger, U., Prévôt, A. S. H. and El Haddad, I.: Characterization of Gas-
1070 Phase Organics Using Proton Transfer Reaction Time-of-Flight Mass Spectrometry: Cooking
1071 Emissions, *Environ. Sci. Technol.*, 50(3), 1243–1250, doi:10.1021/acs.est.5b04618, 2016.
- 1072 Knudsen, H. N., Kjaer, U. D., Nielsen, P. A. and Wolkoff, P.: Sensory and chemical
1073 characterization of VOC emissions from building products: impact of concentration and air
1074 velocity, *Atmos. Environ.*, 33(8), 1217–1230, doi:10.1016/S1352-2310(98)00278-7, 1999.



- 1075 Kodjak, D.: Policies to Reduce Fuel Consumption, Air Pollution, and Carbon Emissions from
1076 Vehicles in G20 Nations., 2015.
- 1077 Kriech, A. J., Kurek, J. T., Wissel, H. L., Osborn, L. V. and Blackburn, G. R.: Evaluation of
1078 Worker Exposure to Asphalt Paving Fumes Using Traditional and Nontraditional Techniques,
1079 AIHA J., 63(5), 628–635, doi:10.1080/15428110208984749, 2002.
- 1080 Kroll, J. H. and Seinfeld, J. H.: Chemistry of secondary organic aerosol: Formation and evolution
1081 of low-volatility organics in the atmosphere, Atmos. Environ., 42(16), 3593–3624,
1082 doi:10.1016/j.atmosenv.2008.01.003, 2008.
- 1083 Kumar, J. and Parmar, B. S.: Physicochemical and Chemical Variation in Neem Oils and Some
1084 Bioactivity Leads against *Spodoptera litura* F, J. Agric. Food Chem., 44(8), 2137–2143, 19996.
- 1085 Kwok, E. and Atkinson, R.: Estimation of hydroxyl radical reaction rate constants for gas-phase
1086 organic compounds using a structure-reactivity relationship: An update, Atmos. Environ.,
1087 29(14), 1685–1695, doi:10.1016/1352-2310(95)00069-B, 1995.
- 1088 Lange, C., Stroup-Gardiner, M. and Cr, L.: Quantification of Potentially Odorous Volatile
1089 Organic Compounds from Asphalt Binders Using Head-Space Gas Chromatography, J. Test.
1090 Eval., 33(2), 1–9, doi:10.1520/JTE11800, 2005.
- 1091 Lange, C. R. and Stroup-Gardiner, M.: Temperature-Dependent Chemical-Specific Emission
1092 Rates of Aromatics and Polyaromatic Hydrocarbons (PAHs) in Bitumen Fume, J. Occup.
1093 Environ. Hyg., 4(September), 72–76, doi:10.1080/15459620701385279, 2007.
- 1094 Lewis, R. G.: Pesticides, in Indoor Air Quality Handbook, edited by J. D. Spengler, J. F.
1095 McCarthy, and J. M. Samet, p. 35.1-35.21, McGraw-Hill., 2001.



- 1096 Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., Amann, M.,
1097 Anderson, H. R., Andrews, K. G., Aryee, M., Atkinson, C., Bacchus, L. J., Bahalim, A. N.,
1098 Balakrishnan, K., Balmes, J., Barker-Collo, S., Baxter, A., Bell, M. L., Blore, J. D., Blyth, F.,
1099 Bonner, C., Borges, G., Bourne, R., Boussinesq, M., Brauer, M., Brooks, P., Bruce, N. G.,
1100 Brunekreef, B., Bryan-Hancock, C., Bucello, C., Buchbinder, R., Bull, F., Burnett, R. T., Byers,
1101 T. E., Calabria, B., Carapetis, J., Carnahan, E., Chafe, Z., Charlson, F., Chen, H., Chen, J. S.,
1102 Cheng, A. T.-A., Child, J. C., Cohen, A., Colson, K. E., Cowie, B. C., Darby, S., Darling, S.,
1103 Davis, A., Degenhardt, L., Dentener, F., Des Jarlais, D. C., Devries, K., Dherani, M., Ding, E. L.,
1104 Dorsey, E. R., Driscoll, T., Edmond, K., Ali, S. E., Engell, R. E., Erwin, P. J., Fahimi, S., Falder,
1105 G., Farzadfar, F., Ferrari, A., Finucane, M. M., Flaxman, S., Fowkes, F. G. R., Freedman, G.,
1106 Freeman, M. K., Gakidou, E., Ghosh, S., Giovannucci, E., Gmel, G., Graham, K., Grainger, R.,
1107 Grant, B., Gunnell, D., Gutierrez, H. R., Hall, W., Hoek, H. W., Hogan, A., Hosgood, H. D.,
1108 Hoy, D., Hu, H., Hubbell, B. J., Hutchings, S. J., Ibeanusi, S. E., Jacklyn, G. L., Jasrasaria, R.,
1109 Jonas, J. B., Kan, H., Kanis, J. A., Kassebaum, N., Kawakami, N., Khang, Y.-H., Khatibzadeh,
1110 S., Khoo, J.-P., Kok, C., et al.: A comparative risk assessment of burden of disease and injury
1111 attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: a systematic
1112 analysis for the Global Burden of Disease Study 2010., *Lancet*, 380(9859), 2224–2260,
1113 doi:10.1016/S0140-6736(12)61766-8, 2012.
- 1114 Lu, S. H., Liu, Y., Shao, M. and Huang, S.: Chemical speciation and anthropogenic sources of
1115 ambient volatile organic compounds (VOCs) during summer in Beijing, *Front. Environ. Sci.*
1116 *Eng. China*, 1(2), 147–152, 2007.
- 1117 Ma, P. K., Zhao, Y., Robinson, A. L., Worton, D. R., Goldstein, A. H., Ortega, A. M., Jimenez,
1118 J.-L., Zotter, P., Prévôt, A. S. H., Szidat, S. and Hayes, P. L.: Evaluating the impact of new



- 1119 observational constraints on P-S/IVOC emissions, multi-generation oxidation, and chamber wall
1120 losses on SOA modeling for Los Angeles, CA, Atmos. Chem. Phys. Discuss., 1–35,
1121 doi:10.5194/acp-2016-957, 2016.
- 1122 Markakis, K., Im, U., Unal, A., Melas, D., Yenigun, O. and Incecik, S.: A computational
1123 approach for the compilation of a high spatially and temporally resolved emission inventory for
1124 the Istanbul Greater Area, in Paper presented at the 7th International Conference of Air Quality
1125 Science and Application, Istanbul., 2009.
- 1126 May, A. A., Nguyen, N. T., Presto, A. A., Gordon, T. D., Lipsky, E. M., Karve, M., Gutierrez,
1127 A., Robertson, W. H., Zhang, M., Brandow, C., Chang, O., Chen, S., Cicero-Fernandez, P.,
1128 Dinkins, L., Fuentes, M., Huang, S.-M., Ling, R., Long, J., Maddox, C., Massetti, J., McCauley,
1129 E., Miguel, A., Na, K., Ong, R., Pang, Y., Rieger, P., Sax, T., Truong, T., Vo, T., Chattopadhyay,
1130 S., Maldonado, H., Maricq, M. M. and Robinson, A. L.: Gas- and particle-phase primary
1131 emissions from in-use, on-road gasoline and diesel vehicles, Atmos. Environ., 88, 247–260,
1132 doi:10.1016/j.atmosenv.2014.01.046, 2014.
- 1133 McDonald, B. C., Gentner, D. R., Goldstein, A. H. and Harley, R. A.: Long-term trends in motor
1134 vehicle emissions in U.S. urban areas, Environ. Sci. Technol., 47(17), 10022–10031,
1135 doi:10.1021/es401034z, 2013.
- 1136 McDonald, B. C., Goldstein, A. H. and Harley, R. A.: Long-term trends in california mobile
1137 source emissions and ambient concentrations of black carbon and organic aerosol., Environ. Sci.
1138 Technol., 49(8), 5178–88, doi:10.1021/es505912b, 2015.
- 1139 Menut, L.: Adjoint modeling for atmospheric pollution process sensitivity at regional scale, J.
1140 Geophys. Res., 108(D17), 8562, doi:10.1029/2002JD002549, 2003.



- 1141 Miller, J. D. and Facanha, C.: The State of Clean Transport Policy - A 2014 Synthesis of Vehicle
1142 and Fuel Policy developments., 2014.
- 1143 Ministry of Environmental Protection: Ambient air quality standards (GB 3095-2012), 2012.
- 1144 Mitro, S. D., Dodson, R. E., Singla, V., Adamkiewicz, G., Elmi, A. F., Tilly, M. K. and Zota, A.
1145 R.: Consumer Product Chemicals in Indoor Dust: A Quantitative Meta-analysis of U.S. Studies,
1146 Environ. Sci. Technol., 50(19), 10661–10672, doi:10.1021/acs.est.6b02023, 2016.
- 1147 Murphy, B. N., Donahue, N. M., Robinson, A. L. and Pandis, S. N.: A naming convention for
1148 atmospheric organic aerosol, Atmos. Chem. Phys., 14(11), 5825–5839, doi:10.5194/acp-14-
1149 5825-2014, 2014.
- 1150 National Research Council: Air Quality Management in the United States, National Academies
1151 Press, Washington, D.C., 2004.
- 1152 Nazaroff, W. W. and Weschler, C. J.: Cleaning products and air fresheners: exposure to primary
1153 and secondary air pollutants, Atmos. Environ., 38(18), 2841–2865,
1154 doi:10.1016/j.atmosenv.2004.02.040, 2004.
- 1155 Neligan, R. E.: Hydrocarbons in the Los Angeles atmosphere. A comparison between the
1156 hydrocarbons in automobile exhaust and those found in the Los Angeles atmosphere., Arch.
1157 Environ. Health, 5(6), 581–591, doi:10.1080/00039896.1962.10663334, 1962.
- 1158 Ng, N. L., Kroll, J. H., Keywood, M. D., Bahreini, R., Varutbangkul, V., Flagan, R. C., Seinfeld,
1159 J. H., Lee, A. and Goldstein, A. H.: Contribution of first- versus second-generation products to
1160 secondary organic aerosols formed in the oxidation of biogenic hydrocarbons., Environ. Sci.
1161 Technol., 40(7), 2283–2297, doi:10.1021/es052269u, 2006.



- 1162 Nielsen, O.-K., Winther, M., Mikkelsen, M. H., Hoffmann, L., Nielsen, M., Gyldenkerne, S.,
1163 Fauser, P., Jensen, M. T., Plejdrup, M. S. and Illerup, J. B.: Annual Danish Emission Inventory
1164 Report to UNECE. Inventories from the base year of the protocols to year 2006., 2008.
- 1165 Pankow, J. F. and Asher, W. E.: SIMPOL.1: a simple group contribution method for predicting
1166 vapor pressures and enthalpies of vaporization of multifunctional organic compounds, Atmos.
1167 Chem. Phys., 8(10), 2773–2796, doi:10.5194/acp-8-2773-2008, 2008.
- 1168 Parker, D., Sonne, J. and Sherwin, J.: Demonstration of Cooling Savings of Light Colored Roof
1169 Surfacing in Florida Commercial Buildings: Retail Strip Mall, Cocoa, Florida., 1997.
- 1170 Pernigotti, D., Belis, C. A. and Spanò, L.: SPECIEUROPE: The European data base for PM
1171 source profiles, Atmos. Pollut. Res., 7(2), 307–314, doi:10.1016/j.apr.2015.10.007, 2016.
- 1172 Piccot, S. D., Watson, J. J. and Jones, J. W.: A global inventory of volatile organic compound
1173 emissions from anthropogenic sources, J. Geophys. Res., 97, 9897–9912,
1174 doi:10.1029/92JD00682, 1992.
- 1175 Platt, S. M., Haddad, I. El, Pieber, S. M., Huang, R.-J., Zardini, A. A., Clairotte, M., Suarez-
1176 Bertoa, R., Barmet, P., Pfaffenberger, L., Wolf, R., Slowik, J. G., Fuller, S. J., Kalberer, M.,
1177 Chirico, R., Dommen, J., Astorga, C., Zimmermann, R., Marchand, N., Hellebust, S., Temime-
1178 Roussel, B., Baltensperger, U. and Prévôt, A. S. H.: Two-stroke scooters are a dominant source
1179 of air pollution in many cities., Nat. Commun., 5, 3749, doi:10.1038/ncomms4749, 2014.
- 1180 Pomerantz, M., Pon, B., Akbari, H. and Chang, S.-C.: The Effect of Pavements' Temperatures
1181 on Air Temperatures in Large Cities, Berkeley., 2000.
- 1182 Pope, C. A. I. and Dockery, D. W.: Health effects of fine particulate air pollution: lines that



- 1183 connect, J. Air Waste Manag. Assoc., 56(6), 709–742, 2006.
- 1184 Poppendieck, D., Hubbard, H., Ward, M., Weschler, C. and Corsi, R. L.: Ozone reactions with
1185 indoor materials during building disinfection, Atmos. Environ., 41(15), 3166–3176,
1186 doi:10.1016/j.atmosenv.2006.06.060, 2007a.
- 1187 Poppendieck, D. G., Hubbard, H. F., Weschler, C. J. and Corsi, R. L.: Formation and emissions
1188 of carbonyls during and following gas-phase ozonation of indoor materials, Atmos. Environ.,
1189 41(35), 7614–7626, doi:10.1016/j.atmosenv.2007.05.049, 2007b.
- 1190 Pöschl, U. and Shiraiwa, M.: Multiphase Chemistry at the Atmosphere–Biosphere Interface
1191 Influencing Climate and Public Health in the Anthropocene, Chem. Rev., 115(10), 4440–4475,
1192 doi:10.1021/cr500487s, 2015.
- 1193 Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop,
1194 A. P., Lane, T. E., Pierce, J. R. and Pandis, S. N.: Rethinking organic aerosols: semivolatile
1195 emissions and photochemical aging., Science (80-.), 315(5816), 1259–1262,
1196 doi:10.1126/science.1133061, 2007.
- 1197 Sadezky, A., Chaimbault, P., Mellouki, A., Römpp, A., Winterhalter, R., Le Bras, G. and
1198 Moortgat, G. K.: Formation of secondary organic aerosol and oligomers from the ozonolysis of
1199 enol ethers, Atmos. Chem. Phys., 6(12), 5009–5024, doi:10.5194/acp-6-5009-2006, 2006.
- 1200 Salthammer, T. and Fuhrmann, F.: Photocatalytic Surface Reactions on Indoor Wall Paint,
1201 Environ. Sci. Technol., 41(18), 6573–6578, doi:10.1021/ES070057M, 2007.
- 1202 San Joaquin Valley Air Pollution Control District: 2008 Area Source Emissions Inventory
1203 Methodology 540 – ASPHALT PAVING., 2008.



- 1204 Sarwar, G., Olson, D. A., Corsi, R. L. and Weschler, C. J.: Indoor Fine Particles: The Role of
1205 Terpene Emissions from Consumer Products, *J. Air Waste Manage. Assoc.*, 54(3), 367–377,
1206 doi:10.1080/10473289.2004.10470910, 2004.
- 1207 von Schneidemesser, E., Coates, J., Denier van der Gon, H. A. C., Visschedijk, A. J. H. and
1208 Butler, T. M.: Variation of the NMVOC speciation in the solvent sector and the sensitivity of
1209 modelled tropospheric ozone, *Atmos. Environ.*, 135, 59–72,
1210 doi:10.1016/j.atmosenv.2016.03.057, 2016.
- 1211 Shin, H.-M., McKone, T. E. and Bennett, D. H.: Contribution of low vapor pressure-volatile
1212 organic compounds (LVP-VOCs) from consumer products to ozone formation in urban
1213 atmospheres, *Atmos. Environ.*, 108, 98–106, doi:10.1016/j.atmosenv.2015.02.067, 2015.
- 1214 Sillman, S.: The relation between ozone, NO and hydrocarbons in urban and polluted rural
1215 environments, , 33, 1999.
- 1216 Singer, B. C., Coleman, B. K., Destailats, H., Hodgson, A. T., Lunden, M. M., Weschler, C. J.
1217 and Nazaroff, W. W.: Indoor secondary pollutants from cleaning product and air freshener use in
1218 the presence of ozone, *Atmos. Environ.*, 40(35), 6696–6710,
1219 doi:10.1016/j.atmosenv.2006.06.005, 2006.
- 1220 Song, Y., Shao, M., Liu, Y., Lu, S., Kuster, W., Goldan, P. and Xie, S.: Source apportionment of
1221 ambient volatile organic compounds in Beijing, *Environ. Sci. Technol.*, 41(12), 4348–4353,
1222 2007.
- 1223 Sparks, L. E., Guo, Z., Chang, J. C. and Tichenor, B. A.: Volatile Organic Compound Emissions
1224 from Latex Paint - Part 2. Test House Studies and Indoor Air Quality (IAQ) Modeling, *Indoor*
1225 *Air*, 9(1), 18–25, doi:10.1111/j.1600-0668.1999.t01-3-00004.x, 1999.



- 1226 Sullivan, D. A.: Water and solvent evaporation from latex and latex paint films, *J. Paint*
1227 *Technol.*, 47, 60–67, 1975.
- 1228 Tessum, C. W., Hill, J. D. and Marshall, J. D.: Life cycle air quality impacts of conventional and
1229 alternative light-duty transportation in the United States, *Proc. Natl. Acad. Sci.*, 111(52), 18490–
1230 18495, doi:10.1073/pnas.1406853111, 2014.
- 1231 The Asphalt Institute: Asphalt Usage Survey for the United States and Canada., 2015.
- 1232 The Asphalt Institute & European Bitumen Association: The bitumen industry: A global
1233 perspective: Production, chemistry, use, specification and occupational exposure, Belgium.,
1234 2015.
- 1235 Toftum, J., Freund, S., Salthammer, T. and Weschler, C. J.: Secondary organic aerosols from
1236 ozone-initiated reactions with emissions from wood-based materials and a “green” paint, *Atmos.*
1237 *Environ.*, 42(33), 7632–7640, doi:10.1016/j.atmosenv.2008.05.071, 2008.
- 1238 Tsimpidi, A. P., Karydis, V. A., Zavala, M., Lei, W., Molina, L., Ulbrich, I. M., Jimenez, J. L.
1239 and Pandis, S. N.: Evaluation of the volatility basis-set approach for the simulation of organic
1240 aerosol formation in the Mexico City metropolitan area, *Atmos. Chem. Phys.*, 10(2), 525–546,
1241 doi:10.5194/acp-10-525-2010, 2010.
- 1242 U.S. Environmental Protection Agency: 2011 National Emissions Inventory (NEI) Data, 2011.
- 1243 U.S. Environmental Protection Agency: SPECIATE 4.4., 2014.
- 1244 U.S. Environmental Protection Agency and Office of Air Quality Planning and Standards:
1245 National Air Pollutant Emission Estimates 1940-1989, Research Triangle Park, North Carolina.,
1246 1991.



- 1247 U.S. EPA: Emissions Factors & AP 42, Compilation of Air Pollutant Emission Factors, Volume
1248 1: Stationary Point and Area Sources, Chapter 4: Evaporation Loss Sources., 1995.
- 1249 U.S. EPA: Regulations for Emissions from Nonroad Vehicles and Engines, [online] Available
1250 from: [https://www.epa.gov/regulations-emissions-vehicles-and-engines/regulations-emissions-](https://www.epa.gov/regulations-emissions-vehicles-and-engines/regulations-emissions-nonroad-vehicles-and-engines)
1251 [nonroad-vehicles-and-engines](https://www.epa.gov/regulations-emissions-vehicles-and-engines/regulations-emissions-nonroad-vehicles-and-engines) (Accessed 15 August 2017), 2017.
- 1252 Warneke, C., de Gouw, J. A., Holloway, J. S., Peischl, J., Ryerson, T. B., Atlas, E., Blake, D.,
1253 Trainer, M. and Parrish, D. D.: Multiyear trends in volatile organic compounds in Los Angeles,
1254 California: Five decades of decreasing emissions, *J. Geophys. Res. Atmos.*, 117(D21), D00V17,
1255 doi:10.1029/2012JD017899, 2012.
- 1256 Washenfelder, R. A., Young, C. J., Brown, S. S., Angevine, W. M., Atlas, E. L., Blake, D. R.,
1257 Bon, D. M., Cubison, M. J., de Gouw, J. A., Dusanter, S., Flynn, J., Gilman, J. B., Graus, M.,
1258 Griffith, S., Grossberg, N., Hayes, P. L., Jimenez, J. L., Kuster, W. C., Lefer, B. L., Pollack, I.
1259 B., Ryerson, T. B., Stark, H., Stevens, P. S. and Trainer, M. K.: The glyoxal budget and its
1260 contribution to organic aerosol for Los Angeles, California, during CalNex 2010, *J. Geophys.*
1261 *Res. Atmos.*, 116, D00V02, doi:10.1029/2011JD016314, 2011.
- 1262 Weiss, M., Bonnel, P., Hummel, R., Provenza, A. and Manfredi, U.: On-Road Emissions of
1263 Light-Duty Vehicles in Europe, *Environ. Sci. Technol.*, 45(19), 8575–8581,
1264 doi:10.1021/es2008424, 2011.
- 1265 Wensing, M., Uhde, E. and Salthammer, T.: Plastics additives in the indoor environment—flame
1266 retardants and plasticizers, *Sci. Total Environ.*, 339(1), 19–40,
1267 doi:10.1016/j.scitotenv.2004.10.028, 2005.
- 1268 Weschler, C. J.: Chemistry in indoor environments: 20 years of research, *Indoor Air*, 21(3), 205–



- 1269 218, doi:10.1111/j.1600-0668.2011.00713.x, 2011.
- 1270 Weschler, C. J. and Nazaroff, W. W.: Semivolatile organic compounds in indoor environments,
1271 Atmos. Environ., 42(40), 9018–9040, doi:10.1016/j.atmosenv.2008.09.052, 2008.
- 1272 Weschler, C. J. and Nazaroff, W. W.: SVOC partitioning between the gas phase and settled dust
1273 indoors, Atmos. Environ., 44(30), 3609–3620, doi:10.1016/j.atmosenv.2010.06.029, 2010.
- 1274 Wilke, O., Jann, O. and Brodner, D.: VOC- and SVOC-emissions from adhesives, floor
1275 coverings and complete floor structures, Indoor Air, 14(s8), 98–107, doi:10.1111/j.1600-
1276 0668.2004.00314.x, 2004.
- 1277 Wolkoff, P.: Impact of air velocity, temperature, humidity, and air on long-term voc emissions
1278 from building products, Atmos. Environ., 32(14–15), 2659–2668, doi:10.1016/S1352-
1279 2310(97)00402-0, 1998.
- 1280 Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-
1281 VanWertz, G., Kreisberg, N. M., Knot, C., Olson, K., Koss, A., Goldstein, A. H., Hering, S. V.,
1282 Gouw, J. de, Baumann, K., Lee, S.-H., Nenes, A., Weber, R. J. and Ng, N. L.: Effects of
1283 anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the
1284 southeastern United States., Proc. Natl. Acad. Sci. U. S. A., 112(1), 37–42,
1285 doi:10.1073/pnas.1417609112, 2015.
- 1286 Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R.,
1287 Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P.
1288 F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimo, A., Hatakeyama, S., Takegawa,
1289 N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams,
1290 P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y. M.



- 1291 and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in
1292 anthropogenically-influenced Northern Hemisphere midlatitudes, *Geophys. Res. Lett.*, 34(13),
1293 L13801, doi:10.1029/2007GL029979, 2007.
- 1294 Zhao, Y., Hennigan, C. J., May, A. A., Tkacik, D. S., de Gouw, J. A., Gilman, J. B., Kuster, W.,
1295 Borbon, A. and Robinson, A. L.: Intermediate-volatility organic compounds: a large source of
1296 secondary organic aerosol., *Environ. Sci. Technol.*, 48(23), 13743–13750,
1297 doi:10.1021/es5035188, 2014.
- 1298 Zhao, Y., Nguyen, N. T., Presto, A. A., Hennigan, C. J., May, A. A. and Robinson, A. L.:
1299 Intermediate Volatility Organic Compound Emissions from On-road Gasoline Vehicles and
1300 Small Off-road Gasoline Engines, *Environ. Sci. Technol.*, 50(8), 4554–4563, 2016.
- 1301 Zhao, Y., Saleh, R., Saliba, G., Presto, A. A., Gordon, T. D., Drozd, G. T., Goldstein, A. H.,
1302 Donahue, N. M. and Robinson, A. L.: Reducing secondary organic aerosol formation from
1303 gasoline vehicle exhaust., *Proc. Natl. Acad. Sci. U. S. A.*, 114(27), 6984–6989,
1304 doi:10.1073/pnas.1620911114, 2017.
- 1305 Zhou, Y., Levy, J. I., Hammitt, J. K. and Evans, J. S.: Estimating population exposure to power
1306 plant emissions using CALPUFF: a case study in Beijing, China, *Atmos. Environ.*, 37(6), 815–
1307 826, doi:10.1016/S1352-2310(02)00937-8, 2003.
- 1308 Zotter, P., El-Haddad, I., Zhang, Y., Hayes, P. L., Zhang, X., Lin, Y.-H., Wacker, L., Schnelle-
1309 Kreis, J., Abbaszade, G., Zimmermann, R., Surratt, J. D., Weber, R., Jimenez, J. L., Szidat, S.,
1310 Baltensperger, U. and Prévôt, A. S. H.: Diurnal cycle of fossil and nonfossil carbon using
1311 radiocarbon analyses during CalNex, *J. Geophys. Res. Atmos.*, 119(11), 6818–6835,
1312 doi:10.1002/2013JD021114, 2014.



1313 **Figure captions**

1314

1315 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle fleets. Trends in motor vehicle
1316 population, fuel use, and VOC emissions for model years conforming to pre-LEV, LEV-I, -II, and -III emissions
1317 standards in California's South Coast air basin for 1990-2020 from CARB's EMFAC database (California Air
1318 Resources Board, 2014).

1319

1320 **Figure 2.** Emissions and potential air quality impacts of product/process-related sources and motor vehicles over
1321 time in the South Coast air basin (i.e. Los Angeles). (a) Total VOC emissions (excluding ethane). (b) Potential SOA
1322 from product/process-related emissions compared to on-road motor vehicles, with uncertainties based on the ranges
1323 of compound class-specific SOA yields, and (c) Ozone formation potential via compound-specific maximum ozone
1324 incremental reactivity values (SAPRC, no uncertainty given). Product/process-related results are produced using
1325 CARB emissions data and EPA source profiles; see methods for further detail. Note: None of the panels include
1326 VOCs, IVOCs, or SVOCs, from products/processes that we identify in Sections 2 and 4 as missing from emissions
1327 inventories, but SOA and ozone formation from motor vehicle emissions does include I/SVOCs. Maximum potential
1328 SOA from gasoline vehicles in 1990 was ~41 tons day⁻¹ (off graph). Pre-1990 potential SOA and ozone for motor
1329 vehicles is excluded due to the lack of fleet-resolved data.

1330

1331 **Figure 3.** Average emissions from product/process-related sources for 2005-2020 based on data from CARB
1332 inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b)
1333 major product/process-related source categories in the inventory (values in Table S3). "Miscellaneous" contains
1334 70% mineral spirits (C₇₋₁₂ hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified
1335 in Section 2 and 4.

1336

1337 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
1338 glycol (b), and acetone (c). Based on applied layers of 0.01-1 mm and vertical transport coefficients for
1339 neutral/stable outdoor conditions (10-50 m hr⁻¹), with longer timescales in indoor environments or thicker layers



1340 (e.g. asphalt, building materials). The left axis is reproduced on the right in days. Timescales are limited by gas-
1341 phase transport from the surface, but absorption into polymeric or porous substrates could extend timescales
1342 (Weschler and Nazaroff, 2008). Volatility range boundaries for VOC-IVOC and IVOC-SVOC occur between C_{12} -
1343 C_{13} and C_{19} - C_{20} , respectively, for n-alkanes or compounds with equivalent volatilities. See Appendix B for
1344 calculations and Table S9.

1345

1346 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
1347 represent average yields of the product/process-related source categories in Figure 3, and do not include "missing"
1348 emissions. The red markers show yields of sampled consumer products calculated from speciation obtained after
1349 GC-MS analysis. All are shown as a function of mass emitted, not product composition.

1350 **FIGURES & TABLES**

1351 **Table 1.** Composition and emittable fraction of twelve commercially available consumer products, and results of
 1352 their carbon isotope analysis. Please see Table S1 for detailed mass distribution profiles and Table S7 for detailed
 1353 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 ^c	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%	-	-

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

[§]Asphalt-related products.

[†]Applied as aerosols.

^aLargely 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: Emittable fraction is confirmed with MSDS where possible.

1354

1355 **Table 2.** Analysis of material safety data sheets commercially available with products.

1356

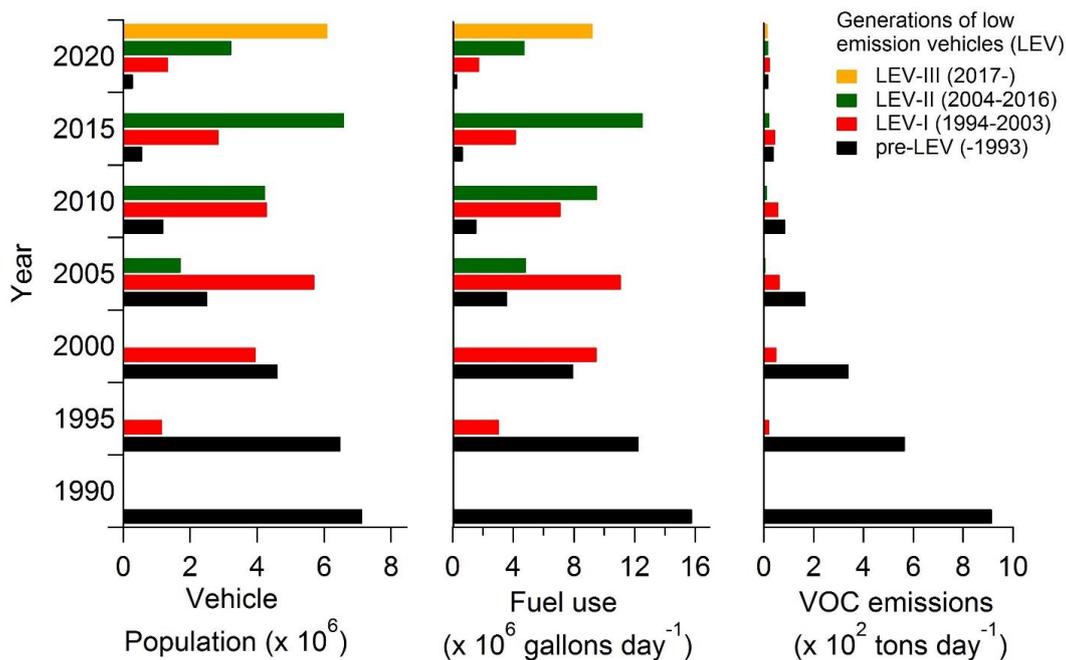
Product Category	Fraction of products with aromatic content	MSDS Aromatics %wt. Mean (range)	Fraction of products with I/SVOC content	MSDS I/SVOCs %wt. Mean (range)
Paints*	0%	0	24% ^a	2.5 (1-5)
Adhesives	27%	8 (1-30)	7% ^b	20 (10-30)
Cleaning Products	8%	5 (1-10)	23% ^c	3.5 (1-7)
Sealants	53%	9 (1-30)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

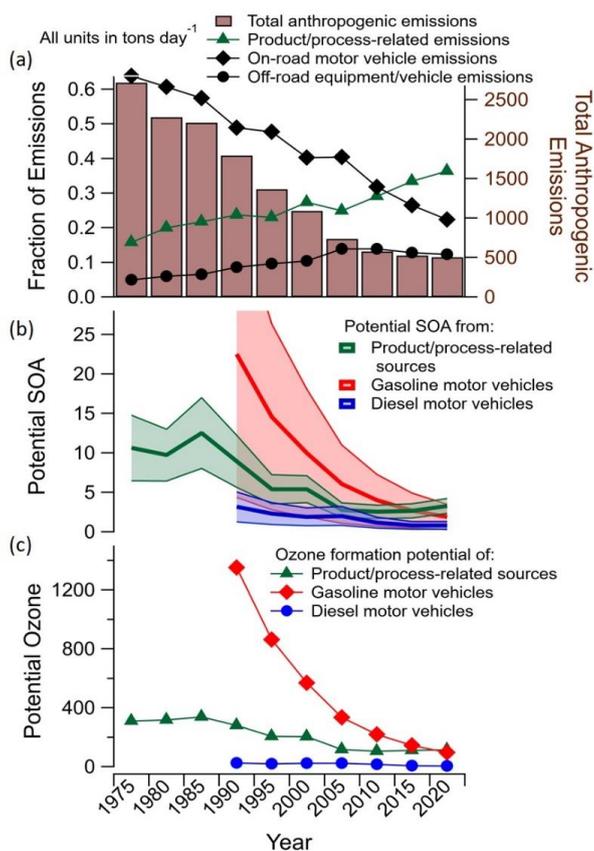
1357



1358

1359

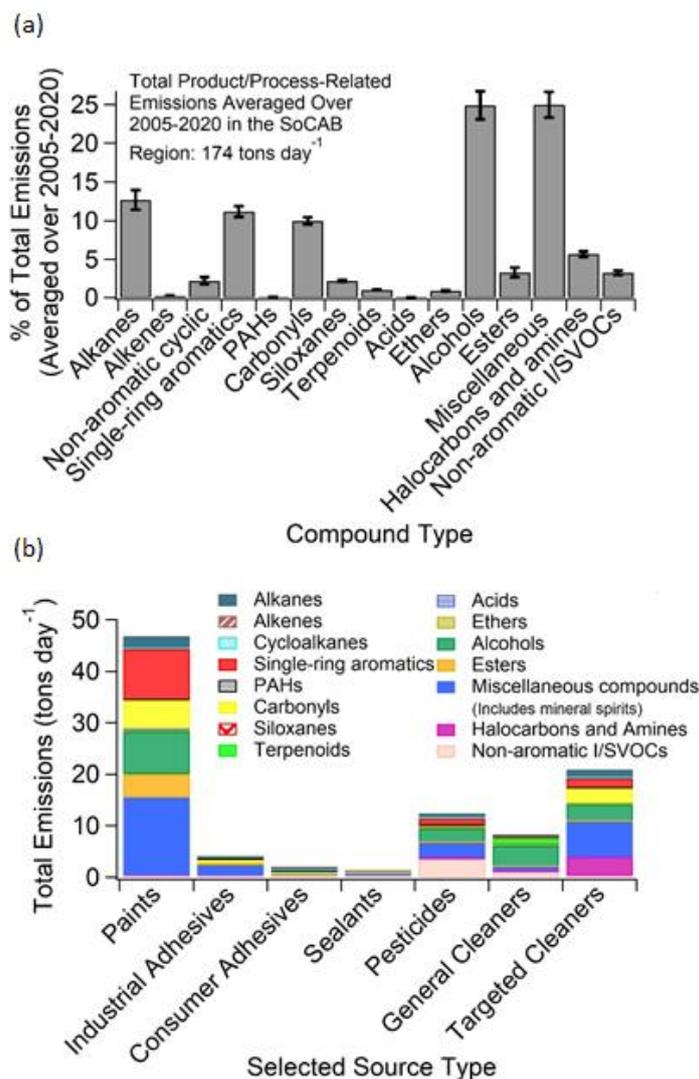
1360 **Figure 1.** Characteristic evolution and modernization of in-use motor vehicle fleets. Trends in motor vehicle
 1361 population, fuel use, and VOC emissions for model years conforming to pre-LEV, LEV-I, -II, and -III emissions
 1362 standards in California's South Coast air basin for 1990-2020 from CARB's EMFAC database (California Air
 1363 Resources Board, 2014).



1364

1365 **Figure 2.** Emissions and potential air quality impacts of product/process-related sources and motor vehicles over
1366 time in the South Coast air basin (i.e. Los Angeles). (a) Total VOC emissions (excluding ethane). (b) Potential SOA
1367 from product/process-related emissions compared to on-road motor vehicles, with uncertainties based on the ranges
1368 of compound class-specific SOA yields, and (c) Ozone formation potential via compound-specific maximum ozone
1369 incremental reactivity values (SAPRC, no uncertainty given). Product/process-related results are produced using
1370 CARB emissions data and EPA source profiles; see methods for further detail. Note: None of the panels include
1371 VOCs, IVOCs, or SVOCs, from products/processes that we identify in Sections 2 and 4 as missing from emissions
1372 inventories, but SOA and ozone formation from motor vehicle emissions does include I/SVOCs. Maximum potential
1373 SOA from gasoline vehicles in 1990 was ~41 tons day⁻¹ (off graph). Pre-1990 potential SOA and ozone for motor
1374 vehicles is excluded due to the lack of fleet-resolved data.

1375



1376

1377

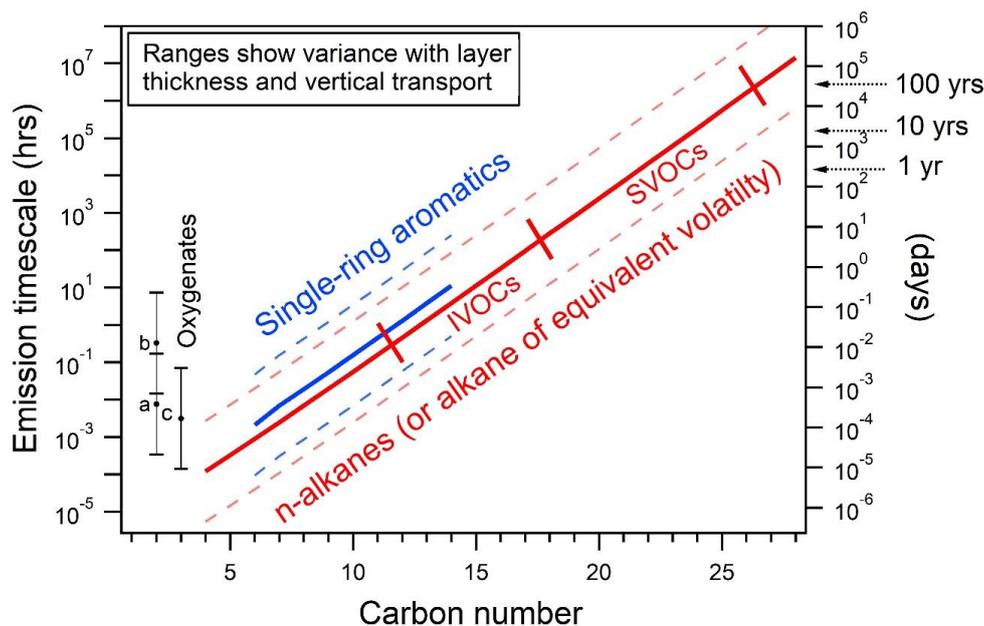
1378 **Figure 3.** Average emissions from product/process-related sources for 2005-2020 based on data from CARB

1379 inventory and SPECIATE database as a function of (a) compound class (shown with standard deviations) and (b)

1380 major product/process-related source categories in the inventory (values in Table S3). “Miscellaneous” contains

1381 70% mineral spirits (C₇₋₁₂ hydrocarbons). I/SVOCs does not include an estimate of the missing emissions identified

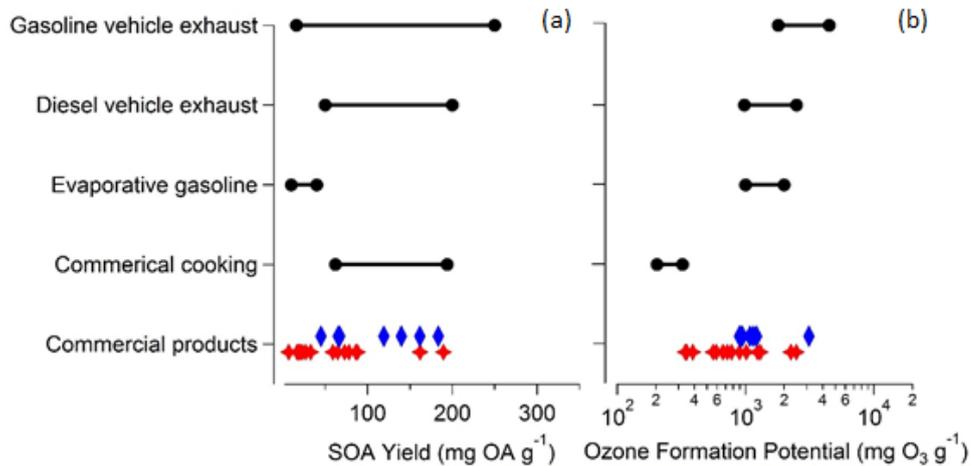
1382 in Section 2 and 4.



1383

1384 **Figure 4.** Emission timescales for single-ring aromatics, alkanes, and prominent solvents: ethanol (a), ethylene
1385 glycol (b), and acetone (c). Based on applied layers of 0.01-1 mm and vertical transport coefficients for
1386 neutral/stable outdoor conditions ($10\text{-}50\text{ m hr}^{-1}$), with longer timescales in indoor environments or thicker layers
1387 (e.g. asphalt, building materials). The left axis is reproduced on the right in days. Timescales are limited by gas-
1388 phase transport from the surface, but absorption into polymeric or porous substrates could extend timescales
1389 (Weschler and Nazaroff, 2008). Volatility range boundaries for VOC-IVOC and IVOC-SVOC occur between C_{12} -
1390 C_{13} and C_{19} - C_{20} , respectively, for n-alkanes or compounds with equivalent volatilities. See Appendix B for
1391 calculations and Table S9.

1392



1393

1394

1395 **Figure 5.** Comparison of (a) SOA yields and (b) Ozone formation potential of five major sources. Blue markers
1396 represent average yields of the product/process-related source categories in Figure 3, and do not include "missing"
1397 emissions. The red markers show yields of sampled consumer products calculated from speciation obtained after
1398 GC-MS analysis. All are shown as a function of mass emitted, not product composition.

1399

1400