

1 **Supplemental Information for:**
2 **Anthropogenic Secondary Organic Aerosols Contribute Substantially to Air Pollution**
3 **Mortality**
4
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6
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8 Section S1 US Emission Inventories

9 *Anthropogenic VOC emissions*

10 The US emissions of VOCs is based on a mass balance estimate of the petrochemical
11 industry reported by McDonald et al. (2018). Briefly, fuel sales and chemical product use are
12 estimated from publicly available reports on energy use, chemical production, economic surveys,
13 and freight shipments. Mobile source emission factors are from prior work quantifying both
14 on-road and off-road engines (McDonald et al., 2013, 2015). Evaporative sources of
15 transportation fuels are considered in addition to tailpipe exhaust (Pierson et al., 1999). VCP
16 emission factors are based on literature values, including from the indoor environment, and
17 reported in McDonald et al. (2018). Other fossil energy sources of VOCs, such as from oil
18 refineries and industry, are taken from official inventories reported by the California Air
19 Resources Board (CARB, 2013) or US Environmental Protection Agency (NEI, 2015).
20 McDonald et al. (2018) reported fossil-VOC emissions for the Los Angeles basin in the year
21 2010.

22

23 *Speciation of VOC emissions*

24 The total VOC emissions are speciated to estimate BTEX and IVOC emissions from
25 petrochemical VOC sources. Briefly, gasoline and diesel exhaust, gasoline fuel, and headspace
26 vapors are based on profiles reported in the literature from the Caldecott Tunnel (Gentner et al.,
27 2012, 2013). Speciation profiles of VCPs are based on California Air Resources Board surveys
28 of architectural coatings (Davis, 2007) and consumer products (CCPR, 2015). Other industrial

29 solvent uses and point/area source emissions are from the EPA SPECIATE (v4.4) database (EPA,
30 2014).

31

32 *Extrapolating IVOC/BTEX ratios from 2010 Los Angeles to other field campaigns*

33 In the ASOA mass closure estimation, three separate field campaigns are utilized from
34 the US: NEAQS 2002 (Boston/New York City), CalNex 2010 (Los Angeles), and WINTER
35 2015 (New York City outflow). These field campaigns span two megacities (Los Angeles and
36 New York City), ~one decade, and two seasons (summer versus winter). Here, we discuss how
37 each of these variables could affect the IVOC/BTEX emissions ratio. We focus the discussion on
38 mobile sources and VCPs because these are the dominant contributors to BTEX and IVOCs.

39 The IVOC/BTEX emissions ratio could be affected by the population density of a city. It
40 is well-established that per capita transportation fuel use decreases with increasing population
41 density (Gately et al., 2015), whereas VCP usage is expected to scale with population. Relative
42 to Los Angeles, the per capita fuel use in New York City is ~2 times lower (Gately et al., 2015),
43 resulting in lower on-road transportation VOC emissions relative to VCPs. Because aromatics
44 are mainly found in gasoline, whereas the IVOCs have a higher contribution from VCPs, the
45 IVOC/BTEX ratio is expected to be higher in New York City than Los Angeles.

46 To assess impacts of annual trends on the IVOC/BTEX ratio, we utilize long-term trend
47 analyses of mobile source VOC emissions in Los Angeles (McDonald et al., 2013, 2015; Hassler
48 et al., 2016). The main effect is that on-road gasoline emissions have decreased with time, both
49 from the tailpipe of vehicles (McDonald et al., 2013) and of gasoline-related VOCs in ambient
50 air measurements (Warneke et al., 2012). We utilize the EPA Trends Report to scale VOC

emissions for other anthropogenic sectors, including VCPs and industrial sources (<https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data>). The EPA Trends Report suggests that VCP (or solvent) emissions decreased by ~30% between 2002 and 2010, including efforts to reduce the VOC content of architectural coatings (Matheson, 2002). After 2010, the emissions have been slightly increasing, likely due to population growth. Because both mobile sources and VCP emissions are decreasing with time, the IVOC/BTEX emissions ratio is not significantly altered.

Lastly, the effects of seasonality influence on-road transportation emissions through: (i) increased VOC emissions in winter relative to summer from cold-starting engines, and (ii) lower evaporative emissions due to colder ambient temperatures. We estimate that exhaust emissions from passenger vehicles increases by ~50% due to higher cold-start emissions in winter relative to summer based on the EPA MOVES model (MOVES, 2015). Evaporated gasoline and headspace vapors are known to exhibit a temperature-dependence (Rubin et al., 2006), and estimated to be ~20% and ~80% lower, respectively, based on typical wintertime temperatures of New York City relative to summertime Los Angeles. Due to compensating factors between cold-start engines and evaporated fuels, the IVOC/BTEX emissions are not significantly affected by seasonality.

Overall, when taking into account differences in population density between Los Angeles and New York City, trends of mobile source and VCP emissions over time, and seasonality, the IVOC/BTEX emission ratios range between ~2.3 to 2.7, which is relatively small. This sensitivity analysis helps explain why the enhancement observed in SOA scales with BTEX levels in the urban atmosphere.

73

74 **Section S2 Beijing Emission Inventory**

75 *Anthropogenic VOC emissions*

76 The total VOC emissions of Beijing were developed following the bottom-up framework
77 of the Multi-resolution Emission Inventory for China (MEIC) model (available at
78 <http://www.meicmodel.org>), based on a technology-based methodology. The details of activity
79 rates, emission factors, technology distribution, and control measures configured in the MEIC
80 model are summarized in a series of papers (Zhang et al., 2009; Zheng et al., 2014, 2018; Liu et
81 al., 2015; Li et al., 2017, 2019).

82 In the MEIC model, a detailed four-level source classification system, representing
83 sector, fuel/product, technology/solvent type, and end-of-pipe pollutant abatement facilities, was
84 established by including over 700 emitting sources for each province. All anthropogenic sources,
85 including power plants, industrial sources, volatile chemical products, fossil fuel burning in
86 residential stoves, transportation were all considered.

87 Power plants are treated as point sources in the MEIC model. The VOC emissions were
88 derived from the China coal-fired Power Plant Emissions Database (CPED, (Liu et al., 2015)),
89 which is developed based on information of each unit on fuel type, fuel quality, combustion
90 technology, etc.

91 Volatile chemical products are comprised of solvent use applied for architecture, vehicles,
92 wood, and other industrial purposes, glue use, printing, pesticide use, and domestic solvent use.
93 The market share of waterborne and solvent-based paint is further taken into account for each
94 source category. For the on-road transportation sector, the improved emissions developed by

95 Zheng et al. (2014) were integrated into the framework of MEIC, which estimated the vehicle
96 population and emission factors at a county level. Both the VOC emissions in running mode and
97 evaporation were considered. Emission standards covering pre-Euro I and Euro I to Euro V in
98 Beijing were applied for each vehicle type (Zheng et al., 2018; Li et al., 2019). Regarding
99 oxygenated volatile organic compounds (OVOCs), the emission factors for on-road vehicles
100 were corrected, as current emission factors are only for non-methane hydrocarbons (NMHC).
101 Correction ratios of 1.32, 1.08, 1.10, and 1.06 were applied for heavy-duty and light-duty diesel
102 vehicles, and heavy-duty and light-duty gasoline vehicles, respectively, to the original values to
103 comply with the follow-up speciation for the total VOC, following the method of Li et al. (2014,
104 2019).

105

106 *Speciation of VOC emissions*

107 Emissions by individual chemical species were developed based on the
108 profile-assignment approach (Li et al., 2014, 2019). First, a “composite” profile database for
109 China was established by integrating the local profiles and supplementing it with the SPECIATE
110 v4.5 database for absent sources ((Simon et al., 2010), available at:
111 <https://www.epa.gov/air-emissions-modeling/speciate-version-45-through-40>). The detailed
112 procedure for developing the composite profile database is illustrated in Li et al. (2014). In brief,
113 for sources where there are significant differences in technology or legislation between China
114 and western countries, only local profiles are used; otherwise, all candidate profiles are included
115 for further compilation in the composite profile database. Local profiles covering most of the
116 important sources were gathered and reviewed, including biofuel combustion, coal combustion,

117 asphalt production, oil production, refinery, paint use, gasoline evaporation, gasoline vehicle
118 exhaust, diesel vehicles, and so on, as detailed illustrated in Li et al. (2019).

119 Then, profiles for all combustion-related sources, including fossil fuel combustion in
120 power plants, industry, residential, and transportation sectors were reviewed, and incomplete
121 profiles that were absent from the OVOC fractions were corrected by appending the component
122 of “OVOC” with fractions derived from the “complete” profiles for the same source. After
123 OVOC correction, all “candidate” profiles were averaged by species to establish the composite
124 profile database. Finally, the composite profile to each source was assigned by setting up the
125 source linkage between the profile database and the inventory. Emissions by individual chemical
126 species for each source were then further developed.

127

128 **Section S3 London/United Kingdom Emission Inventory**

129 *Anthropogenic VOC emissions*

130 The National Atmospheric Emissions Inventory (NAEI) estimates UK emissions of
131 VOCs from anthropogenic sources following methods in the EMEP/EEA Emissions Inventory
132 Guidebook (EMEP/EEA, 2016) for submission under the revised EU Directive 2016/2284/EU on
133 National Emissions Ceilings (NECD), available at:
134 <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32016L2284&from=EN>, and
135 the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range
136 Transboundary Air Pollution (CLRTAP), available at:
137 http://www.ceip.at/ms/ceip_home1/ceip_home/reporting_instructions/reporting_programme/.
138 The NECD and CLRTAP define those VOC sources to be included and excluded from the

139 national inventory (for example, emissions of NMVOCs from biogenic sources are not included).
140 The Guidebook provides estimation methodologies and default emission factors for each source
141 category, although countries can use country-specific emission factors where these are deemed
142 relevant. The NAEI currently covers organic emissions from around 400 individual source
143 categories, with a large contribution from a diverse range of industrial processes and solvents,
144 but with very few individually dominant sources. The inventory then speciates emissions into
145 ~650 individual compounds, or groups of compounds. Groupings of organics, for example,
146 expressed as ‘sum of all C14 compounds,’ make up a substantial fraction of IVOC emissions,
147 rather than being reported as individual compounds.

148 Emissions from the use of solvents and other volatile chemicals in industry and in
149 consumer products, fuel production and distribution, food and drink manufacture and other
150 non-combustion industrial processes accounted for 72% of all UK NMVOC emissions in 2017,
151 according to the NAEI. Both the solvent and industrial process sectors cover a diverse range of
152 emission source categories: the NAEI identifies 136 separate categories across the two sectors

153 For the road transport sector, the NAEI reports exhaust emissions of NMVOCs and its
154 emissions from evaporative losses of fuel vapor from petrol vehicles. Emissions from re-fueling
155 at filling stations are reported separately under the fugitive emissions from the fuel distribution
156 sector. The method used for road transport in the NAEI follows the method in the European
157 COPERT 5 model and described in the EMEP/EEA Emissions Inventory Guidebook. The
158 method uses average speed-related emission factors for hot exhaust emissions of total
159 hydrocarbons for detailed vehicle categories (vehicle type, weight and/or engine size) and Euro
160 standards for petrol cars, diesel cars, petrol and diesel light goods vehicles, rigid and articulated

161 HGVs, buses and coaches, and mopeds and motorcycles, and combines these with detailed traffic
162 and fleet activity data derived from information provided by DfT. Separate estimates are made of
163 methane emissions for each vehicle type and subtracted from the THC emissions to derive the
164 NMVOC emissions.

165 Evaporative emissions from vehicles are estimated in the NAEI, using the Guidebook
166 method for three different processes: diurnal losses, hot soak, and running losses. Emissions are
167 dependent on ambient temperature and fuel vapor pressure and different factors are provided for
168 vehicles with and without carbon canisters for evaporative emission controls. All vehicles from
169 Euro 1 onwards are fitted with these devices; so, evaporative emission have been decreasing
170 from the early 1990s with the penetration of these vehicles in the fleet. The method also takes
171 into account the reduction in Reid Vapour Petrol of petrol sold in the UK since 2000, as required
172 for compliance with the EU Fuel Quality Directive 98/70/EC, amended by Directive
173 2009/30/EC.

174

175 *Speciation of VOC emissions*

176 The NAEI is considered to adequately reflect annual real world emissions of BTEX (see,
177 for example, eddy covariance flux comparisons in London by Langford et al. (2010) and Vaughn
178 et al. (2017)); so, those values are taken directly from the NAEI and used here. IVOCs, and
179 particularly long chain hydrocarbons, are included in many cases in the inventory as groups, but
180 their emissions are known to be significantly underestimated when compared against field
181 observations. We use the observations of Dunmore et al. (2015), made in wintertime central
182 London in 2012, as guide to uprate NAEI emissions for IVOC species based on the estimated

183 discrepancies between inventory and field observation reported for each carbon number above
184 C10. This leads to some significant multipliers being applied to the inventory values, sometimes
185 of the order 60 to 70. We assume that the same multipliers apply to all sources, since field data
186 does not provide any means to attribute different factors to road transport IVOCs compared with
187 IVOCs from VCP sources.

188 Since the NAEI represents a reporting of emissions for the purposes of compliance with
189 international treaties, some fraction of those emissions are not released on the mainland UK. For
190 this paper, offshore BTEX and IVOC emissions, arising for example from offshore oil and gas
191 activity, aircraft in cruise, or shipping and emissions associated with overseas Crown
192 Dependencies are removed from the UK total, since they play no part in determining the
193 chemical environment of London. The annual NAEI totals are then divided equally to give a
194 daily national emission.

195

196 **S4 Ozone Sensitivity to ASOA Simulations**

197 A potential issue in the attribution of premature mortality to AOSA is that reducing
198 emissions that lead to ASOA is that this may impact ozone concentrations. A sensitivity analysis
199 was conducted, where the ASOA emissions were reduced by 20% (Fig. S12). In general, there is
200 a less than 1% reduction in total ozone concentration in the boundary layer. This is due to the
201 fact that the most important AVOCs that contribute to ozone formation are light alkenes (e.g.,
202 ethylene and propylene, Fig. 5), which are not ASOA precursors. Though the reaction rate
203 constant of the ASOA precursors is generally high (Table S11), the concentration of the
204 precursors is low and they thus account for a low percentage of the total ozone production

205 potential (Table S5 through Table S9). For example, the measured OH reactivity (Sect. 3) for two
206 different urban regions was between 15 to 25 s⁻¹ (Griffith et al., 2016; Whalley et al., 2016)
207 while the OH reactivity for the ASOA precursors for the same region was between 2 to 4 s⁻¹. The
208 small contribution to the OH reactivity is in line to the minimal impact to the ozone
209 concentration observed in Fig. S12.

Supporting Information Tables

Table S1. List of instruments whose observations are used in this study. In some cases $\Delta\text{SOA}/\Delta\text{CO}$ (Table S4), SOA versus O_x slope (Table S4), or VOC emission ratios (Table S5 through Table S8) had already been reported, and, in those cases, we use the previous literature reports in our analyses.

Location	SOA	O_x	HCHO	PAN	VOCs	CO
Houston, TX, USA (2000)	Q-AMS ^a	CL & UV Absorption ^b	DOAS ^c	GC-ECD ^d	GC-FID, GC-MS ^e	Infrared Absorption ^f
Mexico City, Mexico (2006)	HR-ToF-AMS ^g	CL ^h	TDLAS ⁱ	CIMS ^j	WAS ^k	UV RF ^l
Los Angeles, CA, USA (2010)	HR-ToF-AMS ^g	CL & UV Absorption ^m	Average of DOAS ^c & Hantzsch Reaction ⁿ	GC-ECD ^d	GC-MS ^o	UV RF ^l
Beijing, China (2011)	HR-ToF-AMS ^g	CL & UV Absorption ^p	PTR-MS ^q	GC-ECD ^r	GC-FID ^s	IR Absorption ^p
London, UK (2012)	C-ToF-AMS ^t	CL & UV Absorption ^u	Hantzsch Reaction ⁿ	GC-ECD ^v	GC-FID & GC \times GC-FID ^w	UV RF ^l
Houston, TX, USA (2013)	HR-ToF-AMS ^g	CL ^x	Average of LIF ^y & CAMS ^z	CIMS ^j	WAS ^k	DACOM ^{aa}
Seoul, South Korea (2016)	HR-ToF-AMS ^g	CL ^h	CAMS ^z	CIMS ^j	WAS ^k	DACOM ^{aa}

^aQuadrupole Aerosol Mass Spectrometer (Q-AMS) (Jayne et al., 2000)

^bChemiluminescence (CL) and UV Absorption (Williams et al., 1997)

^cDifferential Optical Absorption Spectrometry (DOAS) (Stutz and Platt, 1996, 1997)

^dGas chromatography-electron capture detector (GC-ECD) (Williams et al., 2000; Roberts et al., 2002)

^eGas chromatography-flame ionization detector (GC-FID) and gas chromatography mass spectrometer (Roberts et al., 2001)

^fTECO Model 48s IR gas-filter

^gHigh Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) (DeCarlo et al., 2006)

^hChemiluminescence (CL) and UV Absorption (Weinheimer et al., 1994)

ⁱTunable diode laser absorption spectroscopic (TDLAS) measurements (Fried et al., 2003)

^jChemical ionization mass spectrometer (CIMS) (Huey L Tanner D Slusher D Dibb J Arimoto R Chen G Davis D Buhr M Nowak J Mauldin R Eisele F, 2004; Slusher et al., 2004; Kim et al., 2007)

^kWhole air sample, followed by analysis with GC-FID and/or GC-MS (Blake et al., 2003)

^lUV Resonance Fluorescence (RF) (Gerbig et al., 1999)

233 ^mChemiluminescence (CL) and UV Absorption (Hayes et al., 2013)
 234 ⁿHantzsch reaction (Cárdenas et al., 2000)
 235 ^oGas chromatograph mass spectrometer (Gilman et al., 2010)
 236 ^pChemiluminescence (CL), UV Absorption, and IR Absorption (Hu et al., 2016)
 237 ^qProton transfer reaction mass spectrometer (PTR-MS) (Warneke et al., 2011)
 238 ^rGas chromatography electron capture detector (GC-ECD) (Zhang et al., 2017)
 239 ^sGas chromatography flame ionization detector (GC-FID) (Wang et al., 2014)
 240 ^tCompact Time-of-Flight Aerosol Mass Spectrometer (C-ToF-AMS) (Drewnick et al., 2005)
 241 ^uChemiluminescence (CL) and UV Absorption (Whalley et al., 2016)
 242 ^vGas chromatography electron capture detector (GC-ECD) (Whalley et al., 2016)
 243 ^wGas chromatography flame ionization detector (GC-FID) (Dunmore et al., 2015)
 244 ^xChemiluminescence (CL) (Ryerson et al., 1999; Pollack et al., 2010)
 245 ^yLaser induced fluorescence (LIF) (Cazorla et al., 2015)
 246 ^zCompact Atmospheric Multi-species Spectrometer (CAMS) difference frequency absorption
 247 spectrometer (Weibring et al., 2010)
 248 ^{aa}Tunable diode laser absorption spectroscopy (Sachse et al., 1987)

249 **Table S2.** Concentrations of PM₁ components shown in Fig. 1. References for the measurements
 250 can be found in Table 1.

Dataset Location	Average Concentration ($\mu\text{g sm}^{-3}$) of submicron aerosol under standard temperature and pressure				
	SOA	HOA	SO ₄	NO ₃	NH ₄
Houston, TX, USA (2000)	2.7	0.7	4.9	0.4	1.5
Northeast USA (2002)	4.9	0.5	2.0	0.3	0.7
Tokyo, Japan (2004)	6.0	1.5	4.4	0.9	4.0
Mexico City, Mexico (2006)	11.2	4.8	1.9	6.0	2.5
Paris, France (2009)	1.9	1.1	1.2	0.5	0.6
Los Angeles, CA, USA (2010)	5.0	2.0	2.9	3.6	2.1
Changdao Island, China (2011)	9.4	4.4	8.3	12.2	6.5
Beijing, China (2011)	17.1	8.9	22.0	16.8	13.7
London, UK (2012)	2.7	1.6	1.4	2.7	1.3
Houston, TX, USA (2013)	3.7	0.0	2.7	0.1	0.6
New York City, NY, USA (2015)	0.8	0.7	1.2	1.4	0.4
Seoul, South Korea (2016)	11.9	1.3	5.0	7.9	4.4

251

252

Table S3. Table summarizing the results of recent GEOS-Chem performance evaluations for modeling BSOA.

Study	Observed Data	Species	Details
Fisher et al. (2016) ^a	SEAC ⁴ RS, below 1 km (spatial pattern), below 500 m (bias)	Isoprene	Spatial patterns well captured, and biases are +34% for isoprene and +3% for monoterpenes
		Monoterpene	
		Organic Nitrates from Isoprene	Spatial patterns well captured, and biases are -0.6% for first- and -35% for second-generation isoprene nitrates
	SEAC ⁴ RS, 0 - 4 km vertical profiles	Isoprene	Agreed well but GEOS-Chem somewhat overestimated observed concentrations near 1km
		Monoterpene	
		HCHO	
	SOAS, at the surface	Organic Nitrates from Isoprene	Agreed within measurement uncertainties
		Isoprene	Underestimated isoprene and monoterpenes (-28% and -54%), but overestimated first- and second- generation isoprene nitrates (+85% and +43%)
		Monoterpene	
		HCHO	
		Organic Nitrates from Isoprene	
Travis et al. (2016)	SEAC ⁴ RS, 0 - 12 km	First Generation from Isoprene Nitrates	Good agreement for ISOPOOH and ISOPN, underestimation of HPALDs by a factor of two
		ISOPOOH	
		HPALDS	
Marais et al. (2016)	SOAS, at the surface	IEPOX-SOA	Good agreement for isoprene derived aerosols, mean concentrations were almost the same
		ISOPOOH-SOA	
	SEAC ⁴ RS, below 2 km (spatial pattern)	IEPOX-SOA	Spatial patterns well captured

^aThis study decreased isoprene emissions by 15% and doubled monoterpene emissions of MEGANv2.1.

257 **Table S3 cont.**

Study	Observed Data	Species	Details
Kaiser et al. (2018) ^a	SEAC ⁴ RS	Isoprene	All were overestimated, except for first generation isoprene nitrates
		HCHO	
		ISOPOOH	
		MVK + MACR	
		First Generation Isoprene Nitrates	
Pai et al. (2020)	15 airborne campaigns (SEAC ⁴ RS, GoAmazon, SENEX, OP3, etc.)	OA under biogenic dominant conditions	Slight overestimation, but generally very similar in magnitude

258 ^aNEI NO_x emissions other than power plants decreased by 60%, soil NO_x emissions were
259 reduced by 50% across the Midwestern US. With the decrease of NO_x emissions, ISOPOOH
260 concentrations were increased in GEOS-Chem.

Table S4. Dilution-corrected SOA concentrations at 0.5 equivalent days and slopes of SOA versus O_x, HCHO, and PAN used in Fig. 4 and Fig. 5. References for the values can be found either in Table 1 or found in Fig. S2 through Fig. S4. Uncertainty is 1σ, and either represents propagation in uncertainty in measurements (see Sect. 2 Main Paper) for ΔSOA/ΔCO or uncertainty in slopes for SOA versus the three photochemical species.

Dataset Location	ΔSOA/ΔCO at 0.5 eq. days	SOA vs. O _x Slopes	SOA vs. HCHO Slopes	SOA vs. PAN Slopes
Houston, TX, USA (2000)		0.04±0.01 ^a	0.32±0.08	1.41±0.46
Northeast USA (2002)	16±3 ^b 48±9 ^c			
Mexico City, Mexico (2003)		0.14±0.01 ^a		
Tokyo, Japan (2004)		0.19±0.01 ^a		
Mexico City, Mexico (2006)	58±10	0.16±0.01	1.60±0.06	5.60±0.30
Paris, France (2009)		0.14±0.01 ^a		
Pasadena, CA, USA (2010)	59±11	0.16±0.01	1.93±0.02	5.41±0.12
Changdao Island, China (2011)	23±4			
Beijing, China (2011)	31±6	0.21±0.01	3.90±0.15	7.42±0.46
London, UK (2012)	54±10	0.13±0.01	0.36±0.02	3.37±0.41
Houston, TX, USA (2013)		0.16±0.01	1.52±0.13	6.92±0.58
New York City, NY, USA (2015)	33±6			
Seoul, South Korea (2016)	107±19	0.29±0.02	3.73±0.26	10.13±0.52

^aMissing reported uncertainty; therefore, assuming ±0.01, as that is typical for other campaigns

^bFrom de Gouw et al. (2005). ^cFrom Kleinman et al. (2007).

268 **Table S5.** Emission ratios of BTEX aromatics used in this study. If no reference is listed, then
 269 the emission ratio was calculated using Eq. 3.

Dataset Location	Emission Ratios (ppbv aromatic/ppmv CO)					References
	Benzene	Toluene	Ethylbenzene	m+p-xylene	o-xylene	
Houston, TX, USA (2000)	2.6	3.5	0.6	2.8	0.8	
NE USA, Ship (2002)	0.9	2.0	0.2	0.6	0.3	Baker et al. (2008)
NE USA, Aircraft (2002)	0.8	2.9	0.4	1.2	0.5	Warneke et al. (2007)
Mexico City, Mexico (2006)	0.9	7.5	0.9	1.1	0.4	Apel et al. (2010)
Los Angeles, CA, USA (2010)	1.3	3.4	0.6	2.1	0.8	de Gouw et al. (2017)
Changdao Island, China (2011)	2.3	1.9	0.5	1.3	0.4	Yuan et al. (2013)
Beijing, China (2011)	1.2	2.4	1.0	1.6	0.6	Wang et al. (2014)
London, UK (2012)	1.8	6.3	1.2	2.2	1.1	
Houston, TX, USA (2013)	2.3	3.0	0.6	3.9	1.2	
New York City, NY, USA (2015)	0.8	2.9	0.4	1.2	0.5	Warneke et al. (2007) ^a
Seoul, South Korea (2016)	1.1	13.1	2.4	3.3	2.3	

270 ^aUsing the emissions from Warneke et al. (2007) instead of Schroder et al. (2018) as Schroder et
 271 al. found significant uncertainty in the emissions calculated from observations.

272 **Table S6.** Emission ratios of alkanes used in this study. If no reference is listed, then the
 273 emission ratio was calculated using Eq. 3.

Dataset Location	Emission Ratios (ppbv alkane/ppmv CO)							References
	Ethane	Propane	n-Butane	i-Butane	n-Pentane	i-Pentane	n-Hexane	
Houston, TX, USA (2000)	40.9	24.3	9.0	14.7	3.1	10.0	3.1	
NE USA, Ship (2002)	8.3	2.3	1.8	1.3	1.0	2.8	0.9	Baker et al. (2008)
NE USA, Aircraft (2002)	9.9	9.0	2.4	1.3	2.0	5.4	0.6	Warneke et al. (2007)
Mexico City, Mexico (2006)	7.4	41.5	15.1	4.8	2.1	2.7	1.5	Apel et al. (2010)
Los Angeles, CA, USA (2010)	16.5	13.4	5.0	3.2	3.4	8.7	1.4	de Gouw et al. (2017)
Changdao Island, China (2011)	7.7	4.5	2.5	1.2	1.0	1.5	0.5	Yuan et al. (2013)
Beijing, China (2011)	4.3	3.9	2.5	2.5	1.2	2.0	0.6	Wang et al. (2014)
London, UK (2012)	33.0	17.8	17.3	8.4	4.6	11.3	1.3	
Houston, TX, USA (2013)	86.5	37.3	14.6	10.6	7.0	10.5	3.0	
Seoul, South Korea (2016)	16.1	0.4	6.0	3.4	3.1	3.7	1.7	

274

275 **Table S7.** Emission ratios of alkenes used in this study. If no reference is listed, then the
 276 emission ratio was calculated using Eq. 3.

Dataset Location	Emission Ratios (ppbv alkene/ppmv CO)		References
	Ethene	Propene	
Houston, TX, USA (2000)	24.4	28.4	
NE USA, Ship (2002)	4.4	1.1	Baker et al. (2008)
NE USA, Aircraft (2002)	4.9	1.4	Warneke et al. (2007)
Mexico City, Mexico (2006)	8.4	2.6	Apel et al. (2010)
Los Angeles, CA, USA (2010)	11.2	4.1	de Gouw et al. (2017)
Changdao Island, China (2011)	5.3	1.4	Yuan et al. (2013)
Beijing, China (2011)	4.4	1.4	Wang et al. (2014)
London, UK (2012)	10.3	6.2	
Houston, TX, USA (2013)	12.0	15.8	
Seoul, South Korea (2016)	5.4	2.1	

277

278 **Table S8.** Emission ratios of non-BTEX aromatics used in this study. If no reference is listed,
 279 then the emission ratio was calculated using Eq. 3.

Dataset Location	Emission Ratios (ppbv aromatic/ppmv CO)			References
	Trimethylbenzenes	Ethyltoluenes	Propylbenzene	
NE USA, Aircraft (2002)	0.71	0.58	0.14	Warneke et al. (2007)
Los Angeles, CA, USA (2010)	1.47	0.56	0.13	de Gouw et al.(2017)
Beijing, China (2011)	0.57	0.41	0.09	Wang et al. (2014)
London, UK (2012)	0.49	0.23	0.58	
New York City, NY, USA (2015)	0.71	0.58	0.14	Warneke et al. (2007)

280

281

282 **Table S9.** Normalized mass concentration of primary organic aerosol (POA) measured in various
 283 campaigns, used to determine SVOC emission ratios.

Dataset Location	Normalized Mass Concentration ($\mu\text{g sm}^{-3}$)		References
	HOA	Other POA	
NE USA (2002)	12.2	-	de Gouw et al. (2005)
Los Angeles, CA, USA (2010)	5.3	7.7	Hayes et al. (2013)
Beijing, China (2011)	6.1	9.9	Hu et al. (2016)
London, UK (2012)	17.9	14.1	Young et al. (2015)
New York City, NY, USA (2015)	5.6	14.4	Schroder et al. (2018)

284

285 **Table S10.** Sensitivity analysis of slopes and R^2 by removing one city each. All slopes are
 286 statistically similar at the 95% confidence interval.

Dataset Location	Slope ($\mu\text{g sm}^{-3} \text{ s}$)	R^2
All	24.8 \pm 3.4	0.88
Without Seoul	34.0 \pm 10.5	0.64
Without NYC	24.8 \pm 3.8	0.8
Without London	24.2 \pm 3.4	0.90
Without Beijing	24.2 \pm 3.5	0.89
Without Chinese Outflow	24.0 \pm 3.6	0.88
Without LA	24.2 \pm 3.3	0.90
Without Mexico City	24.2 \pm 3.4	0.90
Without NE US Aircraft	24.8 \pm 2.9	0.92
Without NE US, Boat	24.2 \pm 4.0	0.86

287

288 **Table S11.** Rate constants used throughout this study.

Compound	Rate Constant (cm ³ molec. ⁻¹ s ⁻¹)	References
<i>Alkanes</i>		
Ethane	$6.9 \times 10^{-12} \times \exp(-1000/T)$	Atkinson et al. (2006)
Propane	$7.6 \times 10^{-12} \times \exp(-585/T)$	Atkinson et al. (2006)
n-Butane	$9.8 \times 10^{-12} \times \exp(-425/T)$	Atkinson et al. (2006)
i-Butane	$1.17 \times 10^{-17} \times T^2 \times \exp(213/T)$	Atkinson and Arey (2003)
n-Pentane	$2.52 \times 10^{-17} \times T^2 \times \exp(158/T)$	Atkinson and Arey (2003)
i-Pentane	3.6×10^{-12}	Atkinson and Arey (2003)
n-Hexane	$2.54 \times 10^{-14} \times T \times \exp(-112/T)$	Atkinson and Arey (2003)
<i>Alkenes</i>		
Ethene	$7.84 \times 10^{-12,a}$	Atkinson et al. (2006)
Propene	$2.86 \times 10^{-11,a}$	Atkinson et al. (2006)
<i>Aromatics</i>		
Benzene	$2.3 \times 10^{-12} \times \exp(-190/T)$	Atkinson et al. (2006)
Toluene	$1.8 \times 10^{-12} \times \exp(340/T)$	Atkinson et al. (2006)
Ethylbenzene	7×10^{-12}	Atkinson and Arey (2003)
m+p-xylene	$1.87 \times 10^{-11,b}$	Atkinson and Arey (2003)
o-xylene	1.36×10^{-11}	Atkinson and Arey (2003)
Trimethylbenzenes	$2.73 \times 10^{-12} \times \exp(730/T)$	Bohn and Zetzsch (2012)
Ethyltoluenes	1.2×10^{-11}	Atkinson and Arey (2003)
Propylbenzene	5.8×10^{-12}	Atkinson and Arey (2003)
NO _x /NO _y		
OH + NO ₂	$1.23 \times 10^{-11,a}$	Mollner et al. (2010)

289 ^aShowing the rate constant at 298 K, 1013 hPa. However, for this study, we used the temperature
290 and pressure dependent formulation listed in each respective reference.

291 ^bThis is the average of m-xylene and p-xylene rate constants.

292 **Table S12.** Table of GBD parameters, which is the mean of the draw values (see associated file)
 293 from the IHME website:
 294 [http://ghdx.healthdata.org/record/global-burden-disease-study-2010-gbd-2010-ambient-air-pollut](http://ghdx.healthdata.org/record/global-burden-disease-study-2010-gbd-2010-ambient-air-pollution-risk-model-1990-2010)
 295 [ion-risk-model-1990-2010](http://ghdx.healthdata.org/record/global-burden-disease-study-2010-gbd-2010-ambient-air-pollution-risk-model-1990-2010).

Parameter	IHD	Stroke	COPD	LC	ALRI
α	1.4273	1.2641	15.224	114.74	2.2023
β	0.04764	0.00722	0.00095	0.000141	0.000284
ρ	0.376	1.314	0.684	0.741	1.183
PM _{2.5,Threshold}	7.462	7.387	7.374	7.380	7.283

296

297 **Table S13.** Table of GEMM parameters. The GEMM parameters are from Burnett et al. (2018),
 298 with the Chinese male cohort.

Cause of Death	Age Range (years)	θ	Standard Error θ	α	μ	π
NCD + LRI	>25	0.1430	0.01807	1.6	15.5	36.8
	27.5	0.1585	0.01477	1.6	15.5	36.8
	32.5	0.1577	0.01470	1.6	15.5	36.8
	37.5	0.1570	0.01463	1.6	15.5	36.8
	42.5	0.1558	0.01450	1.6	15.5	36.8
	47.5	0.1532	0.01425	1.6	15.5	36.8
	52.5	0.1499	0.01394	1.6	15.5	36.8
	57.5	0.1462	0.01361	1.6	15.5	36.8
	62.5	0.1421	0.01325	1.6	15.5	36.8
	67.5	0.1374	0.01284	1.6	15.5	36.8
	72.5	0.1319	0.01234	1.6	15.5	36.8
	77.5	0.1253	0.01174	1.6	15.5	36.8
	85	0.1141	0.01071	1.6	15.5	36.8
	>25	0.2969	0.01787	1.9	12	40.2
IHD	27.5	0.5070	0.02458	1.9	12	40.2
	32.5	0.4762	0.02309	1.9	12	40.2
	37.5	0.4455	0.02160	1.9	12	40.2
	42.5	0.4148	0.02011	1.9	12	40.2
	47.5	0.3841	0.01862	1.9	12	40.2
	52.5	0.3533	0.01713	1.9	12	40.2
	57.5	0.3226	0.01564	1.9	12	40.2
	62.5	0.2919	0.01415	1.9	12	40.2

300 **Table 13 cont.**

Cause of Death	Age Range (years)	θ	Standard Error θ	α	μ	π
IHD	67.5	0.2612	0.01266	1.9	12	40.2
	72.5	0.2304	0.01117	1.9	12	40.2
	77.5	0.1997	0.00968	1.9	12	40.2
	85	0.1536	0.00745	1.9	12	40.2
Stroke	>25	0.2720	0.07697	6.2	16.7	23.7
	27.5	0.4513	0.11919	6.2	16.7	23.7
	32.5	0.4240	0.11197	6.2	16.7	23.7
	37.5	0.3966	0.10475	6.2	16.7	23.7
	42.5	0.3693	0.09752	6.2	16.7	23.7
	47.5	0.3419	0.09030	6.2	16.7	23.7
	52.5	0.3146	0.08307	6.2	16.7	23.7
	57.5	0.2872	0.07585	6.2	16.7	23.7
	62.5	0.2598	0.06863	6.2	16.7	23.7
	67.5	0.2325	0.06190	6.2	16.7	23.7
	72.5	0.2051	0.05418	6.2	16.7	23.7
	77.5	0.1778	0.04695	6.2	16.7	23.7
	85	0.1368	0.03611	6.2	16.7	23.7
COPD	>25	0.2510	0.06762	6.5	2.5	3.2
Lung Cancer	>25	0.2942	0.06147	6.2	9.3	29.8
LRI	>25	0.4468	0.11735	6.4	5.7	8.4

301

302 **Table S14.** Calculated premature mortality from PM with all aerosol (base mortality) and
 303 removing ASOA, using the IER method.

Location^a	Base Mortality	Mortality reduced due to removing ASOA	Percent mortality reduced due to removing ASOA
North America	43,408	18,479	43%
Central America	11,808	3,395	29%
South America	31,214	10,100	32%
Africa	258,294	14,869	6%
Western Europe	305,754	31,880	10%
Eastern Europe	195,749	16,003	8%
South Asia	938,967	75,085	8%
Southeastern Asia	135,433	31,886	24%
East Asia	1,315,720	122,190	9%
Oceania	95	27	28%
Rest of the World	72,385	13,337	18%
Total	3,308,957	337,224	10%

304 ^aLocations defined by:
 305 http://themasites.pbl.nl/tridion/en/themasites/_disabled_image/background/regions/index-2.html

306 **Table S15.** Calculated premature mortality from PM with all aerosol (base mortality) and
 307 removing ASOA, using the GEMM method.

Location ^a	Base Mortality	Mortality reduced due to removing ASOA	Percent mortality reduced due to removing ASOA
North America	178,793	24,892	14%
Central America	58,516	7,298	12%
South America	145,395	22,372	15%
Africa	765,946	34,528	5%
Western Europe	768,991	50,427	7%
Eastern Europe	465,341	25,552	5%
South Asia	2,285,903	166,228	7%
Southeastern Asia	347,191	50,802	15%
East Asia	2,487,349	220,264	9%
Oceania	3,375	428	13%
Rest of the World	269,769	35,051	13%
Total	7,776,570	638,219	8%

308 ^aLocations defined by:

309 http://themasites.pbl.nl/tridion/en/themasites/_disabled_image/background/regions/index-2.html

310 **Table S16.** List of total final consumption, in millions of tonnes of oil equivalent, of oil products
 311 and oil, for each organization. Total final consumption includes imports, and does not include
 312 exports (IEA, 2019).

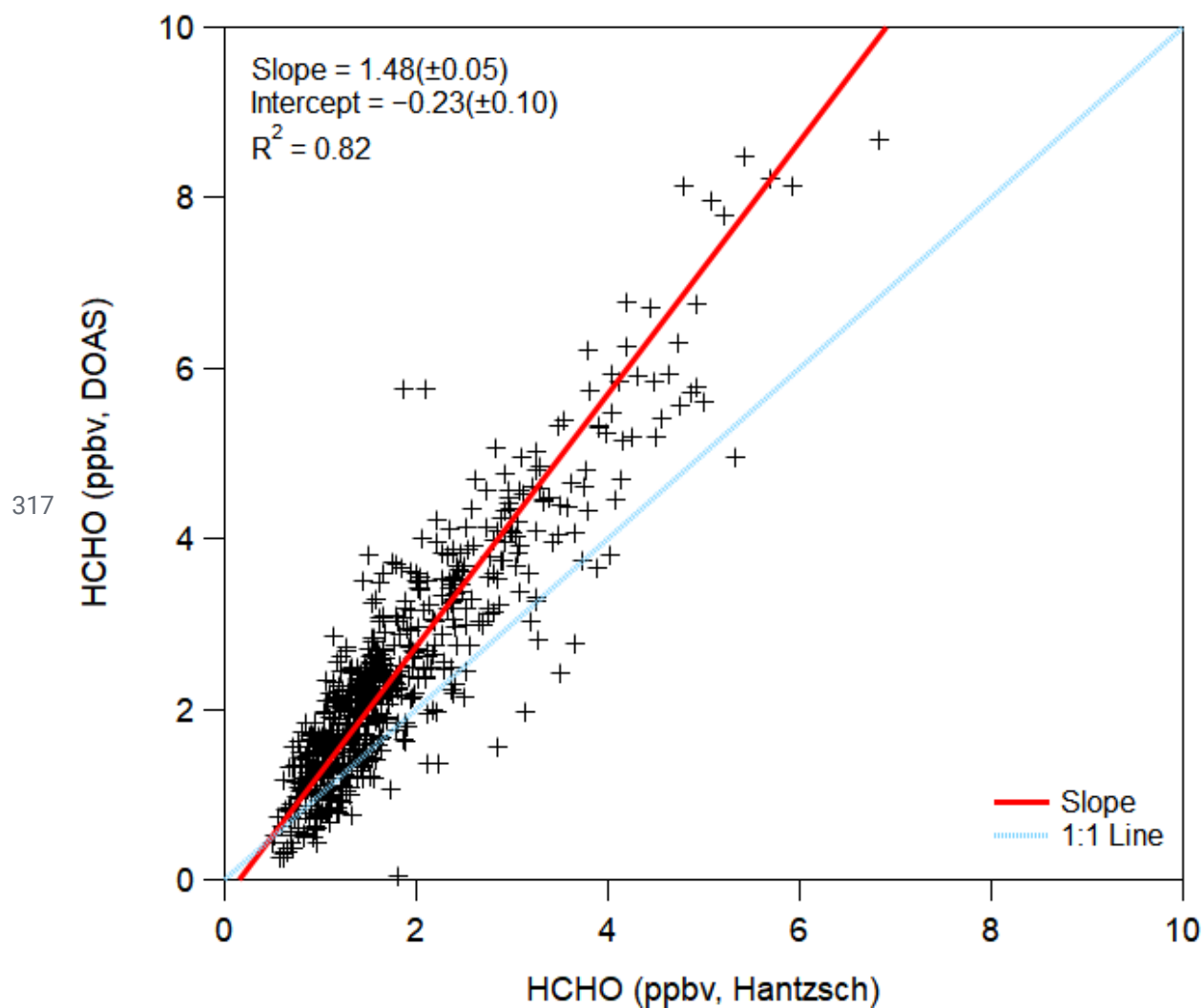
Organization	Industry	Transportation	Non-Energy
World	307	2533	645
OECD	89	1147	326
Africa	18.4	115.4	7.9
Non-OECD	28.3	135	20
Middle East	33.5	126.3	47.5
Non-OECD Europe and Eurasia	35	101	53

313

314 Supplemental figures for this study

315

316



318 **Figure S1.** Comparison of HCHO measured by the DOAS (Stutz and Platt, 1996, 1997) and
319 Hantzsch reaction (Cárdenas et al., 2000) methods during the CalNex 2010 study in Pasadena,
320 CA, ground site (Ryerson et al., 2013).

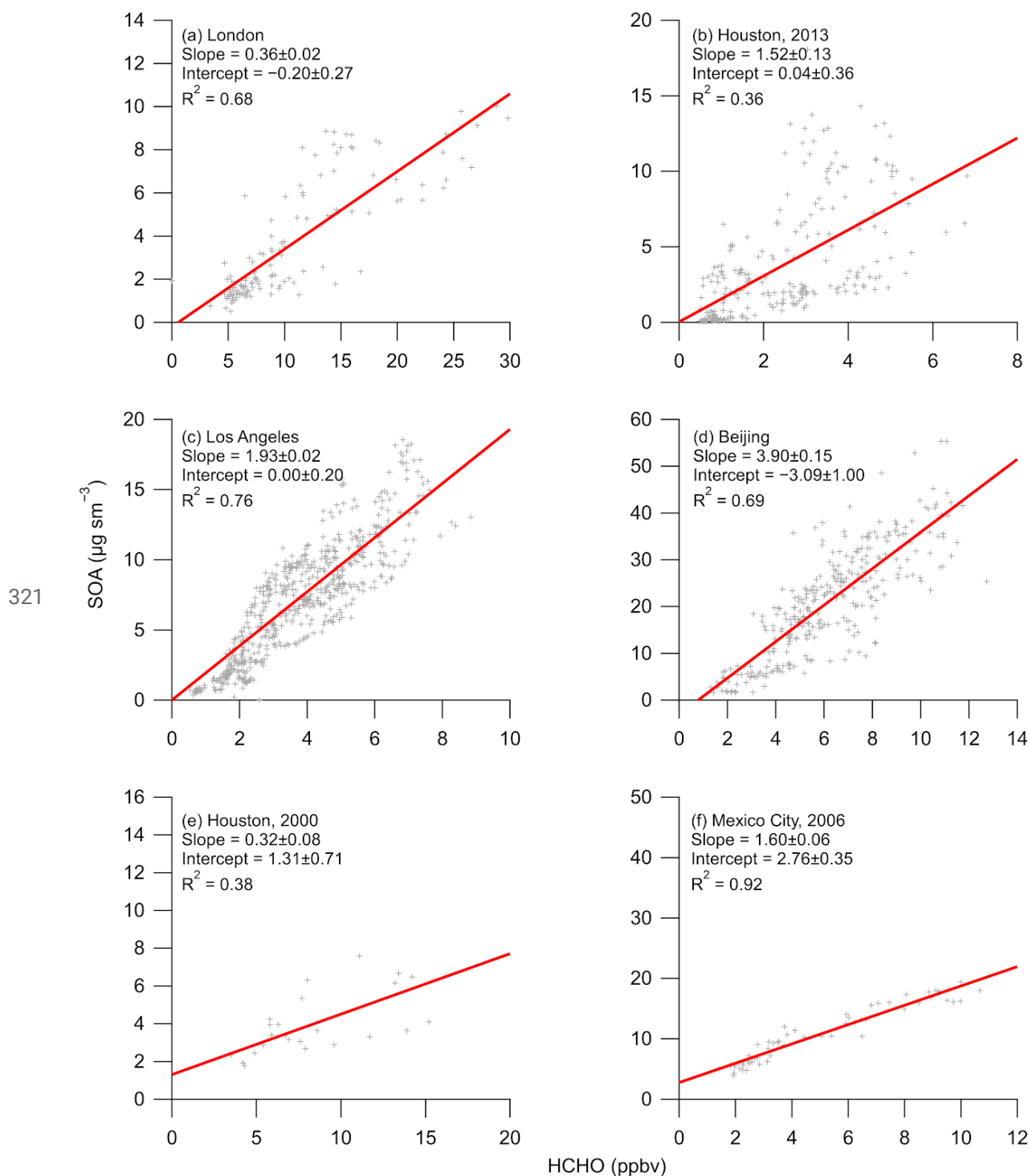


Figure S2. Regression plot of SOA versus HCHO from different campaigns around the world that have not been previously published. Note, for (c), HCHO is $1.24 \times$ Hantzsch HCHO, to account for the differences between the two HCHO measurements during CalNex. Note, for (a), SOA is $0.5 \times$ OA, estimated from Young et al. (2015), and for (f), SOA is $0.8 \times$ OA, estimated from DeCarlo et al. (2010).

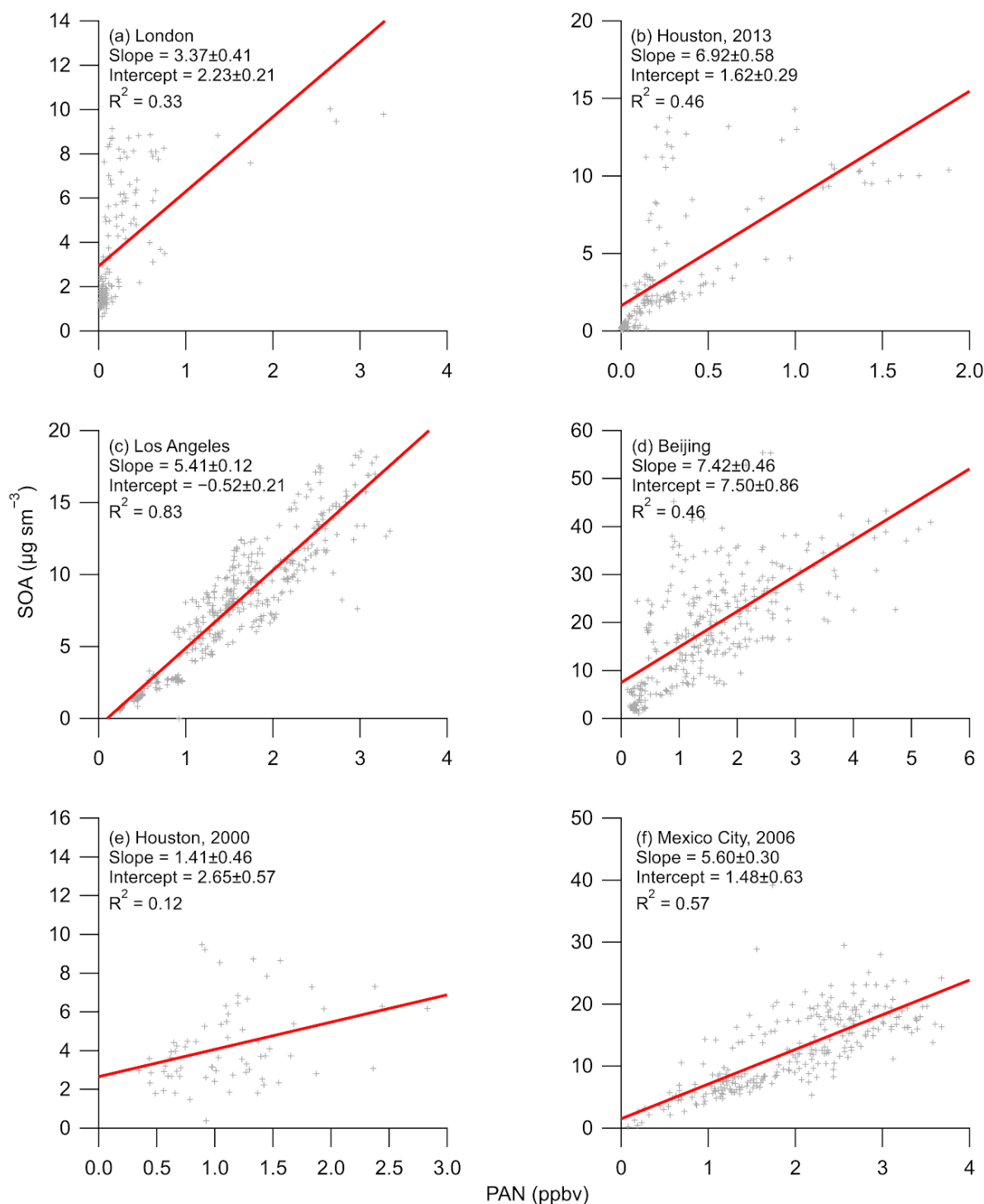
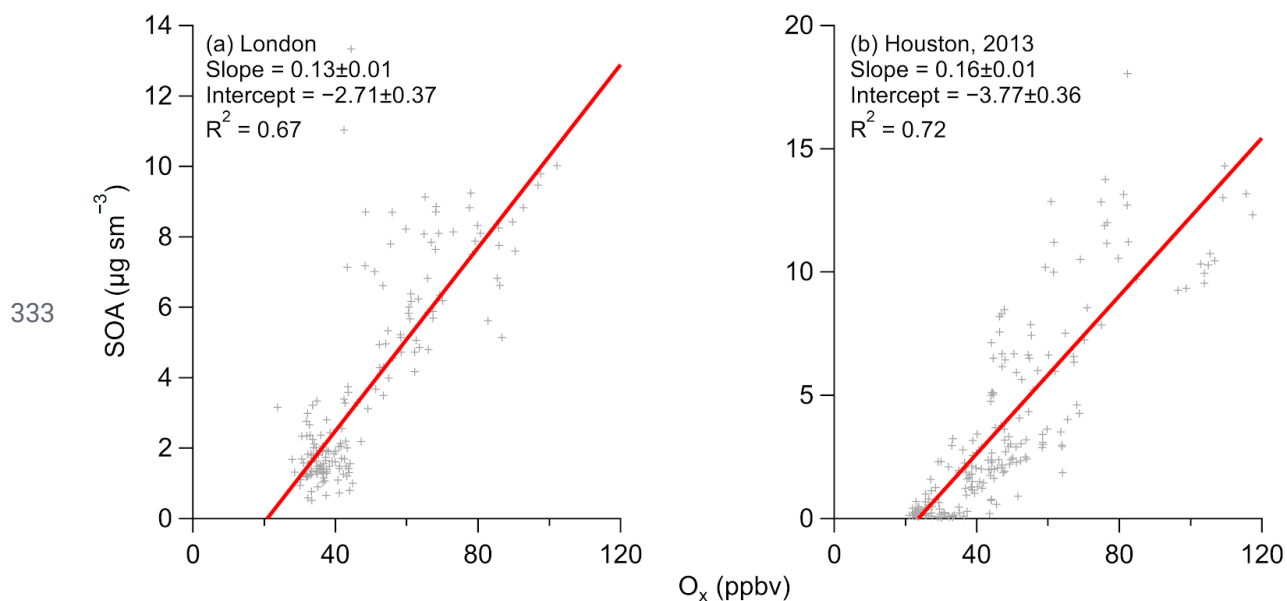


Figure S3. Regression plot of SOA versus PAN from different campaigns around the world that have not been previously published. Note, for (a), SOA is $0.5 \times \text{OA}$, estimated from Young et al. (2015), and for (f), SOA is $0.8 \times \text{OA}$, estimated from DeCarlo et al. (2010).

332



334 **Figure S4.** Regression plot of SOA versus Ox from different campaigns around the world that
 335 have not been previously published. Note, for (a), SOA is $0.5 \times \text{OA}$, estimated from Young et al.
 336 (2015).

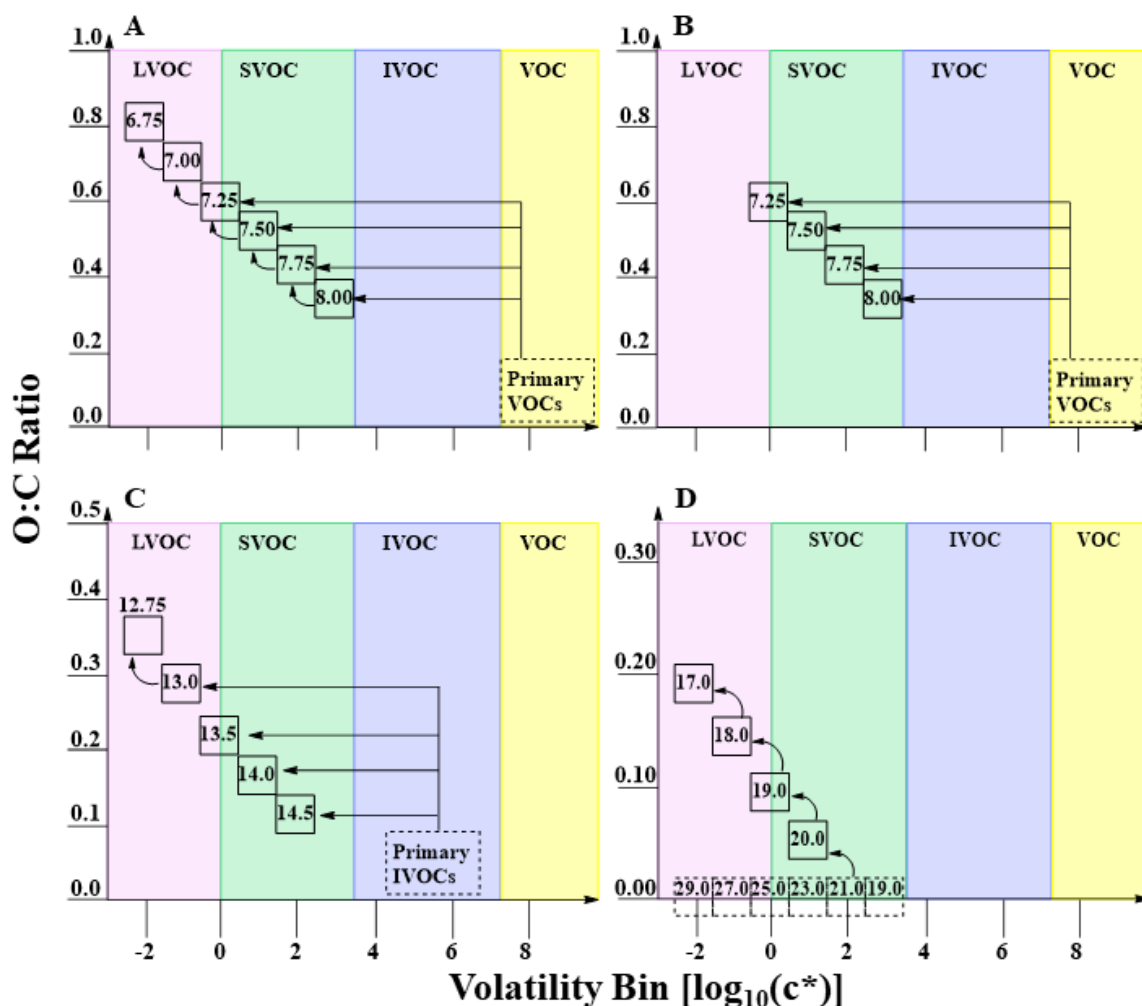
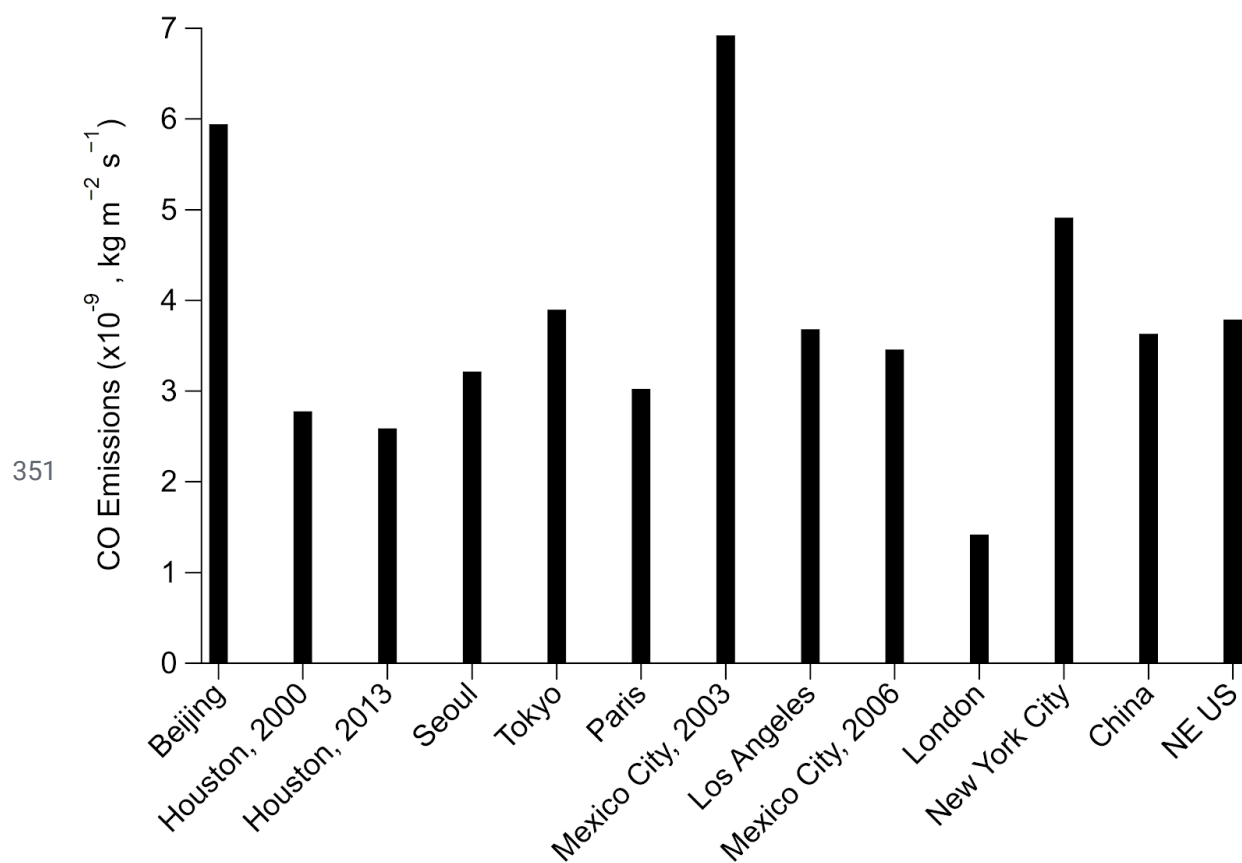


Figure S5. 2-D VBS space defined by oxygen to carbon (O:C) ratio and saturation concentration $[\log_{10}(c^*)]$ for different oxidation mechanisms and primary sources of OA precursors. Dashed boxes represent primary emissions, while the full boxes represent the secondary oxidation products. (A) and (B) represent different parameterizations for treating traditional anthropogenic and biogenic sources of SOA. Both parameterizations depict the oxidation of an 8-carbon precursor VOC. (A) represents the TSI, or aging, parameterization; (B) represents the MA, or wall-loss corrected, parameterization. (C) Represents the initial oxidation and aging pathway of P-IVOCs following the ZHAO parameterization. It should be noted that the carbon number corresponds to first generation aging and subsequent oxidation results in a 0.25 reduction in carbon number. (D) Represents the decadal aging of SVOCs by hydroxyl radicals. In (D), the full aging pathway of only the C21 species is depicted as an example, though all primary species are allowed to age until the $\log_{10}(c^*) = -2$ bin. All emitted P-SVOC species undergo the same decadal aging scheme which begins from the saturation concentration bin of the emitted species.



352 **Figure S6.** CO emissions for the cities investigated here from HTAP (Janssens-Maenhout et al.,
 353 2015).

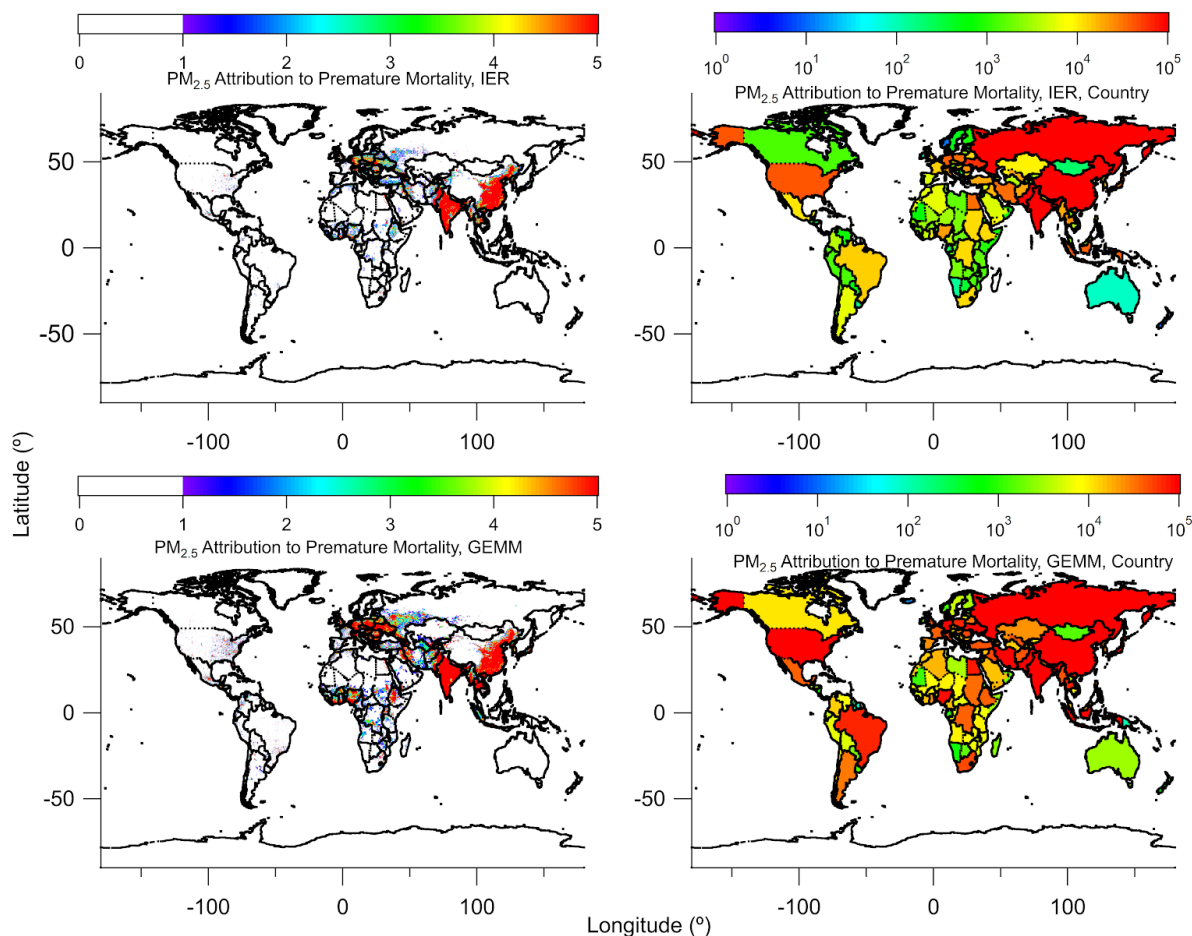


Figure S7. (top) Total deaths associated to PM_{2.5} (left) per 10×10 km² area and (right) summed up for each country, using the Integrated Exposure-Response (IER) method (Burnett et al., 2014). These values are derived from satellite. (bottom) Same as above, but using the Global Exposure Mortality Model (GEMM) (Burnett et al., 2018) for PM_{2.5} per 10×10 km² area (left) and summed up for each country (right). Premature mortality was determined with PM_{2.5} derived by the methods described in van Donkelaar (2016), which includes satellite and ground-based observations of aerosol.

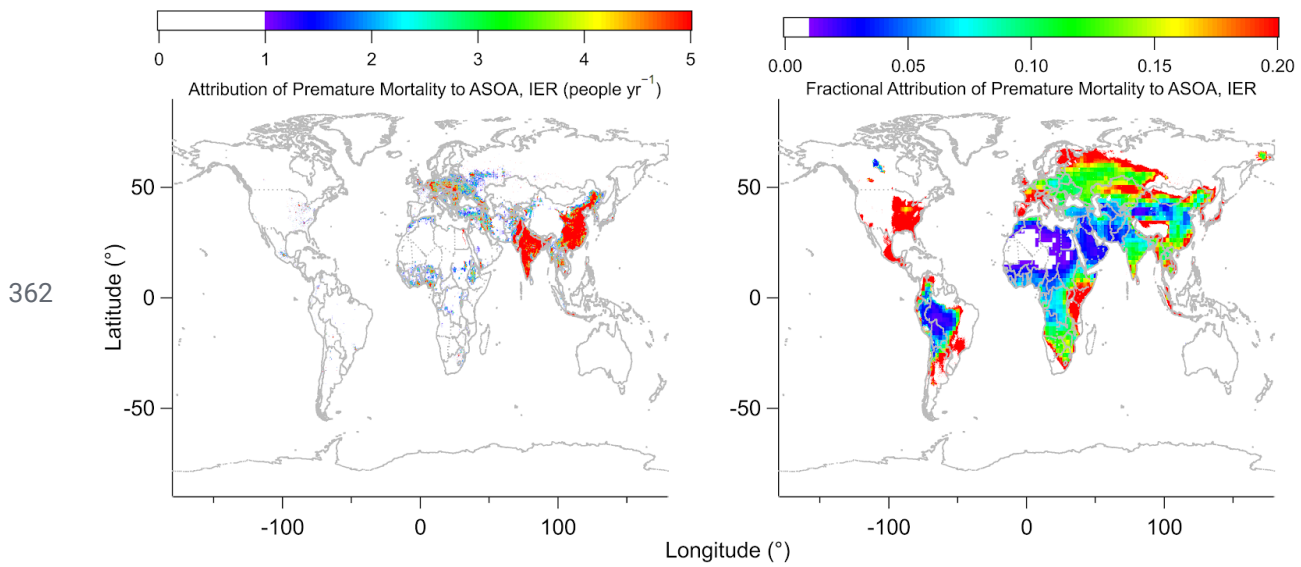


Figure S8. Same as Fig. 8, where top are the results per 10×10 km² area for the attribution of premature mortality to ASOA (people yr⁻¹, left) and fractional attribution of premature mortality to ASOA for one year (right) by the IER method. See Fig. 8 for per country comparison.

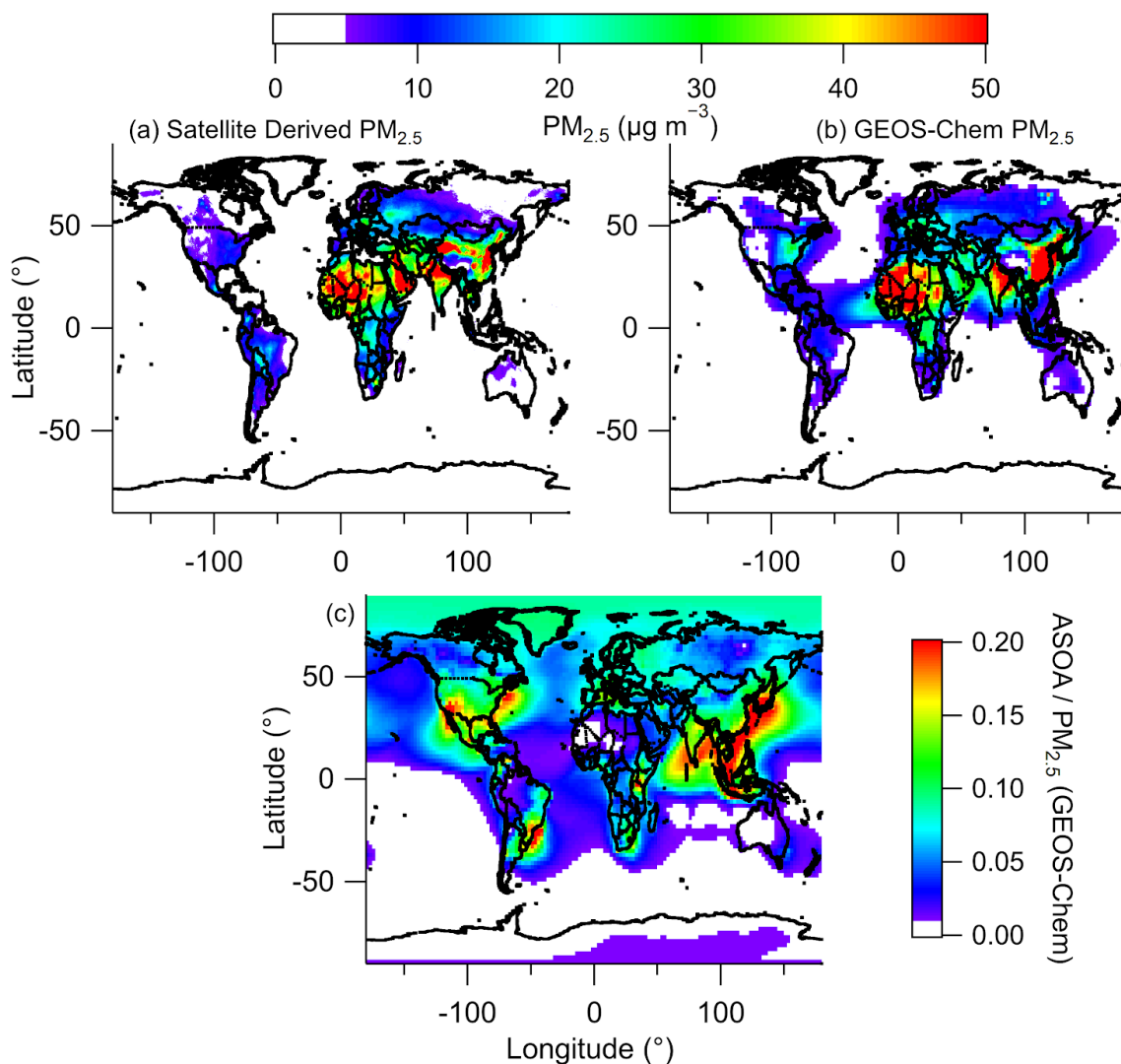


Figure S9. Comparison of satellite retrieved PM_{2.5} (upper left) versus modeled PM_{2.5} (upper right). (Bottom) Fractional contribution of ASOA to total modeled PM_{2.5}.

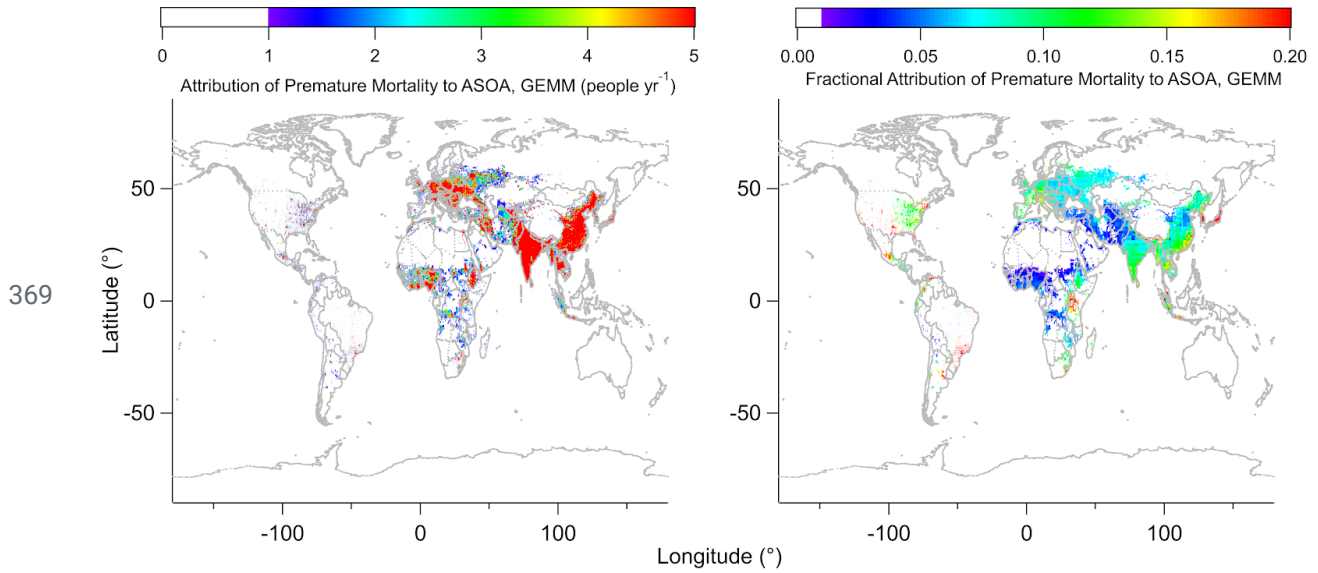
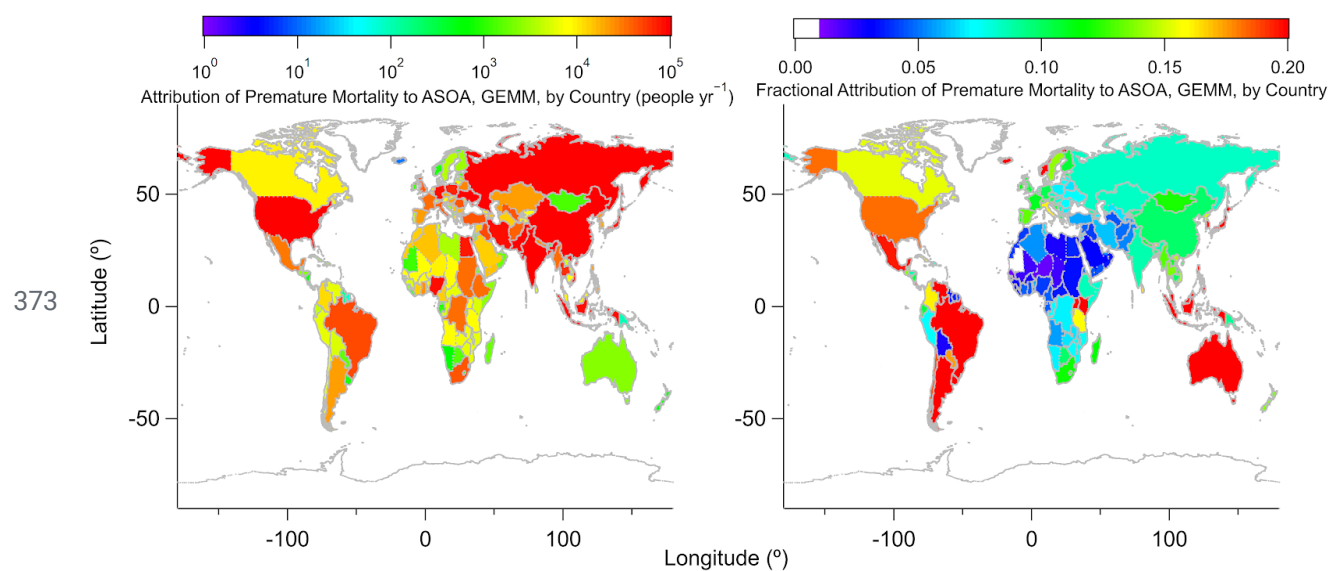
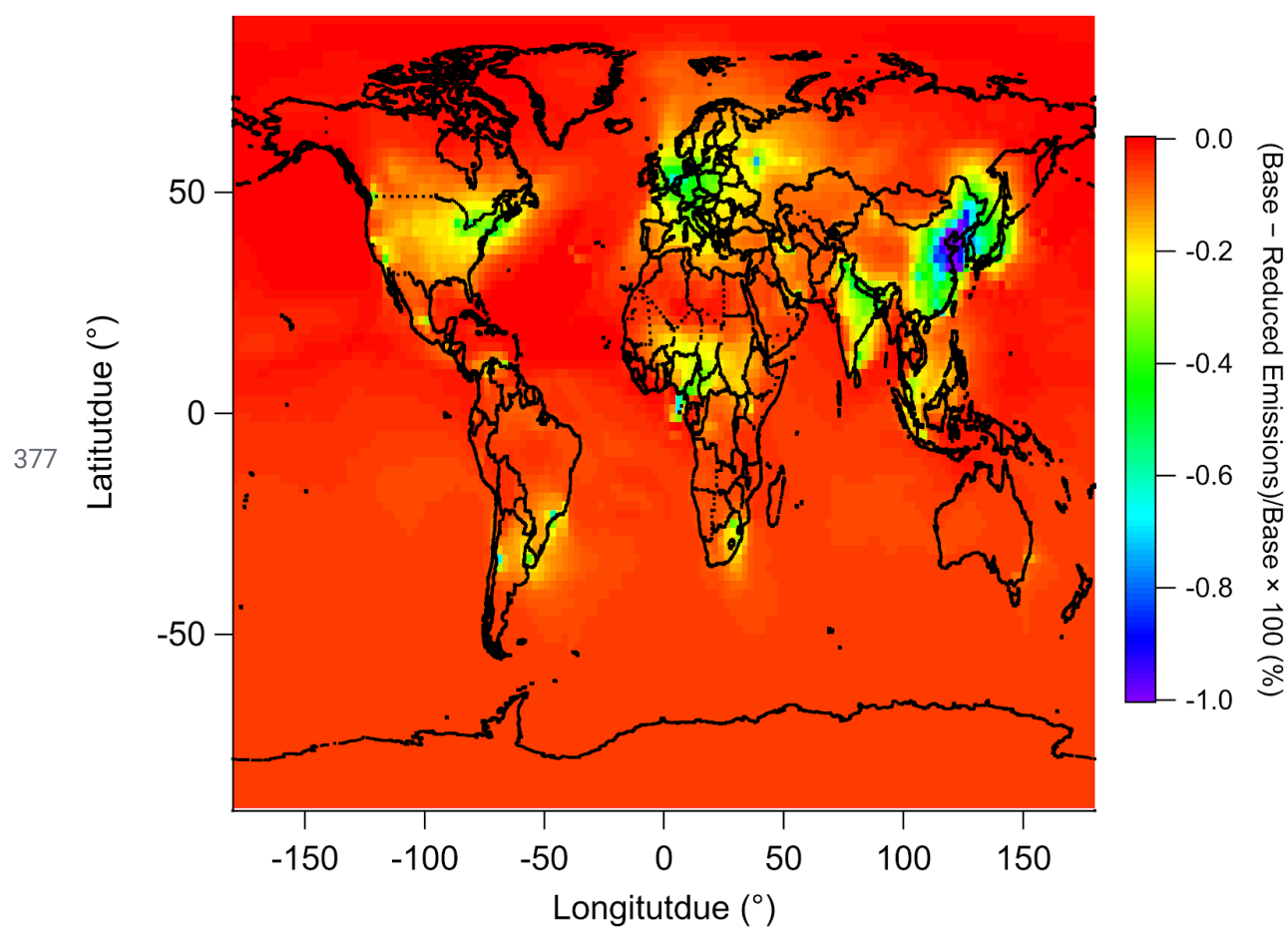


Figure S10. Same as Fig. S8, but using the GEMM from Burnett et al. (2018). (top). (Left) Attribution of premature mortality to ASOA per 10×10 km² area (people yr⁻¹) and (Right) fractional attribution of premature mortality to ASOA per 10×10² km for one year.



374 **Figure S11.** Same as Fig. S10 but summed up for each country for the (left) attribution of
 375 premature mortality to ASOA (people yr⁻¹) and (right) the fractional attribution of premature
 376 mortality to ASOA for one year.



378 **Figure S12.** Comparison for surface level ozone upon reducing SOA precursors by 20%.

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