

# 1 Anthropogenic—Secondary Organic Aerosols from Anthropogenic Volatile Organic 2 Compounds Contribute Substantially to Air Pollution Mortality

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57

## 58 **Abstract**

59 Anthropogenic secondary organic aerosol (ASOA), formed from anthropogenic emissions of  
60 organic compounds, constitutes a substantial fraction of the mass of submicron aerosol in  
61 populated areas around the world and contributes to poor air quality and premature mortality.  
62 However, the precursor sources of ASOA are poorly understood, and there are large uncertainties  
63 in the health benefits that might accrue from reducing anthropogenic organic emissions. We  
64 show that the production of ASOA in 11 urban areas on three continents is strongly correlated  
65 with the reactivity of specific anthropogenic volatile organic compounds. The differences in  
66 ASOA production across different cities can be explained by differences in the emissions of  
67 aromatics and intermediate- and semi-volatile organic compounds, indicating the importance of  
68 controlling these ASOA precursors. With an improved modeling representation of ASOA driven  
69 by the observations, we attribute 340,000 PM<sub>2.5</sub> premature deaths per year to ASOA, which is  
70 over an order of magnitude higher than prior studies. A sensitivity case with a more recently  
71 proposed model for attributing mortality to PM<sub>2.5</sub> (the Global Exposure Mortality Model) results  
72 in up to 900,000 deaths. A limitation of this study is the extrapolation from cities with detailed  
73 studies and regions where detailed emission inventories are available to other regions where  
74 uncertainties in emissions are larger. In addition to further development of institutional air  
75 quality management infrastructure, comprehensive air quality campaigns in the countries in  
76 South and Central America, Africa, South Asia, and the Middle East are needed for further  
77 progress in this area.

## 78 **1. Introduction**

79 Poor air quality is one of the leading causes of premature mortality worldwide (Cohen et  
80 al., 2017; Landrigan et al., 2018). Roughly 95% of the world's population live in areas where  
81  $PM_{2.5}$  (fine particulate matter with diameter smaller than 2.5  $\mu m$ ) exceeds the World Health  
82 Organization's 10  $\mu g m^{-3}$  annual average guideline (Shaddick et al., 2018). This is especially true  
83 for urban areas, where high population density is co-located with increased emissions of  $PM_{2.5}$   
84 and its gas-phase precursors from human activities. It is estimated that  $PM_{2.5}$  leads to 3 to 4  
85 million premature deaths per year, higher than the deaths associated with other air pollutants  
86 (Cohen et al., 2017). More recent analysis using concentration-response relationships derived  
87 from studies of populations exposure to high levels of ambient  $PM_{2.5}$  suggest the global  
88 premature death burden could be up to twice this value (Burnett et al., 2018).

89 The main method to estimate premature mortality with  $PM_{2.5}$  is to use measured  $PM_{2.5}$   
90 from ground observations along with derived  $PM_{2.5}$  from satellites to fill in missing ground-based  
91 observations (van Donkelaar et al., 2015, 2016). To go from total  $PM_{2.5}$  to species-dependent and  
92 even sector-dependent associated premature mortality from  $PM_{2.5}$ , chemical transport models  
93 (CTMs) are used to predict the fractional contribution of species and/or sector (e.g., Lelieveld et  
94 al., 2015; van Donkelaar et al., 2015, 2016; Silva et al., 2016). However, though CTMs may get  
95 total  $PM_{2.5}$  or even total species, e.g., organic aerosol (OA), correct, the model may be getting the  
96 values right for the wrong reason (e.g., de Gouw and Jimenez, 2009; Woody et al., 2016; Murphy  
97 et al., 2017; Baker et al., 2018; Hodzic et al., 2020). This is especially important for OA in urban  
98 areas, where models have a longstanding issue under predicting secondary OA (SOA) with some  
99 instances of over predicting primary OA (POA) (de Gouw and Jimenez, 2009; Dzepina et al.,

100 2009; Hodzic et al., 2010b; Woody et al., 2016; Zhao et al., 2016a; Janssen et al., 2017; Jathar et  
101 al., 2017). Further, this bias has even been observed for highly aged aerosols in remote regions  
102 (Hodzic et al., 2020). As has been found in prior studies for urban areas (e.g., Zhang et al., 2007;  
103 Kondo et al., 2008; Jimenez et al., 2009; DeCarlo et al., 2010; Hayes et al., 2013; Freney et al.,  
104 2014; Hu et al., 2016; Nault et al., 2018; Schroder et al., 2018) and highlighted here (Fig. 1), a  
105 substantial fraction of the observed submicron PM is OA, and a substantial fraction of the OA is  
106 composed of SOA (approximately a factor of 2 to 3 higher than POA). Thus, to better understand  
107 the sources and apportionment of  $PM_{2.5}$  that contributes to premature mortality, CTMs must  
108 improve their prediction of SOA versus POA, as the sources of SOA precursors and POA can be  
109 different.

110         However, understanding the gas-phase precursors of photochemically-produced  
111 anthropogenic SOA (ASOA, defined as the photochemically-produced SOA formed from the  
112 photooxidation of anthropogenic volatile organic compounds (AVOC) (de Gouw et al., 2005;  
113 DeCarlo et al., 2010)) quantitatively is challenging (Hallquist et al., 2009). Note, for the rest of  
114 the paper, unless explicitly stated otherwise, ASOA refers to SOA produced from the  
115 photooxidation of AVOCs, as there are potentially other relevant paths for the production of SOA  
116 in urban environments (e.g., Petit et al., 2014; Kodros et al., 2018, 2020; Stavroulas et al., 2019).  
117 Though the enhancement of ASOA is largest in large cities, these precursors and production of  
118 ASOA should be important in any location impacted by anthropogenic emissions (e.g., Fig. 1).  
119 ASOA comprises a wide range of condensable products generated by numerous chemical  
120 reactions involving AVOC precursors (Hallquist et al., 2009; Hayes et al., 2015; Shrivastava et  
121 al., 2017). The number of AVOC precursors, as well as the role of “non-traditional” AVOC

122 precursors, along with the condensable products and chemical reactions, compound to lead to  
123 differences in the observed versus predicted ASOA for various urban environments (e.g., de  
124 Gouw and Jimenez, 2009; Dzepina et al., 2009; Hodzic et al., 2010b; Woody et al., 2016; Janssen  
125 et al., 2017; Jathar et al., 2017; McDonald et al., 2018). One solution to improve the prediction in  
126 CTMs is to use a simplified model, where lumped ASOA precursors react, non-reversibly, at a  
127 given rate constant, to produce ASOA (Hodzic and Jimenez, 2011; Hayes et al., 2015; Pai et al.,  
128 2020). This simplified model has been found to reproduce the observed ASOA from some urban  
129 areas (Hodzic and Jimenez, 2011; Hayes et al., 2015) but issues in other urban areas (Pai et al.,  
130 2020). This may stem from the simplified model being parameterized to two urban areas (Hodzic  
131 and Jimenez, 2011; Hayes et al., 2015). These inconsistencies impact the model predicted  
132 fractional contribution of ASOA to total  $PM_{2.5}$  and thus the ability to understand the source  
133 attribution to  $PM_{2.5}$  and premature deaths.

134         The main categories of gas-phase precursors that dominate ASOA have been the subject  
135 of intensive research. The debate on what dominates can in turn impact the understanding of  
136 what precursors to regulate to reduce ASOA, to improve air quality, and to reduce premature  
137 mortality associated with ASOA. Transportation-related emissions (e.g., tailpipe, evaporation,  
138 refueling) were assumed to be the major precursors of ASOA, which was supported by field  
139 studies (Parrish et al., 2009; Gentner et al., 2012; Warneke et al., 2012; Pollack et al., 2013). Yet,  
140 budget closure of observed ASOA mass concentrations could not be achieved with  
141 transportation-related VOCs (Ensberg et al., 2014). The contribution of urban-emitted biogenic  
142 precursors to SOA in urban areas is typically small. Biogenic SOA (BSOA) in urban areas  
143 typically results from advection of regional background concentrations rather than processing of

144 locally emitted biogenic VOCs (e.g., Hodzic et al., 2009, 2010a; Hayes et al., 2013; Janssen et  
145 al., 2017). BSOA is thought to dominate globally (Hallquist et al., 2009), but as shown in Fig. 1,  
146 the contribution of BSOA (1% to 20%) to urban concentrations, while often substantial, is  
147 typically smaller than that of ASOA (17% to 39%) (see Sect. S3.1).

148 Many of these prior studies generally investigated AVOC with high volatility, where  
149 volatility here is defined as the saturation concentration,  $C^*$ , in  $\mu\text{g m}^{-3}$  (de Gouw et al., 2005;  
150 Volkamer et al., 2006; Dzepina et al., 2009; Freney et al., 2014; Woody et al., 2016). More recent  
151 studies have identified lower volatility compounds in transportation-related emissions (e.g., Zhao  
152 et al., 2014, 2016b; Lu et al., 2018). These compounds have been broadly identified as  
153 intermediate-volatile organic compounds (IVOCs) and semi-volatile organic compounds  
154 (SVOCs). IVOCs have a  $C^*$  generally of  $10^3$  to  $10^6 \mu\text{g m}^{-3}$  while SVOCs have a  $C^*$  generally of  
155  $1$  to  $10^2 \mu\text{g m}^{-3}$ . Due to their lower volatility and functional groups, these classes of compounds  
156 generally form ASOA more efficiently than traditional, higher volatile AVOCs; however,  
157 S/IVOCs have also been more difficult to measure (e.g., Zhao et al., 2014; Pagonis et al., 2017;  
158 Deming et al., 2018). IVOCs generally have been the more difficult of the two classes to measure  
159 and identify as these compounds cannot be collected onto filters to be sampled off-line (Lu et al.,  
160 2018) and generally show up as unresolved complex mixture for in-situ measurements using  
161 gas-chromatography (GC) (Zhao et al., 2014). SVOCs, on the other hand, can be more readily  
162 collected onto filters and sampled off-line due to their lower volatility (Lu et al., 2018). Another  
163 potential issue has been an under-estimation of the S/IVOC aerosol production, as well as an  
164 under-estimation in the contribution of photochemically produced S/IVOC from photooxidized  
165 “traditional” VOCs, due to partitioning of these low volatile compounds to chamber walls and

166 tubing (Krechmer et al., 2016; Ye et al., 2016; Liu et al., 2019). Accounting for this  
167 under-estimation increases the predicted ASOA (Ma et al., 2017). The inclusion of these classes  
168 of compounds have led to improvement in some urban SOA budget closure; however, many  
169 studies still have indicated a general short-fall in ASOA budget even when including these  
170 compounds from transportation-related emissions. (Dzepina et al., 2009; Tsimpidi et al., 2010;  
171 Hayes et al., 2015; Cappa et al., 2016; Ma et al., 2017; McDonald et al., 2018).

172         Recent studies have indicated that emissions from volatile chemical products (VCPs),  
173 defined as pesticides, coatings, inks, adhesives, personal care products, and cleaning agents  
174 (McDonald et al., 2018), as well as cooking emissions (Hayes et al., 2015), asphalt emissions  
175 (Khare et al., 2020), and solid fuel emissions from residential wood burning and/or cookstoves  
176 (e.g., Hu et al., 2013, 2020; Schroder et al., 2018), are important. While total amounts of ASOA  
177 precursors released in cities have dramatically declined (largely due to three-way catalytic  
178 converters in cars (Warneke et al., 2012; Pollack et al., 2013; Zhao et al., 2017; Khare and  
179 Gentner, 2018)), VCPs have not declined as quickly (Khare and Gentner, 2018; McDonald et al.,  
180 2018). Besides a few cities in the US (Coggon et al., 2018; Khare and Gentner, 2018; McDonald  
181 et al., 2018), extensive VCP emission quantification has not yet been published.

182         Due to the uncertainty on the emissions of ASOA precursors and on the amount of  
183 ASOA formed from them, the number of premature deaths associated with urban organic  
184 emissions is largely unknown. Since numerous studies have shown the importance of VCPs and  
185 other non-traditional VOC emission sources, efforts have been made to try to improve the  
186 representation and emissions of VCPs (Seltzer et al., 2021), which can reduce the uncertainty in  
187 ASOA precursors and the associated premature deaths estimations. Currently, most studies have



188 not ~~treated~~~~included~~ ASOA ~~explicitly~~~~realistically~~ (e.g., Lelieveld et al., 2015; Silva et al., 2016;  
189 Ridley et al., 2018) in source apportionment calculations of the premature deaths associated with  
190 long-term exposure of  $PM_{2.5}$ . ~~Most~~~~These~~ models represented total OA as non-volatile POA and  
191 “traditional” ASOA precursors (transportation-based VOCs), which largely under-predict ASOA  
192 (Ensberg et al., 2014; Hayes et al., 2015; Nault et al., 2018; Schroder et al., 2018) while  
193 over-predicting POA (e.g., Hodzic et al., 2010b; Zhao et al., 2016a; Jathar et al., 2017). This  
194 does not reflect the current understanding that POA is volatile and contributes to ASOA mass  
195 concentration (e.g., Grieshop et al., 2009; Lu et al., 2018). Though the models are estimating  
196 total OA correctly (Ridley et al., 2018; Hodzic et al., 2020; Pai et al., 2020), the attribution of  
197 premature deaths to POA instead of SOA formed from “traditional” and “non-traditional”  
198 sources, including IVOCs from both sources, could lead to regulations that may not target the  
199 emissions that would reduce OA in urban areas. As  $PM_1$  and SOA mass are highest in urban  
200 areas (Fig. 1), also shown in Jimenez et al. (2009), it is necessary to quantify the amount and  
201 identify the sources of ASOA to target future emission standards that will optimally improve air  
202 quality and the associated health impacts. As these emissions are from human activities, they will  
203 contribute to SOA mass outside urban regions and to potential health impacts outside urban  
204 regions as well. Though there are potentially other important exposure pathways to PM that may  
205 increase premature mortality, such as exposure to solid-fuel emissions indoors (e.g., Kodros et  
206 al., 2018), the focus of this paper is on exposure to outdoor ASOA and its associated impacts to  
207 premature mortality.

208       Here, we investigate the factors that control ASOA using 11 major urban, including  
209 megacities, field studies (Fig. 1 and Table 1). The empirical relationships and numerical models

210 are then used to quantify the attribution of premature mortality to ASOA around the world, using  
211 the observations to improve the modeled representation of ASOA. The results provide insight  
212 into the importance of ASOA to global premature mortality due to  $PM_{2.5}$  and further  
213 understanding of the precursors and sources of ASOA in urban regions.

214

## 215 **2. Methods**

216 Here, we introduce the ambient observations from various campaigns used to constrain  
217 ASOA production (Sect. 2.1), description of the simplified model used in CTMs to better predict  
218 ASOA (Sect. 2.2), and description of how premature mortality was estimated for this study (Sect.  
219 2.3). In the SI, the following can be found: description of the emissions used to calculate the  
220 ASOA budget for five different locations (Sect. S1), description of how the ASOA budget was  
221 calculated for the five different locations (Sect. S2), description of the CTM (GEOS-Chem) used  
222 in this study (Sect. S3 - S4), and error analysis for the observations (Sect. S5).

223

### 224 **2.1 Ambient Observations**

225 For values not previously reported in the literature (Table S4), observations taken  
226 between 11:00 – 16:00 local time were used to determine the slopes of SOA versus  
227 formaldehyde (HCHO) (Fig. S1), peroxy acetyl nitrate (PAN) (Fig. S2), and  $O_x$  ( $O_x = O_3 + NO_2$ )  
228 (Fig. S3). For CalNex, there was an approximate 48% difference between the two HCHO  
229 measurements (Fig. S4). Therefore, the average between the two measurements were used in this  
230 study, similar to what has been done in other studies for other gas-phase species (Bertram et al.,

231 2007). All linear fits, unless otherwise noted, use the orthogonal distance regression fitting  
 232 method (ODR).

233 For values in Table S4 through Table S8 not previously reported in the literature, the  
 234 following procedure was applied to determine the emissions ratios, similar to the methods of  
 235 Nault et al. (2018). An OH exposure ( $OH_{exp} = [OH] \times \Delta t$ ), which is also the photochemical age  
 236 (PA), was estimated by using the ratio of  $NO_x/NO_y$  (Eq. 1) or the ratio of  
 237 m+p-xylene/ethylbenzene (Eq. 2). For the m+p-xylene/ethylbenzene, the emission ratio  
 238 (Table S5) was determined by determining the average ratio during minimal photochemistry,  
 239 similar to prior studies (de Gouw et al., 2017). This was done for only one study, TexAQS 2000.  
 240 This method could be applied in that case as it was a ground campaign that operated both day  
 241 and night; therefore, a ratio at night could be determined when there was minimal loss of both  
 242 VOCs. The average emission ratio for the other VOCs was determined using Eq. 3 after the  
 243  $OH_{exp}$  was calculated in Eq. 1 or Eq. 2. The rate constants used for determining  $OH_{exp}$  and  
 244 emission ratios are found in Table S12.

$$245 \quad OH_{exp} = [OH] \times t = \ln \left( \frac{\left( \frac{[NO_x]}{[NO_y]} \right)}{k_{OH+NO_2}} \right) \quad \text{Eq. 1}$$

$$246 \quad OH_{exp} = [OH] \times t = - \frac{1}{k_{m+p-xylene} - k_{ethylbenzene}} \times \ln \left( \frac{[m+p-xylene]_t}{[ethylbenzene]_t} - \frac{[m+p-xylene]_0}{[ethylbenzene]_0} \right) \quad \text{Eq. 2}$$

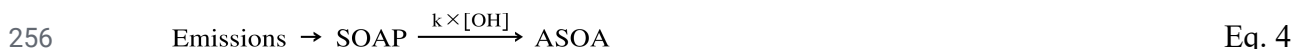
$$248 \quad \frac{[VOC(i)]}{[CO]}(0) = - \frac{[VOC(i)]}{[CO]}(t) \times \left( 1 - \frac{1}{\exp(-k_i \times [OH]_{exp} \times t)} \right) \times k_i + \frac{[VOC(i)]}{[CO]}(t) \times k_i \quad \text{Eq. 3}$$

249

250

## 251 **2.2 Updates to the SIMPLE Model**

252 With the combination of the new dataset, which expands across urban areas on three  
253 continents, the SIMPLE parameterization for ASOA (Hodzic and Jimenez, 2011) is updated in  
254 the standard GEOS-Chem model to reproduce observed ASOA in Fig. 2a. The parameterization  
255 operates as represented by Eq. 4.



257 SOAP represents the lumped precursors of ASOA,  $k$  is the reaction rate coefficient with OH  
258 ( $1.25 \times 10^{-11} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ ), and  $[\text{OH}]$  is the OH concentration in molecules  $\text{cm}^{-3}$ . This rate  
259 constant is also consistent with observed ASOA formation time scale of  $\sim 1$  day that has been  
260 observed across numerous studies (e.g., de Gouw et al., 2005; DeCarlo et al., 2010; Hayes et al.,  
261 2013; Nault et al., 2018; Schroder et al., 2018).

262 SOAP emissions were calculated based on the relationship between  $\Delta\text{SOA}/\Delta\text{CO}$  and  
263  $R_{\text{aromatics}}/\Delta\text{CO}$  in Fig. 2a. First, we calculated  $R_{\text{aromatics}}/\Delta\text{CO}$  (Eq. 5) for each grid cell and time step  
264 as follows:

$$265 \quad \frac{R_{\text{aromatics}}}{\Delta\text{CO}} = \frac{E_{\text{B}} \times k_{\text{B}} + E_{\text{T}} \times k_{\text{T}} + E_{\text{X}} \times k_{\text{X}}}{E_{\text{CO}}} \quad \text{Eq. 5}$$

266 Where  $E$  and  $k$  stand for the emission rate and reaction rate coefficient with OH, respectively, for  
267 benzene (B), toluene (T), and xylenes (X). Ethylbenzene was not included in this calculation  
268 because its emission was not available in HTAPv2 emission inventory. However, ethylbenzene  
269 contributed a minor fraction of the mixing ratio ( $\sim 7\%$ , Table S5) and reactivity ( $\sim 6\%$ ) of the  
270 total BTEX across the campaigns. Reaction rate constants used in this study were  $1.22 \times 10^{-12}$ ,

271  $5.63 \times 10^{-12}$ , and  $1.72 \times 10^{-11} \text{ cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$  for benzene, toluene, and xylene, respectively  
 272 (Atkinson and Arey, 2003; Atkinson et al., 2006). The  $R_{\text{aromatics}}/\Delta\text{CO}$  allows a dynamic  
 273 calculation of the  $E(\text{VOC})/E(\text{CO}) = \text{SOA}/\Delta\text{CO}$ . Hodzic and Jimenez (2011) and Hayes et al.  
 274 (2015) used a constant value of  $0.069 \text{ g g}^{-1}$ , which worked well for the two cities investigated,  
 275 but not for the expanded dataset studied here. Thus, both the aromatic emissions and CO  
 276 emissions are used in this study to better represent the variable emissions of ASOA precursors  
 277 (Fig. S5).

278 Second,  $E_{\text{SOAP}}/E_{\text{CO}}$  can be obtained from the result of Eq. 6, using slope and intercept in  
 279 Fig. 2a, with a correction factor (F) to consider additional SOA production after 0.5 PA  
 280 equivalent days, since Fig. 2a shows the comparison at 0.5 PA equivalent days.

$$281 \quad \frac{E_{\text{SOAP}}}{E_{\text{CO}}} = \left( \text{Slope} \times \frac{R_{\text{Aromatics}}}{\Delta\text{CO}} + \text{Intercept} \right) \times F \quad \text{Eq. 6}$$

282 Where slope is 24.8 and intercept is  $-1.7$  from Fig. 2a. F (Eq. 7) can be calculated as follows:

$$283 \quad F = \frac{ASOA_{t=\infty}}{ASOA_{t=0.5d}} = \frac{SOAP_{t=0}}{SOAP_{t=0} \times (1 - \exp(-k \times \Delta t \times [\text{OH}]))}, \Delta t = 43200 \text{ s} \quad \text{Eq. 7}$$

284 F was calculated as 1.8 by using  $[\text{OH}] = 1.5 \times 10^6 \text{ molecules cm}^{-3}$ , which was used in the  
 285 definition of 0.5 PA equivalent days for Fig. 2a.

286 Finally,  $E_{\text{SOAP}}$  can be computed by multiplying CO emissions ( $E_{\text{CO}}$ ) for every grid point  
 287 and time step in GEOS-Chem by the  $E_{\text{SOAP}}/E_{\text{CO}}$  ratio.

288

### 289 **2.3 Estimation of Premature Mortality Attribution**

290 Premature deaths were calculated for five disease categories: ischemic heart disease  
 291 (IHD), stroke, chronic obstructive pulmonary disease (COPD), acute lower respiratory illness

292 (ALRI), and lung cancer (LC). We calculated premature mortality for the population aged more  
293 than 30 years, using Eq. 8.

$$294 \quad \text{Premature Death} = \text{Pop} \times y_0 \times \frac{RR - 1}{RR} \quad \text{Eq. 8}$$

295 Mortality rate,  $y_0$ , varies according to the particular disease category and geographic region,  
296 which is available from Global Burden of Disease (GBD) Study 2015 database (IHME, 2016).  
297 Population (Pop) was obtained from Columbia University Center for International Earth Science  
298 Information Network (CIESIN) for 2010 (CIESIN, 2017). Relative risk, RR, can be calculated as  
299 shown in Eq. 9.

$$300 \quad RR = 1 + \alpha \times \left( 1 - \exp\left(\beta \times \left(PM_{2.5} - PM_{2.5,Threshold}\right)^\rho\right)\right) \quad \text{Eq. 9}$$

301  $\alpha$ ,  $\beta$ , and  $\rho$  values depend on disease category and are calculated from Burnett et al. (2014) (see  
302 Table S14 and associated file). If the  $PM_{2.5}$  concentrations are below the  $PM_{2.5}$  threshold value  
303 (Table S14), premature deaths were computed as zero. However, there could be some health  
304 impacts at concentrations below the  $PM_{2.5}$  threshold values (Krewski et al., 2009); following the  
305 methods of the GBD studies, these can be viewed as lower bounds on estimates of premature  
306 deaths.

307 We performed an additional sensitivity analysis using the Global Exposure Mortality  
308 Model (GEMM) (Burnett et al., 2018). For the GEMM analysis, we also used age stratified  
309 population data from GWPv3. Premature death is calculated the same as shown in Eq. 8;  
310 however, the relative risk differs. For the GEMM model, the relative risk can be calculated as  
311 shown in Eq. 10.

$$RR = \exp(\theta \times \lambda) \text{ with } \lambda = \frac{\log\left(1 + \frac{z}{\alpha}\right)}{\left(1 + \exp\left(\frac{(\hat{\mu} - z)}{\pi}\right)\right)} \quad \text{Eq. 10}$$

Here  $z = \max(0, PM_{2.5} - PM_{2.5, \text{Threshold}})$ ;  $\theta$ ,  $\pi$ ,  $\hat{\mu}$ ,  $\alpha$ , and  $PM_{2.5, \text{Threshold}}$  depends on disease category and are from Burnett et al. (2018). Similar to the Eq. 9, if the concentrations are below the threshold (2.4  $\mu\text{g m}^{-3}$ , Burnett et al. (2018)), then premature deaths are computed as zero; however, the GEMM has a lower threshold than the GBD method.

For GBD, we do not consider age-specific mortality rates or risks. For GEMM, we calculate age-specific health impacts with age-specific parameters in the exposure response function (Table S15). We combine the age-specific results of the exposure-response function with age distributed population data from GPW (CIESIN, 2017) and a national mortality rate across all ages to assess age-specific mortality.

We calculated total premature deaths using annual average total  $PM_{2.5}$  concentrations derived from satellite-based estimates at the resolution of  $0.1^\circ \times 0.1^\circ$  from van Donkelaar et al. (2016). Application of the remote-sensing based  $PM_{2.5}$  at the  $0.1^\circ \times 0.1^\circ$  resolution rather than direct use of the GEOS-Chem model concentrations at the  $2^\circ \times 2.5^\circ$  resolution helps reduce uncertainties in the quantification of  $PM_{2.5}$  exposure inherent in coarser estimates (Punger and West, 2013). We also calculated deaths by subtracting from this amount the total annual average ASOA concentrations derived from GEOS-Chem (Fig. S11). To reduce uncertainties related to spatial gradients and total concentration magnitudes in our GEOS-Chem simulations of  $PM_{2.5}$ , our modeled ASOA was calculated as the fraction of ASOA to total  $PM_{2.5}$  in GEOS-Chem, multiplied by the satellite-based  $PM_{2.5}$  concentrations (Eq. 11).

$$ASOA_{\text{sat}} = (ASOA_{\text{mod}} / PM_{2.5, \text{mod}}) \times PM_{2.5, \text{sat}} \quad \text{Eq. 11}$$

333 Finally, this process for estimating  $PM_{2.5}$  health impacts considers only  $PM_{2.5}$  mass concentration  
334 and does not distinguish toxicity by composition, consistent with the current US EPA position  
335 expressed in Sacks et al. (2019).

336

### 337 **3. Observations of ASOA Production across Three Continents**

#### 338 **3.1 Observational Constraints of ASOA Production across Three Continents**

339 Measurements during intensive field campaigns in large urban areas better constrain  
340 concentrations and atmospheric formation of ASOA because the scale of ASOA enhancement is  
341 large compared to SOA from a regional background. Generally, ASOA increased with the  
342 amount of urban precursor VOCs and with atmospheric PA (de Gouw et al., 2005; de Gouw and  
343 Jimenez, 2009; DeCarlo et al., 2010; Hayes et al., 2013; Nault et al., 2018; Schroder et al., 2018;  
344 Shah et al., 2018). In addition, ASOA correlates strongly with gas-phase secondary  
345 photochemical species, including  $O_x$ , HCHO, and PAN (Herndon et al., 2008; Wood et al., 2010;  
346 Hayes et al., 2013; Zhang et al., 2015; Nault et al., 2018; Liao et al., 2019) (Table S4; Fig. S1 to  
347 Fig. S3), which are indicators of photochemical processing of emissions.

348 However, as initially discussed by Nault et al. (2018) and shown in Fig. 3, there is large  
349 variability in these various metrics across the urban areas evaluated here. To the best of the  
350 authors' knowledge, this variability has not been explored and its physical meaning has not been  
351 interpreted. As shown in Fig. 3, though, the trends in  $\Delta SOA/\Delta CO$  are similar to the trends in the  
352 slopes of SOA versus  $O_x$ , PAN, or HCHO. For example, Seoul is the highest for nearly all  
353 metrics, and is approximately a factor of 6 higher than the urban area, Houston, that generally



354 showed the lowest photochemical metrics. This suggests that the variability is related to a  
355 physical factor, including emissions and chemistry.

356 The VOC concentration, together with how quickly the emitted VOCs react ( $\sum k_i \times [\text{VOC}]_i$ ,  
357 i.e., the hydroxyl radical, or OH, reactivity of VOCs), where k is the OH rate coefficient for each  
358 VOC, are a determining parameter for ASOA formation over urban spatial scales (Eq. 12).  
359 ASOA formation is normalized here to the excess CO mixing ratio ( $\Delta\text{CO}$ ) to account for the  
360 effects of meteorology, dilution, and non-urban background levels, and allow for easier  
361 comparison between different studies:

$$362 \quad \frac{\Delta \text{ASOA}}{\Delta \text{CO}} \propto [\text{OH}] \times \Delta t \times \left( \sum_i k_i \times \left[ \frac{\text{VOC}}{\text{CO}} \right]_i \times Y_i \right) \quad \text{Eq. 12}$$

363 where Y is the aerosol yield for each compound (mass of SOA formed per unit mass of precursor  
364 reacted), and  $[\text{OH}] \times \Delta t$  is the PA.

365 BTEX are one group of known ASOA precursors (Gentner et al., 2012; Hayes et al.,  
366 2013), and their emission ratio (to CO) was determined for all campaigns (Table S5). BTEX can  
367 thus provide insight into ASOA production. Fig. 2a shows that the variation in ASOA (at PA =  
368 0.5 equivalent days) is highly correlated with the emission reactivity ratio of BTEX ( $R_{\text{BTEX}}$ ,  
369  $\sum_i [\text{VOC}/\text{CO}]_i$ ) across all the studies. However, BTEX alone cannot account for much of the  
370 ASOA formation (see budget closure discussion below), and instead, BTEX may be better  
371 thought of as both partial contributors and also as indicators for the co-emission of other  
372 (unmeasured) organic precursors that are also efficient at forming ASOA.

373  $\text{O}_x$ , PAN, and HCHO are produced from the oxidation of a much wider set of VOC  
374 precursors (including small alkenes, which do not appreciably produce SOA when oxidized).

375 These alkenes have similar reaction rate constants with OH as the most reactive BTEX  
376 compounds (Table S12); however, their emissions and concentration can be higher than BTEX  
377 (Table S7). Thus, alkenes would dominate  $R_{\text{Total}}$ , leading to  $O_x$ , HCHO, and PAN being produced  
378 more rapidly than ASOA (Fig. 2b–d). When  $R_{\text{BTEX}}$  becomes more important for  $R_{\text{Total}}$ , the emitted  
379 VOCs are more efficient in producing ASOA. Thus, the ratio of ASOA to gas-phase  
380 photochemical products shows a strong correlation with  $R_{\text{BTEX}}/R_{\text{Total}}$  (Fig. 2b–d).

381 An important aspect of this study is that most of these observations occurred during  
382 spring and summer, when solid fuel emissions are expected to be lower (e.g., Chafe et al., 2015;  
383 Lam et al., 2017; Hu et al., 2020). Further, the most important observations used here are during  
384 the afternoon, investigating specifically the photochemically produced ASOA. These results here  
385 might partially miss any ASOA produced through nighttime aqueous chemistry or oxidation by  
386 nitrate radical (Kodros et al., 2020). However, two of the studies included in our analysis,  
387 Chinese Outflow (CAPTAIN, 2011) and New York City (WINTER, 2015), occurred in late  
388 winter/early spring, when solid fuel emissions were important (Hu et al., 2013; Schroder et al.,  
389 2018). We find that these observations lie within the uncertainty in the slope between ASOA and  
390  $R_{\text{BTEX}}$  (Fig. 2a). Their photochemically produced ASOA observed under strong impact from solid  
391 fuel emissions shows similar behavior as the ASOA observed during spring and summer time.  
392 Thus, given the limited datasets currently available, photochemically produced ASOA is  
393 expected to follow the relationship shown in Fig. 2a and is expected to also follow this  
394 relationship for regions impacted by solid fuel burning. Future comprehensive studies in regions  
395 strongly impacted by solid fuel burning are needed to further investigate photochemical ASOA  
396 production under those conditions.

397

### 398 **3.2 Budget Closure of ASOA for 4 Urban Areas on 3 Continents Indicates Reasonable** 399 **Understanding of ASOA Sources**

400 To investigate the correlation between ASOA and  $R_{\text{BTEX}}$ , a box model using the emission  
401 ratios from BTEX (Table S5), other aromatics (Table S8), IVOCs (Sect. S1), and SVOCs (Sect.  
402 S1) was run for five urban areas: New York City, 2002, Los Angeles, Beijing, London, and New  
403 York City, 2015 (see Sect. S1 and S3 for more information). The differences in the results shown  
404 in Fig. 4 are due to differences in the emissions for each city. We show that BTEX alone cannot  
405 explain the observed ASOA budget for urban areas around the world. Fig. 4a shows that  
406 approximately  $25\pm 6\%$  of the observed ASOA originates from the photooxidation of BTEX.  
407 BTEX only explaining 25% of the observed ASOA is similar to prior studies that have done  
408 budget analysis of precursor gases and observed SOA (e.g., Dzepina et al., 2009; Ensberg et al.,  
409 2014; Hayes et al., 2015; Ma et al., 2017; Nault et al., 2018). Therefore, other precursors must  
410 account for most of the ASOA produced.

411 Because alkanes, alkenes, and oxygenated compounds with carbon numbers less than 6  
412 are not significant ASOA precursors, we focus on emissions and sources of BTEX, other  
413 mono-aromatics, IVOCs, and SVOCs. These three classes of VOCs, aromatics, IVOCs, and  
414 SVOCs, have been suggested to be significant ASOA precursors in urban atmospheres  
415 (Robinson et al., 2007; Hayes et al., 2015; Ma et al., 2017; McDonald et al., 2018; Nault et al.,  
416 2018; Schroder et al., 2018; Shah et al., 2018), originating from both fossil fuel and VCP  
417 emissions.

418 Using the best available emission inventories from cities on three continents  
419 (EMEP/EEA, 2016; McDonald et al., 2018; Li et al., 2019) and observations, we quantify the  
420 emissions of BTEX, other mono-aromatics, IVOCs, and SVOCs for both fossil fuel (e.g.,  
421 gasoline, diesel, kerosene, etc.), VCPs (e.g., coatings, inks, adhesives, personal care products,  
422 and cleaning agents), and cooking sources (Fig. 5). This builds off the work of McDonald et al.  
423 (2018) for urban regions on three different continents.

424 Note, the emissions investigated here ignore any oxygenated VOC emissions not  
425 associated with IVOCs and SVOCs due to the challenge in estimating the emission ratios for  
426 these compounds (de Gouw et al., 2018). Further, SVOC emission ratios are estimated from the  
427 average POA observed by the AMS during the specific campaign and scaled by profiles in  
428 literature for a given average temperature and average OA (Robinson et al., 2007; Worton et al.,  
429 2014; Lu et al., 2018). As most of the campaigns had an average OA between 1 and 10  $\mu\text{g m}^{-3}$   
430 and temperature of  $\sim 298$  K, this led to the majority of the estimated emitted SVOC gases in the  
431 highest SVOC bin. However, as discussed later, this does not lead to SVOCs dominating the  
432 predicted ASOA due to taking into account the fragmentation and overall yield from the  
433 photooxidation of SVOC to ASOA.

434 Combining these inventories and observations for the various locations provide the  
435 following insights about the potential ASOA precursors not easily measured or quantified in  
436 urban environments (e.g., Zhao et al., 2014; Lu et al., 2018): (1) aromatics from fossil fuel  
437 accounts for 14-40% (mean 22%) of the total BTEX and IVOC emissions for the five urban  
438 areas investigated in-depth (Fig. 5), agreeing with prior studies that have shown that the observed  
439 ASOA cannot be reconciled by the observations or emission inventory of aromatics from fossil

440 fuels (e.g., Ensberg et al., 2014; Hayes et al., 2015). (2) BTEX from both fossil fuels and VCPs  
441 account for 25-95% (mean 43%) of BTEX and IVOC emissions (Fig. 5). China has the lowest  
442 contribution of IVOCs, potentially due to differences in chemical make-up of the solvents used  
443 daily (Li et al., 2019), but more research is needed to investigate the differences in IVOCs:BTEX  
444 from Beijing versus US and UK emission inventories. Nonetheless, this shows the importance of  
445 IVOCs for both emissions and ASOA precursors. (3) IVOCs are generally equal to, if not greater  
446 than, the emissions of BTEX in 4 of the 5 urban areas investigated here (Fig. 5). (4) Overall,  
447 VCPs account for a large fraction of the BTEX and IVOC emissions for all five cities. (5)  
448 Finally, SVOCs account for 27-88% (mean 53%) of VOCs generally considered ASOA  
449 precursors (VOCs with volatility saturation concentrations  $\leq 10^7 \mu\text{g m}^{-3}$ ) (Fig. S6). Beijing has  
450 the highest contribution of SVOCs to ASOA precursors due to the use of solid fuels and cooking  
451 emissions (Hu et al., 2016). Also, this indicates the large contribution of a class of VOCs  
452 difficult to measure (Robinson et al., 2007) that are an important ASOA precursor (e.g., Hayes et  
453 al., 2015), showing further emphasis should be placed in quantifying the emissions of this class  
454 of compounds.

455         These results provide an ability to further investigate the mass balance of predicted and  
456 observed ASOA for these urban locations (Fig. 4). The inclusion of IVOCs, other aromatics not  
457 including BTEX, and SVOCs leads to the ability to explain, on average,  $85\pm 12\%$  of the observed  
458 ASOA for these urban locations around the world (Fig. 4a). Further, VCP contribution to ASOA  
459 is important for all these urban locations, accounting for, on average,  $37\pm 3\%$  of the observed  
460 ASOA (Fig. 4b).

461 This bottom-up mass budget analysis provides important insights to further explain the  
462 correlation observed in Fig. 2. First, IVOCs are generally co-emitted from similar sources as  
463 BTEX for the urban areas investigated in-depth (Fig. 5). The oxidation of these co-emitted  
464 species leads to the ASOA production observed across the urban areas around the world. Second,  
465 S/IVOCs generally have similar rate constants as toluene and xylenes ( $\geq 1 \times 10^{-11} \text{ cm}^3 \text{ molec.}^{-1} \text{ s}^{-1}$ )  
466 (Zhao et al., 2014, 2017), the compounds that contribute the most to  $R_{\text{BTEX}}$ , explaining the rapid  
467 ASOA production that has been observed in various studies (de Gouw and Jimenez, 2009;  
468 DeCarlo et al., 2010; Hayes et al., 2013; Hu et al., 2013, 2016; Nault et al., 2018; Schroder et al.,  
469 2018) and correlation (Fig. 2). Finally, the contribution of VCPs and fossil fuel sources to ASOA  
470 is similar across the cities, expanding upon and further supporting the conclusion of McDonald  
471 et al. (2018) in the importance of identifying and understanding VCP emissions in order to  
472 explain ASOA.

473 This investigation shows that the bottom-up calculated ASOA agrees with observed  
474 top-down ASOA within 15%. As highlighted above, this ratio is explained by the co-emissions  
475 of IVOCs with BTEX from traditional sources (diesel, gasoline, and other fossil fuel emissions)  
476 and VCPs (Fig. 5) along with similar rate constants for these ASOA precursors (Table S12).  
477 Thus, the  $\text{ASOA}/R_{\text{BTEX}}$  ratio obtained from Fig. 2 results in accurate predictions of ASOA for the  
478 urban areas evaluated here, and this value can be used to better estimate ASOA with chemical  
479 transport models (Sect. 4).

480

#### 481 **4. Improved Urban SIMPLE Model Using Multi-Cities to Constrain**

482 The SIMPLE model was originally designed and tested against the observations collected  
483 around Mexico City (Hodzic and Jimenez, 2011). It was then tested against observations  
484 collected in Los Angeles (Hayes et al., 2015; Ma et al., 2017). As both data sets have nearly  
485 identical  $\Delta\text{SOA}/\Delta\text{CO}$  and  $R_{\text{BTEX}}$  (Fig. 2 and Fig. 3), it is not surprising that the SIMPLE model  
486 did well in predicting the observed  $\Delta\text{SOA}/\Delta\text{CO}$  for these two urban regions with consistent  
487 parameters. Though the SIMPLE model generally performed better than more explicit models, it  
488 generally had lower skill in predicting the observed ASOA in urban regions outside of Mexico  
489 City and Los Angeles (Shah et al., 2019; Pai et al., 2020).

490 This may stem from the original SIMPLE model with constant parameters missing the  
491 ability to change the amount and reactivity of the emissions, which are different for the various  
492 urban regions, versus the ASOA precursors being emitted proportionally to only CO (Hodzic and  
493 Jimenez, 2011; Hayes et al., 2015). For example, in the HTAP emissions inventory, the CO  
494 emissions for Seoul, Los Angeles, and Mexico City are all similar (Fig. S8); thus, the original  
495 SIMPLE model would suggest similar  $\Delta\text{SOA}/\Delta\text{CO}$  for all three urban locations. However, as  
496 shown in Fig. 2 and Fig. 3, the  $\Delta\text{SOA}/\Delta\text{CO}$  is different by nearly a factor of 2. The inclusion of  
497 the emissions and reactivity, where  $R_{\text{BTEX}}$  for Seoul is approximately a factor of 2.5 higher than  
498 Los Angeles and Seoul, into the improved SIMPLE model better accounts for the variability in  
499 SOA production, as shown in Fig. 2. Thus, the inclusion and use of this improved SIMPLE  
500 model refines the simplified representation of ASOA in chemical transport models and/or box  
501 models.

502 The “improved” SIMPLE shows higher ASOA compared to the default VBS  
503 GEOS-Chem (Fig. 6a,b). In areas strongly impacted by urban emissions (e.g., Europe, East Asia,

504 India, east and west coast US, and regions impacted by Santiago, Chile, Buenos Aires,  
505 Argentina, Sao Paulo, Brazil, Durban and Cape Town, South Africa, and Melbourne and Sydney,  
506 Australia), the “improved” SIMPLE model predicts up to  $14 \mu\text{g m}^{-3}$  more ASOA, or ~30 to 60  
507 times more ASOA than the default scheme (Fig. 6c,d). As shown in Fig. 1, during intensive  
508 measurements, the ASOA composed 17-39% of  $\text{PM}_{10}$ , with an average contribution of ~25%. The  
509 default ASOA scheme in GEOS-Chem greatly underestimates the fractional contribution of  
510 ASOA to total  $\text{PM}_{2.5}$  (<2%; Fig. 6e). The “improved” SIMPLE model greatly improves the  
511 predicted fractional contribution, showing that ASOA in the urban regions ranges from 15-30%,  
512 with an average of ~15% for the grid cells corresponding to the urban areas investigated here  
513 (Fig. 6f). Thus, the “improved” SIMPLE predicts the fractional contribution of ASOA to total  
514  $\text{PM}_{2.5}$  far more realistically, compared to observations. As discussed in Sect. 2.3 and Eq. 11,  
515 having the model accurately predict the fractional contribution of ASOA to the total PM is very  
516 important, as the total  $\text{PM}_{2.5}$  is derived from satellite-based estimates (van Donkelaar et al.,  
517 2015), and the model fractions are then applied to those total  $\text{PM}_{2.5}$  estimates. The ability for the  
518 “improved” SIMPLE model to better represent the ASOA composition provides confidence  
519 attributing the ASOA contribution to premature mortality.

520

## 521 **5. Preliminary Evaluation of Worldwide Premature Deaths Due to ASOA with Updated** 522 **SIMPLE Parameterization**

523         The improved SIMPLE parameterization is used along with GEOS-Chem to provide an  
524 accurate estimation of ASOA formation in urban areas worldwide and provide an ability to  
525 obtain realistic simulations of ASOA based on measurement data. We use this model to quantify



526 the attribution of  $PM_{2.5}$  ASOA to premature deaths. Analysis up to this point has been for  $PM_1$ ;  
527 however, both the chemical transport model and epidemiological studies utilize  $PM_{2.5}$ . For  
528 ASOA, this will not impact the discussion and results here because the mass of OA (typically  
529 80–90%) is dominated by  $PM_1$  (e.g., Bae et al., 2006; Seinfeld and Pandis, 2006), and ASOA is  
530 formed mostly through condensation of oxidized species, which favors partitioning onto smaller  
531 particles (Seinfeld and Pandis, 2006).

532         The procedure for this analysis is described in Fig. 7 and Sect. 2.3 and S3. Briefly, we  
533 combine high-resolution satellite-based  $PM_{2.5}$  estimates (for exposure) and a chemical transport  
534 model (GEOS-Chem, for fractional composition) to estimate ASOA concentrations and various  
535 sensitivity analysis (van Donkelaar et al., 2015). We calculated ~3.3 million premature deaths  
536 (using the Integrated Exposure-Response, IER, function) are due to long-term exposure of  
537 ambient  $PM_{2.5}$  (Fig. S9, Table S16), consistent with recent literature (Cohen et al., 2017).

538         The attribution of ASOA  $PM_{2.5}$  premature deaths can be calculated one of two ways: (a)  
539 marginal method (Silva et al., 2016) or (b) attributable fraction method (Anenberg et al., 2019).  
540 For method (a), it is assumed that a fraction of the ASOA is removed, keeping the rest of the  
541  $PM_{2.5}$  components approximately constant, and the change in deaths is calculated from the deaths  
542 associated with the total concentration less the deaths calculated using the reduced total  $PM_{2.5}$   
543 concentrations. For method (b), the health impact is attributed to each  $PM_{2.5}$  component by  
544 multiplying the total deaths by the fractional contribution of each component to total  $PM_{2.5}$ . For  
545 method (a), the deaths attributed to ASOA are ~340,000 people per year (Fig. 8); whereas, for  
546 method (b), the deaths are ~370,000 people per year. Both of these are based on the IER response  
547 function (Cohen et al., 2017).

548 Additional recent work (Burnett et al., 2018) has suggested less reduction in the  
549 premature deaths versus  $PM_{2.5}$  concentration relationship at higher  $PM_{2.5}$  concentrations, and  
550 lower concentration limits for the threshold below which this relationship is negligible, both of  
551 which lead to much higher estimates of  $PM_{2.5}$  associated premature deaths. This is generally  
552 termed the Global Exposure Mortality Model (GEMM). Using the two attribution methods  
553 described above (a and b), the ASOA  $PM_{2.5}$  premature deaths are estimated to be ~640,000  
554 (method a) and ~900,000 (method b) (Fig. S9 and Fig. S12 and Table S17).

555 Compared to prior studies using chemical transport models to estimate premature deaths  
556 associated with ASOA (e.g., Silva et al., 2016; Ridley et al., 2018), which assumed non-volatile  
557 POA and “traditional” ASOA precursors, the attribution of premature mortality due to ASOA is  
558 over an order of magnitude higher in this study (Fig. 9). This occurs using either the IER and  
559 GEMM approach for estimating premature mortality (Fig. 9). For regions with larger populations  
560 and more  $PM_{2.5}$  pollution, the attribution is between a factor of 40 to 80 higher. This stems from  
561 the non-volatile POA and “traditional” ASOA precursors over-estimating POA and  
562 under-estimating ASOA compared to observations (Schroder et al., 2018). These offsetting  
563 errors will lead to model predicted total OA similar to observations (Ridley et al., 2018; Schroder  
564 et al., 2018), yet different conclusions on whether POA versus SOA is more important for  
565 reducing  $PM_{2.5}$  associated premature mortality. Using a model constrained to day-time  
566 atmospheric observations (Fig. 2 and Fig. 4, see Sect. 4) leads to a more accurate **than earlier**  
567 estimation of the contribution of photochemically-produced ASOA to  $PM_{2.5}$  associated  
568 premature mortality that has not been possible in prior studies. We note that ozone concentrations  
569 change little as we change the ASOA simulation (see Sect. S4 and Fig. S14).

570 A limitation in this study is the lack of sufficient measurements in South and Southeast  
571 Asia, Eastern Europe, Africa, and South America (Fig. 1), though these areas account for 44% of  
572 the predicted reduction in premature mortality for the world (Table S16). However, as  
573 highlighted in Table S18, these regions likely still consume both transportation fuels and VCPs,  
574 although in lower per capita amounts than more industrialized countries. This consumption is  
575 expected to lead to the same types of emissions as for the cities studied here, though more field  
576 measurements are needed to validate global inventories of VOCs and resulting oxidation  
577 products in the developing world. Transportation emissions of VOCs are expected to be more  
578 dominant in the developing world due to higher VOC emission factors associated with inefficient  
579 combustion engines, such as two-stroke scooters (Platt et al., 2014) and auto-rickshaws (e.g.,  
580 Goel and Guttikunda, 2015).

581 Solid fuels are used for residential heating and cooking, which impact the outdoor air  
582 quality as well (Hu et al., 2013, 2016; Lacey et al., 2017; Stewart et al., 2020), and which also  
583 lead to SOA (Heringa et al., 2011). As discussed in Sect. 3.1, though the majority of the studies  
584 evaluated here occurred in spring to summer time, when solid fuel emissions are decreased, two  
585 studies occurred during the winter/early spring time, where solid fuel emissions were important  
586 (Hu et al., 2013; Schroder et al., 2018). These studies still follow the same relationship between  
587 ASOA and  $R_{\text{BTEX}}$  as the studies that focused on spring/summer time photochemistry. Thus, the  
588 limited datasets available indicate that photochemically produced ASOA from solid fuels follow  
589 a similar relationship to that from other ASOA sources.

590 Also, solid fuel sources are included in the inventories used in our modeling. For the  
591 HTAP emission inventory used here (Janssens-Maenhout et al., 2015), small-scale combustion,

592 which includes heating and cooking (e.g., solid-fuel use), is included in the residential emission  
593 sector. Both CO and BTEX are included in this source, and can account for a large fraction of the  
594 total emissions where solid-fuel use may be important (Fig. S15). Thus, as CO and BTEX are  
595 used in the updated SIMPLE model, and campaigns that observed solid-fuel emissions fall  
596 within the trend for all urban areas, the solid-fuel contribution to photochemically-produced  
597 ASOA is accounted for (as accurately as allowed by current datasets) in the estimation of ASOA  
598 for the attribution to premature mortality.

599 Note that recent work has observed potential nighttime aqueous chemistry and/or  
600 oxidation by nitrate radical from solid fuel emissions to produce ASOA (Kodros et al., 2020).  
601 Thus, missing this source of ASOA may lead to an underestimation of total ASOA versus the  
602 photochemically-produced ASOA we discuss here, leading to a potential underestimation in the  
603 attribution of ASOA to premature mortality. From the studies that investigated “night-time  
604 aging” of solid-fuel emissions to form SOA, we predict that the total ASOA may be  
605 underestimated by 1 to 3  $\mu\text{g m}^{-3}$  (Kodros et al., 2020). This potential underestimation, though, is  
606 less than the current underestimation in ASOA in GEOS-Chem (default versus “Updated”  
607 SIMPLE).

608 Recently, emission factors from Abidjan, Côte d’Ivoire, a developing urban area, showed  
609 the dominance of emissions from transportation and solid fuel burning, with BTEX being an  
610 important fraction of the total emissions, and that all the emissions were efficient in producing  
611 ASOA (Dominutti et al., 2019). Further, investigation of emissions in New Delhi region of India  
612 demonstrated the importance of both transportation and solid fuel emissions (Stewart et al.,  
613 2020; Wang et al., 2020) while model comparisons with observations show an underestimation

614 of OA compared to observations due to a combination of emissions and OA representation (Jena  
615 et al., 2020). Despite emission source differences, SOA is still an important component of  $PM_{2.5}$   
616 (e.g., Singh et al., 2019) and thus will impact air quality and premature mortality in developing  
617 regions. Admittedly, though, our estimates will be less accurate for these regions.

618

## 619 **6. Conclusions**

620 In summary, ASOA is an important, though inadequately constrained component of air  
621 pollution in megacities and urban areas around the world. This stems from the complexity  
622 associated with the numerous precursor emission sources, chemical reactions, and oxidation  
623 products that lead to observed ASOA concentrations. We have shown here that the variability in  
624 observed ASOA across urban areas is correlated with  $R_{\text{BTEX}}$ , a marker for the co-emissions of  
625 IVOC from both transportation and VCP emissions. Global simulations indicate ASOA  
626 contributes to a substantial fraction of the premature mortality associated with  $PM_{2.5}$ . Reductions  
627 of the ASOA precursors will reduce the premature deaths associated with  $PM_{2.5}$ , indicating the  
628 importance of identifying and reducing exposure to sources of ASOA. These sources include  
629 emissions that are both traditional (transportation) as well as non-traditional emissions of  
630 emerging importance (VCPs) to ambient  $PM_{2.5}$  concentrations in cities around the world. Further  
631 investigation of speciated IVOCs and SVOCs for urban areas around the world along with SOA  
632 mass concentration and other photochemical products (e.g.,  $O_x$ , PAN, and HCHO) for other  
633 urban areas, especially in South Asia, throughout Africa, and throughout South America, would  
634 provide further constraints to improve the SIMPLE model and our understanding of the emission  
635 sources and chemistry that leads to the observed SOA and its impact on premature mortality.

## 636 Acknowledgements

637

638 This study was partially supported by grants from NASA NNX15AT96G, NNX16AQ26G, Sloan  
639 Foundation 2016-7173, NSF AGS-1822664, EPA STAR 83587701-0, NERC NE/H003510/1,  
640 NERC NE/H003177/1, NERC NE/H003223/1, NOAA NA17OAR4320101, NCAS  
641 R8/H12/83/037, Natural Science and Engineering Research Council of Canada (NSERC,  
642 RGPIN/05002-2014), and the Fonds de Recherche du Québec —Nature et technologies  
643 (FRQNT, 2016-PR-192364). This manuscript has not been formally reviewed by EPA. The  
644 views expressed in this document are solely those of the authors and do not necessarily reflect  
645 those of the Agency. EPA does not endorse any products or commercial services mentioned in  
646 this publication. We thank Katherine Travis for useful discussions. We acknowledge B J. Bandy,  
647 J. Lee, G. P. Mills, d. D. Montzka, J. Stutz, A. J. Weinheimer E. J. Williams, E. C. Wood, and D.  
648 R. Worsnop for use of their data.

649

## 650 Data Availability

651 TexAQS measurements are available at  
652 <https://esrl.noaa.gov/csl/groups/csl7/measurements/2000TexAQS/LaPorte/DataDownload/> and  
653 upon request. NEAQS measurements are available at  
654 <https://www.esrl.noaa.gov/csl/groups/csl7/measurements/2002NEAQS/>. MILAGRO  
655 measurements are available at <http://doi.org/10.5067/Aircraft/INTEXB/Aerosol-TraceGas>.  
656 CalNex measurements are available at  
657 <https://esrl.noaa.gov/csl/groups/csl7/measurements/2010calnex/Ground/DataDownload/>.  
658 ClearfLo measurements are available at  
659 <https://catalogue.ceda.ac.uk/uuid/6a5f9eedd68f43348692b3bace3eba45>. SEAC<sup>4</sup>RS measurements  
660 are available at <http://doi.org/10.5067/Aircraft/SEAC4RS/Aerosol-TraceGas-Cloud>. WINTER  
661 measurements are available at [https://data.eol.ucar.edu/master\\_lists/generated/winter/](https://data.eol.ucar.edu/master_lists/generated/winter/).  
662 KORUS-AQ measurements are available at  
663 <http://doi.org/10.5067/Suborbital/KORUSAQ/DATA01>. Data from Chinese campaigns are  
664 available upon request, and rest of data used were located in papers cited. GEOS-Chem data  
665 available upon request. Figures will become accessible at  
666 [cires1.colorado.edu/jimenez/group\\_pubs.html](https://cires1.colorado.edu/jimenez/group_pubs.html).

667

## 668 Competing Interests

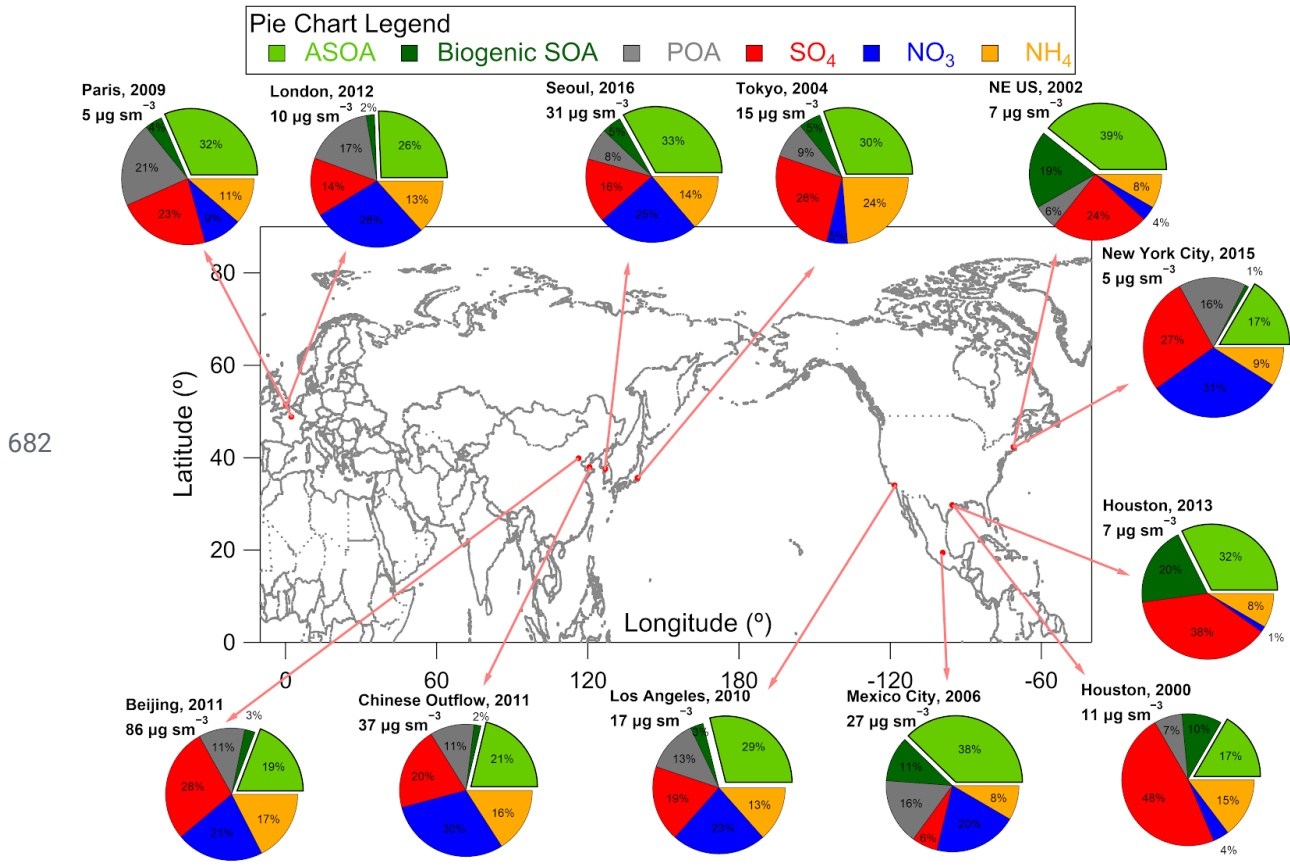
669 The authors declare no competing interests.

670

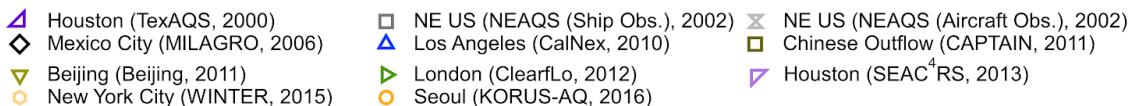
## 671 Author Contribution

672 B.A.N., D.S.J., B.C.M., J.A.dG., and J.L.J designed the experiment and wrote the paper. B.A.N.,  
673 PC.-J., D.A.D., W.H., J.C.S, J.A., D.R.B., M.R.C., H.C., M.M.C., P.F.D, G.S.D., R.D., F.F, A.F.,  
674 J.B.G., G.G., J.F.H, T.F.H., P.L.H., J.H., M.H., L.G.H., B.T.J., W.C.K., J.L., I.B.P., J.P., B.R.,

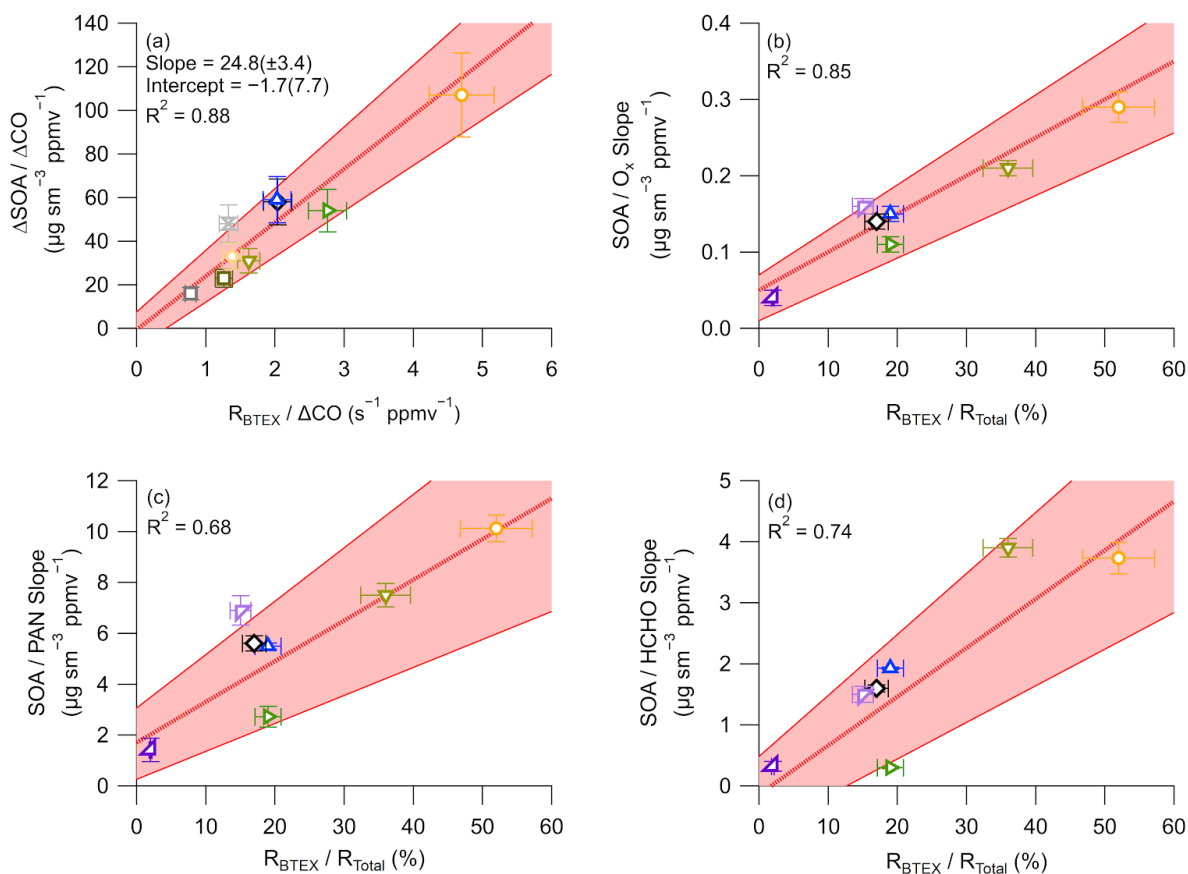
675 C.E.R., D.R., J.M.R., T.B.R, M.S., J.W., C.W., P.W., G.M.W., D.E.Y., B.Y., J.A.dG., and J.L.J.  
676 collected and analyzed the data. D.S.J. and A.H. ran the GEOS-Chem model and B.A.N., D.S.J,  
677 and J.L.J. analyzed the model output. B.A.N., P.L.H., J.M.S., and J.L.J. ran and analyzed the 0-D  
678 model used for ASOA budget analysis of ambient observations. B.C.M., A.L., M.L., and Q.Z.  
679 analyzed and provided the emission inventories used for the 0-D box model. D.S.J., D.K.H., and  
680 M.O.N. conducted the ASOA attribution to mortality calculation, and B.A.N., D.S.J., D.K.H.,  
681 M.O.N., J.A.dG, and J.L.J analyzed the results. All authors reviewed the paper.





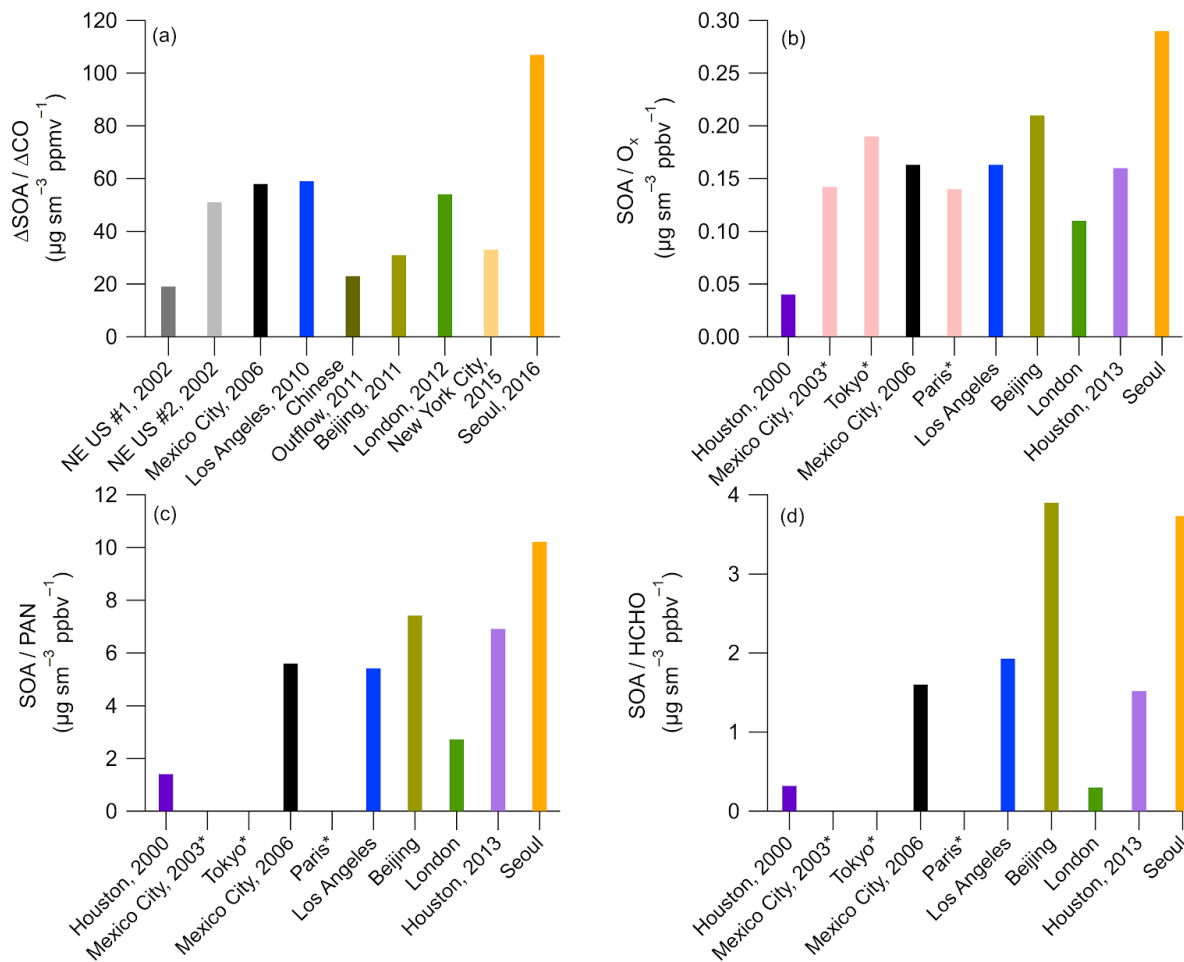


687



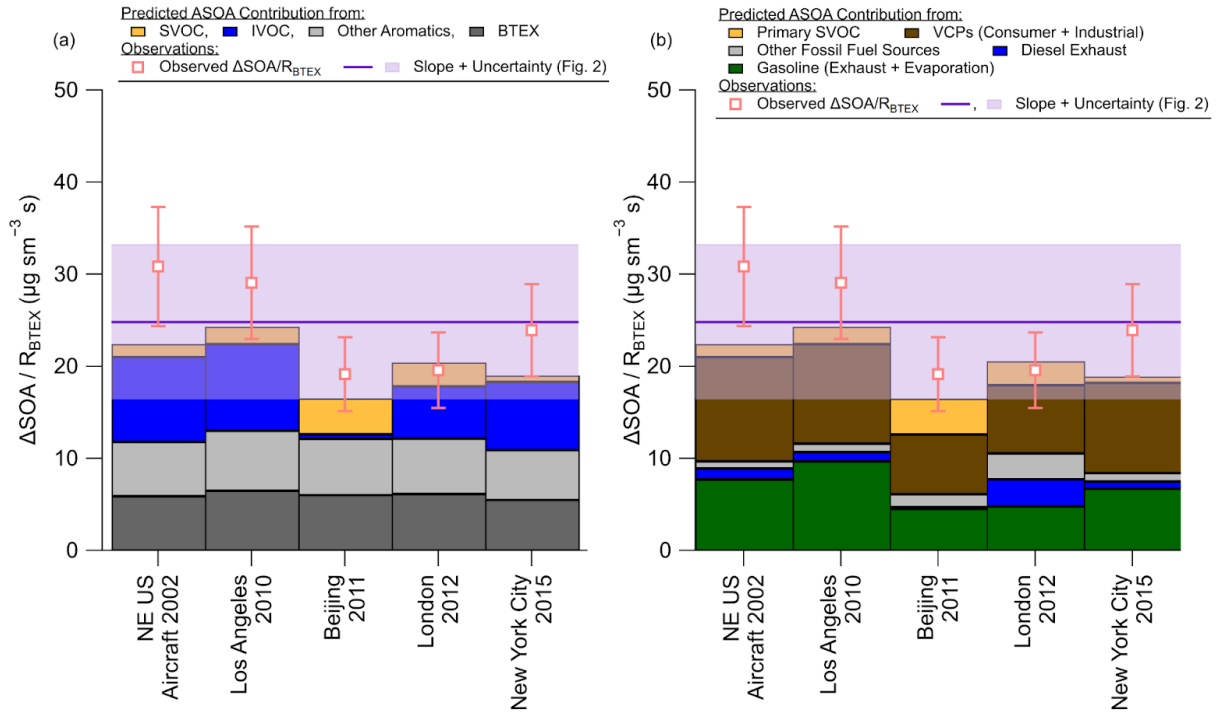
688 **Figure 2.** (a) Scatter plot of background and dilution corrected ASOA concentrations  
 689 ( $\Delta\text{SOA}/\Delta\text{CO}$  at PA = 0.5 equivalent days) versus BTEX emission reactivity ratio ( $R_{\text{BTEX}} =$   
 690  $\sum_i [\text{VOC}/\text{CO}]_i$ ) for multiple major field campaigns on three continents. Comparison of ASOA  
 691 versus (b) O<sub>x</sub>, (c) PAN, and (d) HCHO slopes versus the ratio of the BTEX/Total emission  
 692 reactivity, where total is the OH reactivity for the emissions of BTEX + C2-3 alkenes + C2-6  
 693 alkanes (Table S5 through Table S7), for the campaigns studied here. For all figures, red shading  
 694 is the  $\pm 1\sigma$  uncertainty of the slope, and the bars are  $\pm 1\sigma$  uncertainty of the data (see Sect. S5).

695

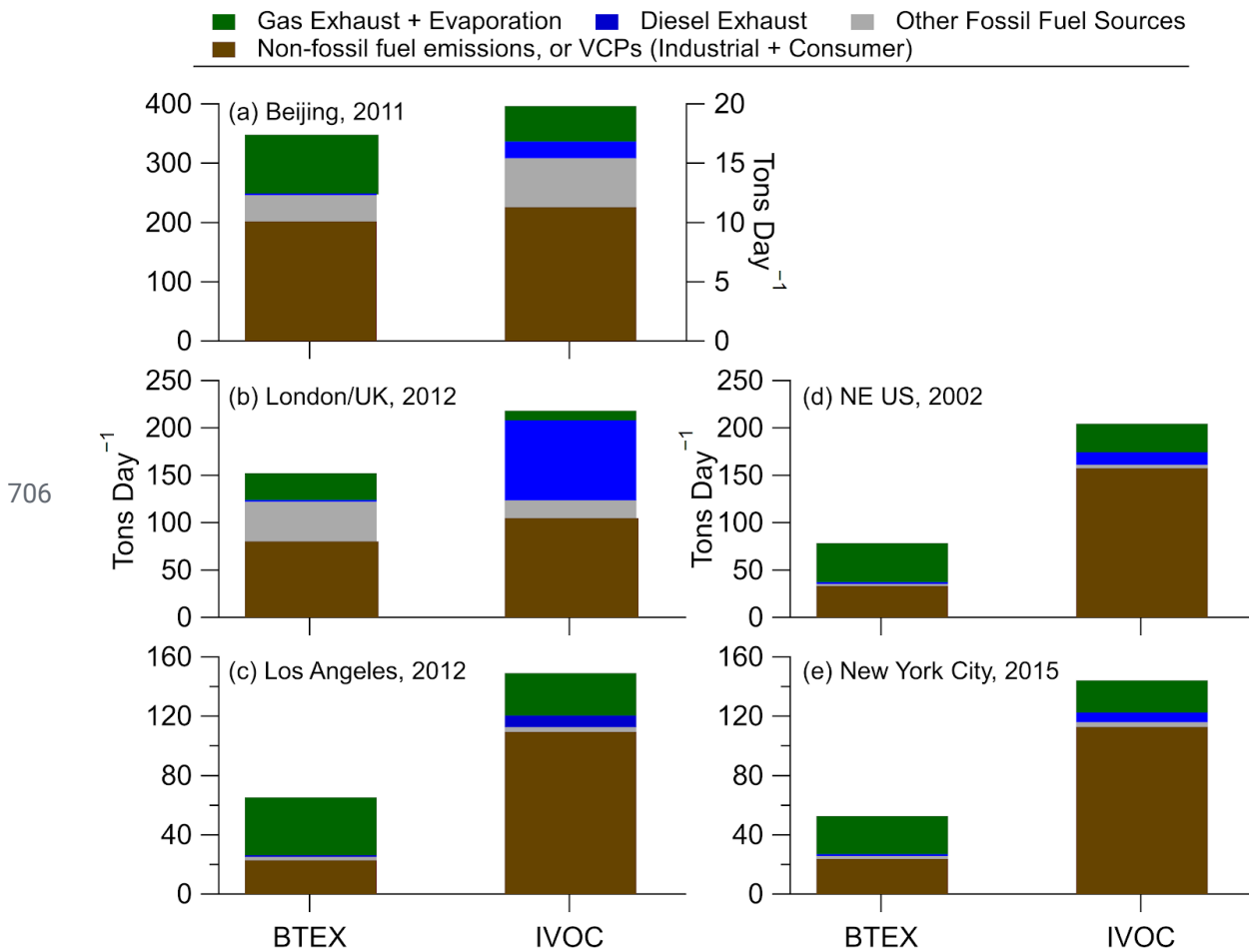


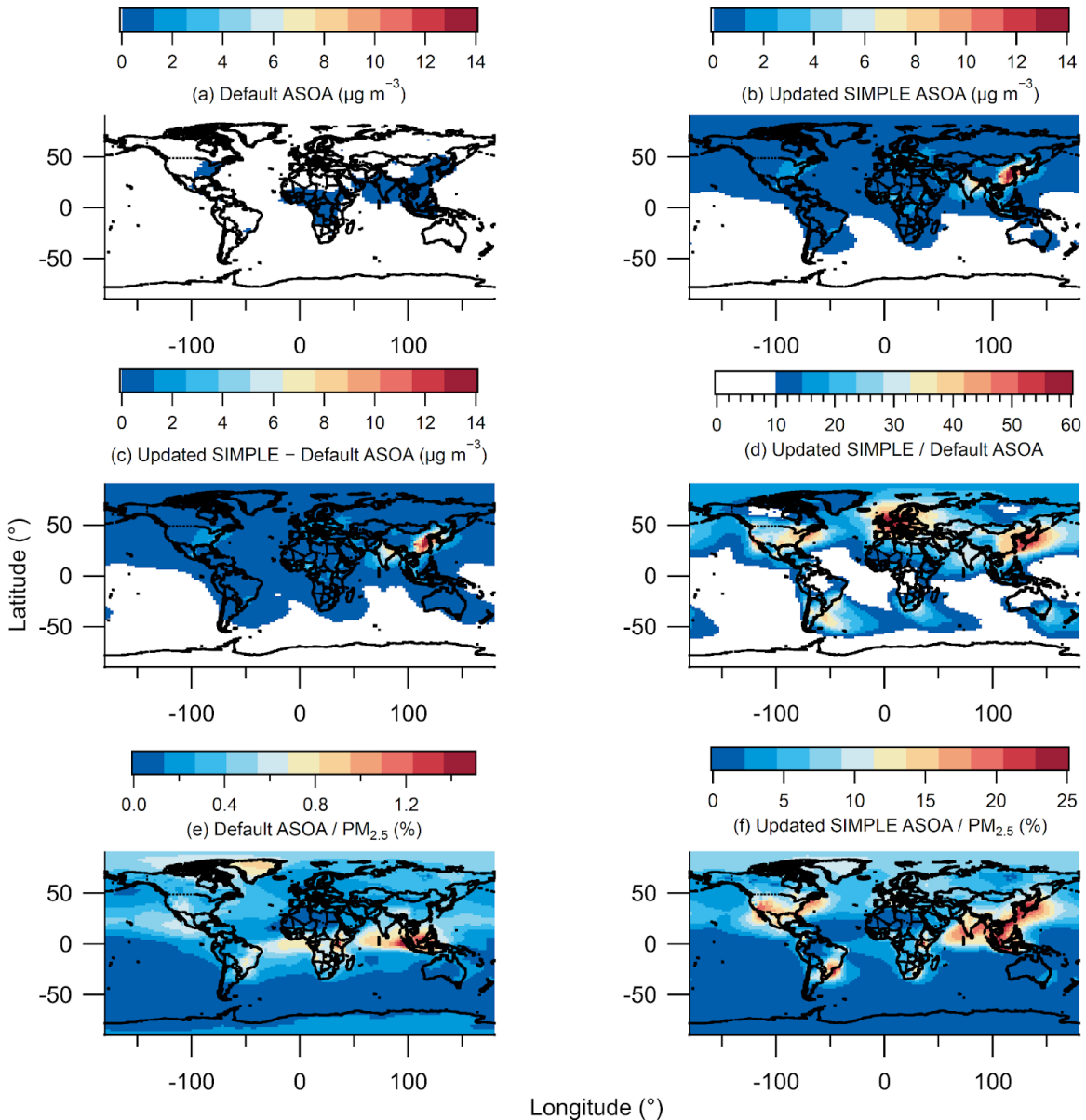
696 **Figure 3.** (a) A comparison of the  $\Delta\text{SOA}/\Delta\text{CO}$  for the urban campaigns on three continents.  
 697 Comparison of (b)  $\text{SOA}/\text{O}_x$ , (c)  $\text{SOA}/\text{HCHO}$ , and (d)  $\text{SOA}/\text{PAN}$  slopes for the urban areas  
 698 (Table S4). For (b) through (d), cities marked with \* have no HCHO, PAN, or hydrocarbon data.

699

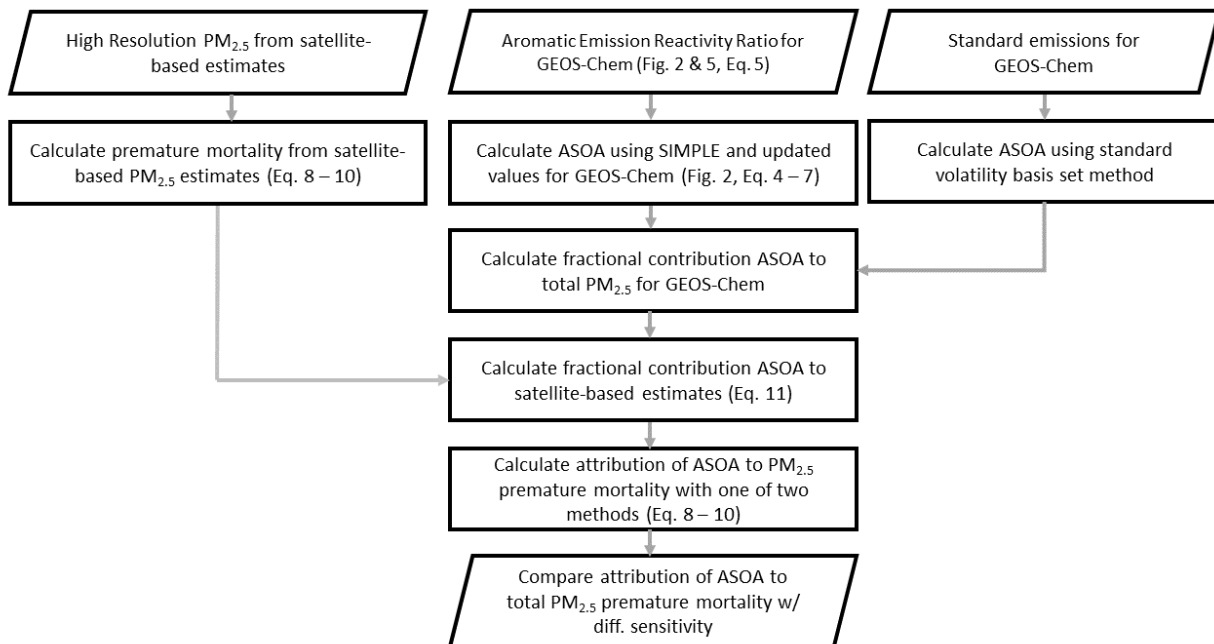


700 **Figure 4.** (a) Budget analysis for the contribution of the observed  $\Delta\text{SOA}/R_{\text{BTEX}}$  (Fig. 2) for cities  
 701 with known emissions inventories for different volatility classes (see SI and Fig. 5 and Fig. S6).  
 702 (b) Same as (a), but for sources of emissions. For (a) and (b), SVOC is the contribution from  
 703 both vehicle and other (cooking, etc.) sources. See SI for information about the emissions,  
 704 ASOA precursor contribution, error analysis, and discussion about sensitivity of emission  
 705 inventory IVOC/BTEX ratios for different cities and years in the US.



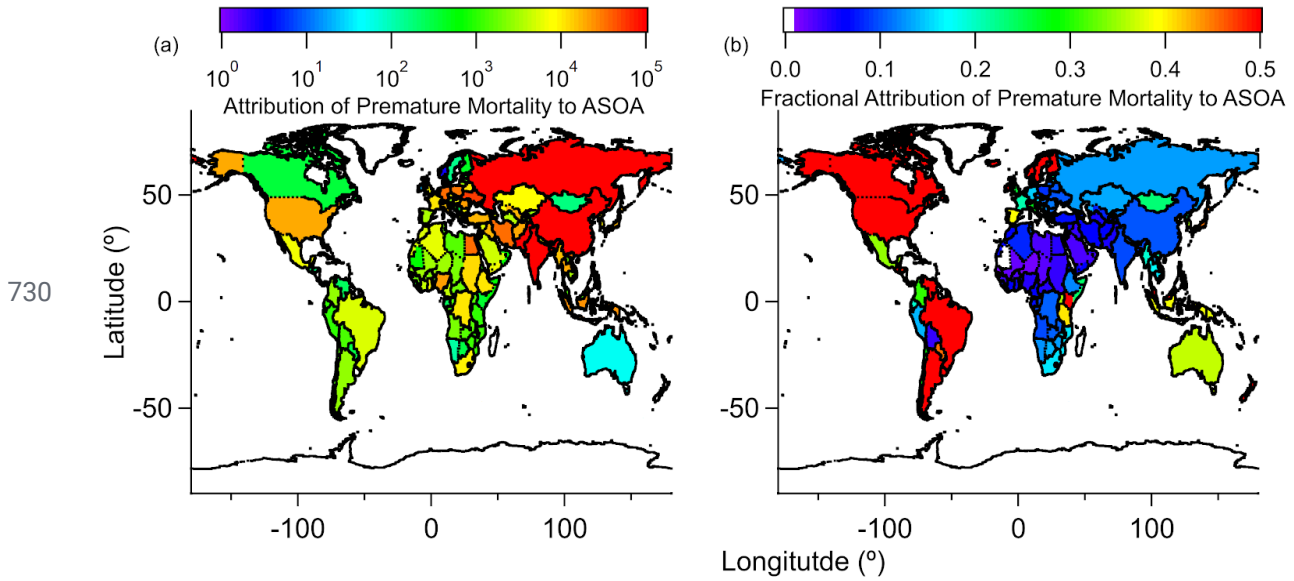


713 **Figure 6.** (a) Annual average modeled ASOA using the default VBS. (b) Annual average  
 714 modeled ASOA using the updated SIMPLE model. (c) Difference between annual average  
 715 modeled updated SIMPLE and default VBS. Note, for (a) - (b), values less than  $0.05 \mu\text{g m}^{-3}$  are  
 716 white, and for (c), values less than  $0.02 \mu\text{g m}^{-3}$  are white. (d) Ratio between annual average  
 717 modeled updated SIMPLE (b) and default VBS (a). (e) Percent contribution of annual average  
 718 modeled ASOA using default VBS to total modelled PM<sub>2.5</sub>. (f) Percent contribution of annual  
 719 average modeled ASOA using updated SIMPLE to total modelled PM<sub>2.5</sub>.

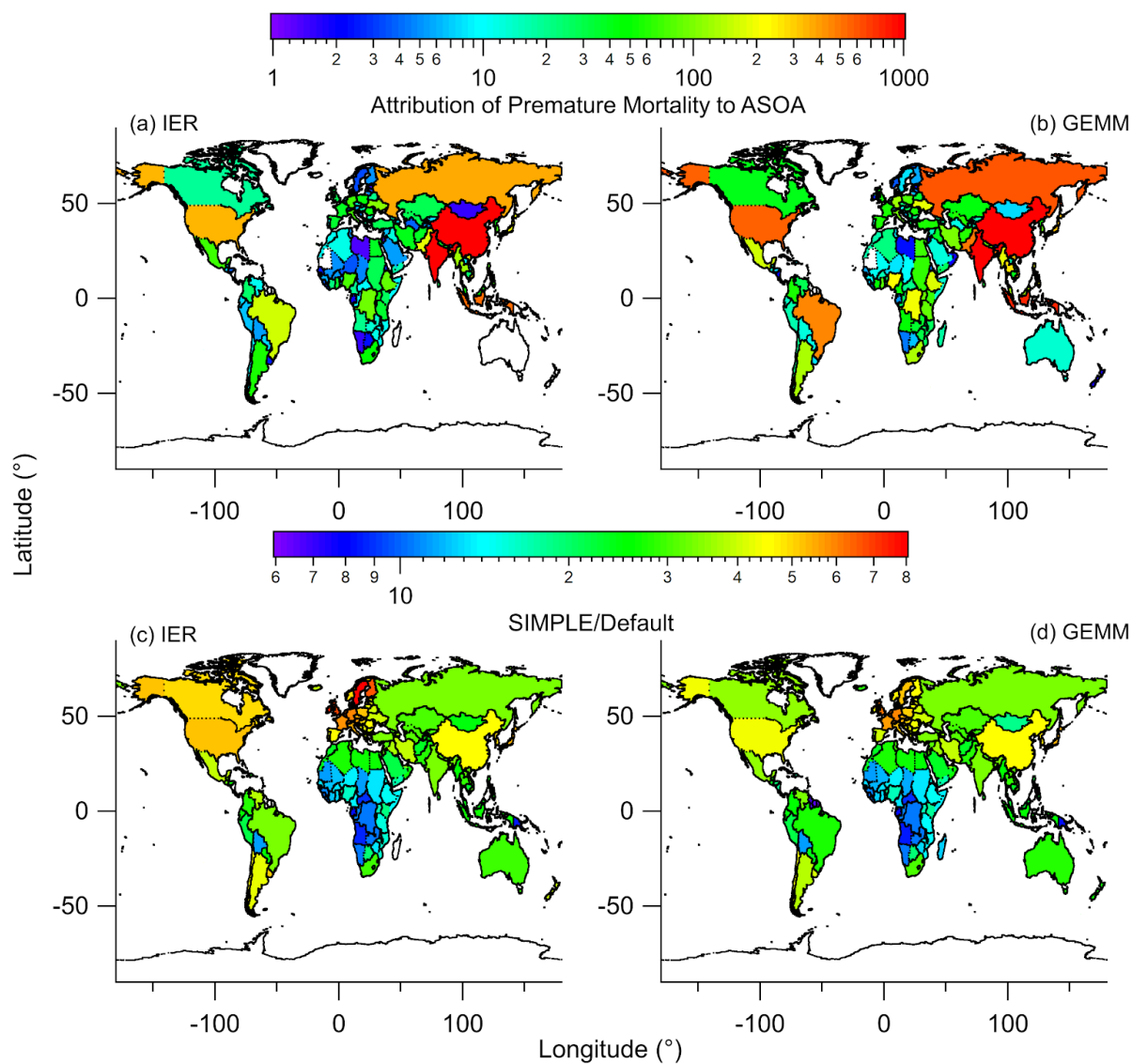


720

721 **Figure 7.** Flowchart describing how observed ASOA production was used to calculate ASOA in  
 722 GEOS-Chem, and how the satellite-based PM<sub>2.5</sub> estimates and GEOS-Chem PM<sub>2.5</sub> speciation was  
 723 used to estimate the premature mortality and attribution of premature mortality by ASOA. See  
 724 Sect. 2 and SI for further information about the details in the figure. SIMPLE is described in  
 725 Eq. 4 and by Hodzic and Jimenez (2011) and Hayes et al. (2015). The one of two methods  
 726 mentioned include either the Integrated Exposure-Response (IER) (Burnett et al., 2014) with  
 727 Global Burden of Disease (GBD) dataset (IHME, 2016) or the new Global Exposure Mortality  
 728 Model (GEMM) (Burnett et al., 2018) methods. For both IER and GEMM, the marginal method  
 729 (Silva et al., 2016) or attributable fraction method (Anenberg et al., 2019) are used.



731 **Figure 8.** Five-year average (a) estimated reduction in  $PM_{2.5}$ -associated premature deaths, by  
 732 country, upon removing ASOA from total  $PM_{2.5}$ , and (b) fractional reduction (reduction  $PM_{2.5}$   
 733 premature deaths / total  $PM_{2.5}$  premature deaths) in  $PM_{2.5}$ -associated premature deaths, by  
 734 country, upon removing ASOA from GEOS-Chem. The IER methods are used here. See Fig. S9  
 735 and Fig. S12 for results using GEMM. See Fig. S10 for  $10 \times 10$  km<sup>2</sup> area results in comparison  
 736 with country-level results.



738 **Figure 9.** Attribution of premature mortality to ASOA using (a) IER or (b) GEMM, using the  
 739 non-volatile primary OA and traditional SOA precursor method in prior studies (e.g., Ridley et  
 740 al., 2018). The increase in attribution of premature mortality to ASOA for the “SIMPLE” model  
 741 (Fig. 8) versus the non-volatile primary OA and traditional SOA precursor method (“Default”),  
 742 for (c) IER and (d) GEMM.



743 **Table 1.** List of campaigns used here. For values previously reported for those campaigns, they  
 744 are noted. For Seasons, W = Winter, Sp = Spring, and Su = Summer.

Location	Field Campaign	Coordinates		Time Period	Season	Previous Publication/Campaign Overview
		Long. (°)	Lat. (°)			
Houston, TX, USA (2000)	TexAQS 2000	-95.4	29.8	15/Aug/2000 - 15/Sept/2000	Su	Jimenez et al. (2009) <sup>a</sup> , Wood et al. (2010) <sup>b</sup>
Northeast USA (2002)	NEAQS 2002	-78.1 - -70.5	32.8 - 43.1	26/July/2002; 29/July/2002 - 10/Aug/2002	Su	Jimenez et al. (2009) <sup>a</sup> , de Gouw and Jimenez (2009) <sup>c</sup> , Kleinman et al. (2007) <sup>c</sup>
Mexico City, Mexico (2003)	MCMA-2003	-99.2	19.5	31/Mar/2003 - 04/May/2003	Sp	Molina et al. (2007), Herndon et al. (2008) <sup>b</sup>
Tokyo, Japan (2004)		139.7	35.7	24/July/2004 - 14/Aug/2004	Su	Kondo et al. (2008) <sup>a</sup> , Miyakawa et al. (2008) <sup>a</sup> , Morino et al. (2014) <sup>b</sup>
Mexico City, Mexico (2006)	MILAGRO	-99.4 - -98.6	19.0 - 19.8	04/Mar/2006 - 29/Mar/2006	Sp	Molina et al. (2010), DeCarlo et al. (2008) <sup>a</sup> , Wood et al. (2010) <sup>b</sup> , DeCarlo et al. (2010) <sup>c</sup>
Paris, France (2009)	MEGAPOLI	48.9	2.4	13/July/2009 - 29/July/2009	Su	Frenay et al. (2014) <sup>a</sup> , Zhang et al. (2015) <sup>b</sup>
Pasadena, CA, USA (2010)	CalNex	-118.1	34.1	15/May/2010 - 16/June/2010	Sp	Ryerson et al. (2013), Hayes et al. (2013) <sup>a,b,c</sup>
Changdao Island, China (2011)	CAPTAIN	120.7	38.0	21/Mar/2011 - 24/Apr/2011	Sp	Hu et al. (2013) <sup>a,c</sup>
Beijing, China (2011)	CareBeijing 2011	116.4	39.9	03/Aug/2011 - 15/Sept/2011	Su	Hu et al. (2016) <sup>a,b,c</sup>
London, UK (2012)	ClearfLo	0.1	51.5	22/July/2012 - 18/Aug/2012	Su	Bohnenstengel et al. (2015)
Houston, TX, USA (2013)	SEAC <sup>4</sup> RS	-96.0 - -94.0	29.2 - 30.3	01/Aug/2013 - 23/Sept/2013	Su	Toon et al. (2016)
New York City, NY, USA (2015)	WINTER	-74.0 - -69.0	39.5 - 42.5	07/Feb/2015	W	Schroder et al. (2018) <sup>a,c</sup>
Seoul, South Korea (2016)	KORUS-AQ	124.6 - 128.0	36.8 - 37.6	01/May/2016 - 10/June/2016	Sp	Nault et al. (2018) <sup>a,b,c,d</sup>

745 <sup>a</sup>Reference used for PM<sub>1</sub> composition. <sup>b</sup>Reference used for SOA/O<sub>x</sub> slope. <sup>c</sup>Reference used for  
 746 ΔOA/ΔCO value. <sup>d</sup>Reference used for SOA/HCHO and SOA/PAN slopes

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