

1           **Characterization of organic aerosol across the global**  
2                   **remote troposphere: A comparison of ATom**  
3                   **measurements and global chemistry models**

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32 **Abstract.**

33 The spatial distribution and properties of submicron organic aerosols (OA) are among the  
34 key sources of uncertainty in our understanding of aerosol effects on climate.  
35 Uncertainties are particularly large over remote regions of the free troposphere and  
36 Southern Ocean, where very little data has been available, and where OA predictions from  
37 AeroCom Phase II global models span two to three orders-of-magnitude, greatly  
38 exceeding the model spread over source regions. The (nearly) pole-to-pole vertical  
39 distribution of non-refractory aerosols was measured with an aerosol mass spectrometer  
40 onboard the NASA DC8 aircraft as part of the Atmospheric Tomography (ATom) mission  
41 during the northern hemisphere summer (August 2016) and winter (February 2017). This  
42 study presents the first extensive characterization of OA mass concentrations and their  
43 level of oxidation in the remote atmosphere. OA and sulfate are the major contributors by  
44 mass to submicron aerosols in the remote troposphere, together with sea salt in the marine  
45 boundary layer. Sulfate was dominant in the lower stratosphere. OA concentrations have  
46 a strong seasonal and zonal variability, with the highest levels measured in the lower  
47 troposphere in the summer and over the regions influenced by the biomass burning from  
48 Africa (up to  $10 \mu\text{g sm}^{-3}$ ). Lower concentrations ( $\sim 0.1\text{-}0.3 \mu\text{g sm}^{-3}$ ) are observed in the  
49 northern mid- and high- latitudes and very low concentrations ( $< 0.1 \mu\text{g sm}^{-3}$ ) in the  
50 southern mid- and high- latitudes. The ATom dataset is used to evaluate predictions of  
51 eight current global chemistry models that implement a variety of commonly used  
52 representations of OA sources and chemistry, as well as of the AeroCom-II ensemble.  
53 The current model ensemble captures the average vertical and spatial distribution of  
54 measured OA concentrations, and the spread of the individual models remains within a  
55 factor of 5. These results are significantly improved over the AeroCom-II model ensemble,  
56 which shows large overestimations over these regions. However, some of the improved  
57 agreement with observations occurs for the wrong reasons, as models have the tendency  
58 to greatly overestimate the primary OA fraction, and underestimate the secondary fraction.  
59 Measured OA in the remote free troposphere are highly oxygenated with organic aerosol  
60 to organic carbon (OA/OC) ratios of  $\sim 2.2\text{-}2.8$  and are 30-60% more oxygenated than in  
61 current models, which can lead to significant errors in OA concentrations. The  
62 model/measurement comparisons presented here support the concept of a more dynamic  
63 OA system as proposed by Hodzic et al. (2016), with enhanced removal of primary OA,  
64 and a stronger production of secondary OA in global models needed to provide a better  
65 agreement with observations.

## 66 1 Introduction

67 Organic aerosols (OA) are a complex mixture of directly emitted primary OA (POA) and  
68 chemically produced secondary OA (SOA) from anthropogenic and biogenic emission  
69 sources. They are associated with adverse health effects (Mauderly and Chow, 2008,  
70 Shiraiwa et al., 2017) and contribute radiative forcing in the climate system (Boucher et  
71 al., 2013). The currently limited understanding of processes involved in the formation,  
72 ageing, and removal of organic compounds results in large uncertainties in (i) the  
73 predicted global OA burden, (ii) relative contributions of emissions vs. chemistry to OA  
74 formation, (iii) spatial distribution, and (iv) impacts on radiation and clouds (Kanakidou et  
75 al., 2005, Hallquist et al., 2009, Heald et al., 2011, Spracklen et al., 2011, Tsigaridis et al.,  
76 2014, Hodzic et al., 2016, Shrivastava et al., 2017, Tsigaridis and Kanakidou, 2018, Zhu  
77 et al., 2019). The uncertainties are particularly large in the estimated global burden of SOA  
78 that range from 12 to 450 Tg y<sup>-1</sup> (see Fig. 9 of Hodzic et al., 2016), and in their direct and  
79 indirect radiative forcing that range from -0.08 to -0.33 W m<sup>-2</sup>, and -0.60 to -0.77 W m<sup>-2</sup>,  
80 respectively (Spracklen et al., 2011, Myhre et al., 2013, Scott et al., 2014, Hodzic et al.,  
81 2016, Tsigaridis and Kanakidou, 2018). Reducing these uncertainties is becoming more  
82 important as OA is on a path to becoming the dominant fraction of the submicron  
83 anthropogenic aerosol mass globally due to the ongoing efforts to reduce SO<sub>2</sub> emissions  
84 and associated sulfate aerosols.

85 Model performance has been especially poor in the remote regions of the atmosphere  
86 where OA measurements available for model evaluation have been sparse (especially  
87 aloft). Using data from 17 aircraft campaigns mostly located in the northern hemisphere  
88 Heald et al. (2011) showed that the skill of the global GEOS-Chem model in predicting the  
89 vertical distribution of OA was significantly decreased in remote regions compared to  
90 polluted near-source regions. The study pointed out the limitations of commonly used SOA  
91 formation mechanisms that are based on chamber data; these have the tendency to  
92 underpredict OA in source regions and overpredict OA in the remote troposphere. For a  
93 subset of 9 recent aircraft campaigns, Hodzic et al. (2016) showed that OA is likely a more  
94 dynamic system than represented in chemistry-climate models, with both stronger  
95 production and stronger removals. These authors suggested that additional removal  
96 mechanisms via e.g. photolytic or heterogeneous reactions of OA particles are needed to  
97 explain low OA concentrations observed in the upper troposphere where direct cloud  
98 scavenging is less efficient. The recent global multi-model comparison study (Tsigaridis

99 et al., 2014) within the AeroCom Phase II project illustrates well the amplitude of model  
100 uncertainties simulating OA mass concentrations and the contrast in model performance  
101 between near-source and remote regions. The results indicate that model dispersion (the  
102 spread between the models with the lowest and highest predicted OA concentrations)  
103 increases with altitude from roughly 1 order of magnitude near the surface to 2-3 orders  
104 of magnitude in the upper troposphere. Our own analyses of the AeroCom-II models  
105 shown in Figure 1a indicate that model dispersion (quantified as the ratio of the average  
106 concentration of the highest model to that of the lowest one, in each region) increases not  
107 only with altitude but also with distance from the northern mid-latitude source (and data-  
108 rich) regions. The model spread is a factor of 10-20 in the free troposphere between the  
109 equator and northern mid-latitudes, and increases to a factor of 200-800 over the Southern  
110 Ocean and near the tropopause. It is not surprising that model spread is lower closer to  
111 source regions where it is mostly driven by uncertainties in emissions and SOA production  
112 yields. Spread is expected to be larger in remote regions where models are also impacted  
113 by uncertainties in transport, chemical ageing and removal. The lowest model dispersion  
114 also coincides with the regions of the northern hemisphere (NH) or the African biomass  
115 burning outflow where models have been evaluated the most (Figure 1b), emphasizing  
116 the need for further model/observation comparison studies in remote regions (of the  
117 southern hemisphere (SH) in particular).

118 Here, we present a unique data set of airborne aerosol mass spectrometer measurements  
119 of OA mass concentrations collected onboard the NASA DC-8 as part of the Atmospheric  
120 Tomography (ATom) mission. The aircraft sampled the vertical structure of the  
121 atmosphere from near-surface (0.2 km) to the lower-stratosphere (LS) regions (12 km  
122 altitude) over both the Pacific and Atlantic basins (to limit the influence of source regions)  
123 with a quasi-global spatial coverage from 82°N to 67°S. This dataset is used to perform  
124 the first systematic global-scale multi-model evaluation of the chemistry-climate models  
125 focusing on OA in the remote troposphere over the remote oceans. We focus on the NH  
126 summer (August 2016, ATom-1) and NH winter (February 2017, ATom-2) deployments.  
127 Overall these ATom missions sampled the marine boundary layer (MBL) for 10% of the  
128 flight tracks, 12% of the time the remote lower stratosphere, and the rest the free  
129 troposphere. The model-observation comparisons are aimed at identifying discrepancies  
130 in terms of OA mass concentrations and vertical distribution, their fractional contribution  
131 to submicron aerosols, and their oxidation level in global models.

132 The modeling framework is described in Section 2. Section 3 describes the ATom dataset  
133 and the spatial and vertical distributions of OA over the Atlantic and Pacific regions.  
134 Section 4 presents the comparisons of ATom-1 and -2 data to multi-model predictions  
135 from both the AeroCom-II models, and the ensemble of eight current model simulations of  
136 the ATom campaign. Section 5 presents the conclusions of the study and discusses its  
137 implications.

## 138 **2 Modeling framework**

### 139 **2.1 ATom model simulations**

140 ATom measurements were compared with results of eight global models that simulated  
141 the time period of the ATom-1 and 2 campaigns (August 2016 and February 2017), using  
142 the emissions and reanalysis meteorology corresponding to this period (and a spin-up  
143 time of at least six to twelve months). These are referred hereafter as ATom models and  
144 include the NASA global Earth system model GEOS5, the aerosol-climate model  
145 ECHAM6-HAM, three versions of the NCAR Community Earth System Model (CESM),  
146 and three versions of the global chemistry GEOS-Chem model. Simulations were  
147 performed at various horizontal resolutions ranging from relatively high ~50km (GEOS5)  
148 and ~100km (CESM2 models) resolutions to somewhat coarser grids of ~200km (CESM1-  
149 CARMA, GEOS-Chem) and ~400km for GC10-TOMAS. The advantage of using the same  
150 host model (in the cases of variants of CESM2 and GEOS-Chem) is that the dynamics  
151 and emissions remain comparable. Models differ greatly in their treatment of emissions,  
152 gas-phase chemistry, aerosol chemistry and physical processes, and aerosol coupling  
153 with radiation and clouds, among others. Table 1 describes the configuration of various  
154 models (e.g. meteorology, emissions), and their treatment of OA. In this section we only  
155 summarize the main features and parameters directly impacting the OA simulations. Some  
156 models do not include SOA chemistry and instead assume that SOA is directly emitted  
157 proportional to the emissions of its precursors (ECHAM6-HAM, CESM2-SMP, GEOS5,  
158 GC10-TOMAS), while others have more complex treatments of organic compounds, their  
159 chemistry, and partitioning into particles (GC12-REF, GC12-DYN, GC10-TOMAS,  
160 CESM1-CARMA, CESM2-DYN). It should be noted that models that directly emit SOA  
161 assume that SOA is a non-volatile species that remains irreversibly in the particle phase.  
162 In all models POA is treated as a non-volatile directly emitted species. In most models  
163 (see below) the primary emitted organic aerosol is artificially aged to transition between  
164 hydrophobic to hydrophilic POA. There are some commonalities between simulations for

165 the treatment of biogenic emissions, which are based in all models on the Model of  
166 Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2012) to  
167 generate meteorology-dependent emissions of volatile organic compounds. None of the  
168 models includes the marine production of OA which is estimated to be ~3 orders of  
169 magnitude smaller than the continental production of OA from both isoprene and  
170 monoterpene precursors (Kim et al., 2017), but could be important in the MBL. This  
171 contribution could however be larger for sea-spray biological material from phytoplankton  
172 with predicted contributions of 0.01 to 0.1  $\mu\text{g m}^{-3}$  to surface submicron aerosol over remote  
173 oceanic regions (Vergara-Temprado et al., 2017, Middlebrook et al., 1998). Below we only  
174 provide a brief description of most important processes that influence OA for each model.

175 GEOS5 was run in a configuration similar to Bian et al. (2019) using the anthropogenic  
176 emissions from HTAP v2 (Janssens-Maenhout et al., 2015) and biomass burning  
177 emissions from the Quick Fire Emission Dataset (QFED v2.54). Aerosols are simulated  
178 within the GOCART bulk aerosol module and include externally mixed particles of black  
179 carbon (BC), organic carbon (OC), sulfate, ammonium, nitrate, dust and sea salt (Colarco  
180 et al., 2010, Bian et al., 2017). The formation of SOA is based on a prescribed 10%  
181 formation yield from the monoterpene emissions. The primary emitted OC and SOA are  
182 separated into hydrophobic (50%) and hydrophilic (50%) species, with a 2.5 days e-folding  
183 time conversion from hydrophobic to hydrophilic organic particles. All SOAs from biogenic,  
184 anthropogenic, and biomass burning sources are treated as hydrophilic particles. Both  
185 types of organic particles are dry deposited. The hydrophilic OA is removed by large-scale  
186 and convective warm clouds, while hydrophobic OA is removed by ice clouds. The  
187 hydrophilic particles undergo hygroscopic growth according to the equilibrium  
188 parameterization of Gerber (1985).

189 The ECHAM6.3-HAM2.3 standard version (Tegen et al., 2019) was run using updated  
190 anthropogenic emissions (Schacht et al., 2019) combining the ECLIPSE (Klimont et al.,  
191 2017) emissions, with the Russian anthropogenic BC emissions from Huang et al. (2015).  
192 For biomass burning the Global Fire Assimilation System (GFAS, Kaiser et al., 2012)  
193 biomass burning emissions are used, however, without the scaling factor of 3.4 suggested  
194 by Kaiser et al. (2012). Aerosol composition and processes are simulated using the  
195 Hamburg Aerosol Model (HAM2, Zhang et al., 2012), that considers an aerosol internal  
196 mixture of sulfate, BC, OC, sea salt, and mineral dust. The aerosol population and their  
197 microphysical interactions are simulated using seven log-normal modes, including the

198 nucleation mode, soluble and insoluble Aitken, accumulation and coarse modes. In the  
199 model configuration used in this publication the formation of SOA is based on a prescribed  
200 15% mass yield from monoterpene emissions only (Dentener et al., 2006). Aerosol  
201 particles are removed by dry and wet deposition. The wet deposition includes the below  
202 cloud scavenging by rain and in-cloud cloud scavenging for large-scale and convective  
203 systems (Croft et al., 2010).

204 The two simulations with the GEOS-Chem 12.0.1 global chemistry model (Bey et al.,  
205 2001) use emissions based on CMIP6 global inventory (CEDS historical emissions up to  
206 2014 and future emissions based on climate scenarios, Hoesly et al., 2018; Feng et al.,  
207 2019) with regional improvements for anthropogenic sources, and on GFED v.4 for  
208 biomass burning emissions (Giglio et al., 2013). Both simulations use the bulk aerosol  
209 representation and differ only in the treatment of SOA formation and removal. The first  
210 configuration (called hereafter GC12-REF) includes the default  
211 (<http://wiki.seas.harvard.edu/geos-chem/index.php>) representation of SOA formation  
212 based on Marais et al. (2016) for isoprene-derived SOA, and on the volatility basis set  
213 (VBS) of Pye et al. (2010) for all other precursors. Note that this GEOS-Chem REF  
214 simulation is similar to the version 12 default “complex option” which includes non-volatile  
215 POA and semi-volatile SOA (semi-volatile POA is an optional switch within this version  
216 used in Pai et al. 2020). The second configuration (referred to as GC12-DYN) includes a  
217 more dynamic representation of the SOA lifecycle based on Hodzic et al. (2016), with the  
218 exception of the treatment of isoprene SOA that is formed in the aqueous aerosols as in  
219 Marais et al. (2016). As in Hodzic et al. (2016) the GC12-DYN model version includes  
220 updated VBS SOA parameterization, updated dry and wet removal of organic vapors, and  
221 photolytic removal of SOA (except for isoprene-SOA that is formed in aqueous aerosols,  
222 where we follow Marais et al. 2016). SOA formation is based on wall-corrected chamber  
223 yields (Zhang et al., 2014) for the traditional precursors (isoprene, monoterpenes,  
224 sesquiterpenes, benzene, toluene, xylene) and on yields derived from an explicit chemical  
225 mechanism for higher molecular weight n-alkanes and n-alkenes species (Hodzic et al.,  
226 2016). The removal of gas-phase oxidized volatile organics uses updated Henry’s law  
227 solubility coefficients from Hodzic et al. (2014), and photolytic removal of SOA (Hodzic et  
228 al., 2015). In addition to OA, the model includes BC and dust, and simulates the chemistry  
229 and gas-particle partitioning of inorganic compounds such as sulfate, ammonium, nitrate  
230 and sea salt using the ISORROPIA II thermodynamic model (Fountoukis and Nenes,  
231 2007). In both GEOS-Chem configurations, BC and primary OC are simulated with a

232 hydrophobic and hydrophilic fraction for each. At the time of emission, 80% of BC and  
233 50% of primary OC are considered as hydrophobic. Hydrophobic aerosols are converted  
234 to hydrophilic aerosols with an e-folding lifetime of 1.15 days. An OA/OC ratio of 2.1 is  
235 assumed to convert POC to POA, and SOA is simulated as OA mass (i.e. no OA/OC ratio  
236 assumption is needed for SOA, except for comparison with OC measurements). Soluble  
237 gases and aerosols are removed by both dry and wet deposition. Wet deposition includes  
238 scavenging in convective updrafts, and in-cloud and below-cloud scavenging from large-  
239 scale precipitation (Liu et al., 2001). Hydrophobic aerosols (BC and POA) are scavenged  
240 in convective updrafts following Wang et al. (2014).

241 GC10-TOMAS is based on the GEOS-Chem version 10.01 coupled with Two Moment  
242 Aerosol Sectional microphysics scheme (TOMAS) and ran in a similar configuration to that  
243 described in Kodros et al. (2016). The model computes the evolution of sulfate, sea salt,  
244 primary and secondary OA, BC, and dust aerosols described by 15 internally mixed size  
245 bins (of which six were analyzed for these comparisons, cf. Table 1). Anthropogenic  
246 emissions are based on the EDGAR v4 global inventory with regional improvements, while  
247 the biomass burning emissions are from GFED v3. SOA are irreversibly made from the  
248 emitted parent precursor, considering a 10% mass yield from monoterpene emissions,  
249 and an emission flux of 0.2 Tg of SOA per Tg of CO for the anthropogenic CO emissions.  
250 The removal of gases and aerosols are treated similar to the GEOS-Chem 12.0.1 model  
251 (GC12-REF, see above).

252 Simulations based on the CESM2.0 Earth system model use the standard version of the  
253 Whole Atmosphere Community Climate Model (WACCM6, Gettelman et al., 2019,  
254 Emmons et al., 2019). Details on the specific of the model configurations are described in  
255 detail in Tilmes et al. (2019) i.e. CESM2-SMP and CESM2-DYN correspond to the  
256 specified dynamics WACCM6-SOAG and WACCM6-VBSext simulations described in that  
257 work, respectively. Emissions are based on the CMIP6 global inventory for the year 2014  
258 for anthropogenic sources, and on the QFED version 2.4 for the wildfires inventory.  
259 Aerosols are represented with the modal aerosol scheme (MAM4, Liu et al., 2012) that  
260 includes BC, primary and secondary OA, sulfate, dust and sea salt. Four modes are  
261 considered including Aitken, accumulation and coarse size modes, and an additional  
262 primary carbon mode. Only the accumulation mode was used in this work. The CESM2-  
263 SMP and CESM2-DYN simulations differ in their treatment of OA. CESM2-SMP forms OA  
264 directly using fixed mass yields from primary emitted precursors (isoprene, monoterpenes,



265 aromatics) without explicitly simulating their oxidation and partitioning. These mass yields  
266 are increased by a factor of 1.5 to match the anthropogenic aerosol indirect forcing (Liu et  
267 al., 2012). The second configuration (referred to as CESM2-DYN) includes the formation  
268 and removal parameterizations of organics of Hodzic et al. (2016), as implemented into  
269 CESM2 by Tilmes et al. (2019) for all species based on low-NO<sub>x</sub> VBS yields only. This is  
270 a similar SOA scheme as used in GC12-DYN (with differences in the treatment of  
271 isoprene-SOA based on Marais et al. 2016 in GC12-DYN, and the use of both low- and  
272 high-NO<sub>x</sub> VBS yields in GC12-DYN). Organic gases and aerosols undergo dry and wet  
273 deposition as described in Liu et al. (2012). It should be noted that CESM2-SMP does not  
274 include deposition of intermediate organic vapors. Aerosol wet scavenging considers in-  
275 cloud scavenging (the removal of cloud-borne particles that were activated at the cloud  
276 base) and below-cloud scavenging for both convective and grid-scale clouds.

277 CESM1-CARMA simulations use the configuration described in Yu et al. (2019) which is  
278 based on CESM1 and the sectional Community Aerosol and Radiation Model for  
279 Atmospheres (CARMA v3.0). Anthropogenic emissions are those from the Greenhouse  
280 gas-Air pollution Interactions and Synergies (GAINS) model, and biomass burning  
281 emissions are from the Global Fire Emission Database (GFED v3, van der Werf et al.,  
282 2010). In CARMA, 20 size bins are used for both pure sulfate particles (bins from 0.2 nm  
283 to 1.3 μm in radius, only used up to 500 nm) and mixed aerosols composed of BC, primary  
284 and secondary OC, dust, sea salt, and sea-spray sulfate (bins from 0.05–8.7 μm in radius,  
285 again, only analyzed up to 500 nm). SOA formation is based on the VBS approach from  
286 Pye et al. (2010). The removal of OA occurs only by dry and wet deposition. Compared to  
287 the CESM2-SMP and CESM2-DYN simulations, the convective removal of aerosols uses  
288 the modified scheme described in Yu et al. (2019) which accounts for aerosol secondary  
289 activation from the entrained air above the cloud base, and the scavenging of activated  
290 aerosols in convective updrafts. The default CESM can transport aerosols from the cloud  
291 base to the top of the cloud in strong convective updrafts in one time step without  
292 scavenging them, while the new scheme allows for a more efficient removal off all aerosols  
293 inside convective clouds. A sensitivity simulation is performed for ATom-1 to quantify the  
294 effect of this improved removal on OA concentrations (Section 4.5).

## 295 **2.2 AeroCom-II model climatology**

296 The ATom measurements are also compared to the global model OA predictions  
297 generated within the Phase II Aerosol Comparisons between Observations and Models

298 (AeroCom-II) project (Schulz et al., 2009). We consider the monthly average results of 28  
299 global models, which is a subset of those presented in Tsigaridis et al. (2014), based on  
300 the availability of model results. It should be noted that the meteorological forcing used in  
301 these models is mostly based on the year 2006, while the anthropogenic and biomass  
302 burning emissions are mostly representative of the year 2000. For comparison purposes,  
303 the monthly mean model outputs for the months of August (ATom-1) and February (ATom-  
304 2) are interpolated along the flight path (latitude, longitude, and altitude), and averaged  
305 the same way as the measurements (see section 3.2).

### 306 **3 Description of ATom measurements**

#### 307 **3.1 Submicron aerosol data**

308 The measurements of non-refractory submicron aerosols were performed onboard the  
309 NASA DC8 aircraft as part of the ATom field study (Wofsy et al., 2018) using the University  
310 of Colorado Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (AMS  
311 in the following, Canagaratna et al., 2007, DeCarlo et al., 2006).

312 We use measurements from both the NH summer (August 2016, ATom-1) and winter  
313 (February 2017, ATom-2) deployments. Figure 2a shows the flight path and the vertical  
314 extent of the ATom-1 dataset colored by OA mass concentrations (see Figure S1 for  
315 ATom-2). The aircraft performed systematic vertical sampling with ~140 vertical profiles  
316 per campaign throughout the troposphere from the near surface ~0.2 km to the upper  
317 troposphere/lower stratosphere region at ~13 km altitude. Details on the operation of the  
318 CU AMS on board the DC-8 are reported in Schroder et al. (2018), Nault et al. (2018), and  
319 Jimenez et al. (2019b). AMS data was acquired at 1 Hz time resolution and independently  
320 processed and reported at both 1 s and 60 s time resolutions (Jimenez et al., 2019a). The  
321 later product, with more robust peak fitting at low concentrations was exclusively used as  
322 the primary dataset in this work. Detection limits at different time resolutions/geographical  
323 bins relevant to this study are discussed in Section 3.3. The overall  $2\sigma$  accuracies of the  
324 AMS measurement (38% for OA, 34% for sulfate and other inorganics) are discussed in  
325 Bahreini et al. (2008) and Jimenez et al. (2019b).

326 For ATom, the AMS reported the standard non-refractory aerosol species OA, sulfate,  
327 nitrate, ammonium, and chloride, with the response for all the nominally inorganic species  
328 characterized by in-field calibrations. In addition, it also reported methanesulfonic acid  
329 (MSA, Hodshire et al., 2019a describes the AMS MSA methods and calibrations for ATom)

330 and sea salt for for  $D_{\text{geo}} < 450$  nm (based on the method of Ovadnevaite et al., 2012). Both  
331 of these species were important to achieve closure with the volume calculated from the  
332 on-board sizing instruments in the marine boundary layer (Jimenez et al., 2019b). Another  
333 important refractory submicron species not captured by the AMS measurements is BC.  
334 This was measured on ATom with the NOAA SP2 instrument (Katich et al., 2018). It should  
335 be noted that aircraft measurements of aerosol mass concentrations are given in  $\mu\text{g sm}^{-3}$   
336 (i.e., under standard conditions of 1 atm and 273.15 K).

337 For ATom the AMS measured particles with geometric diameters (based on the campaign-  
338 wide average density of  $1640 \text{ kg m}^{-3}$ , Jimenez et al., 2019b) of between  $D_{\text{geo}} \sim 60$  and 295  
339 nm with  $\sim 100\%$  efficiency (and between 35 and 460 nm with 50% efficiency). Here we  
340 denote the AMS aerosol data as “submicron” mass (based on the more usual definition  
341 using aerodynamic diameter, which is larger than the geometric diameter; DeCarlo et al.,  
342 2004), with the assumption that non-refractory aerosol are small contributors to mass  
343 above the AMS size range. As shown in Brock et al., 2019, the accumulation mode for the  
344 ATom sampling environment only extended up to 500 nm, and hence, as expected for a  
345 background tropospheric environment, this approximation is appropriate. Very good  
346 agreement was observed with the integrated volume calculated from the number size  
347 distributions for ATom (Brock et al., 2019). A low bias compared to a typical submicron  
348 definition can occur in thick biomass burning plumes and in the lower stratosphere at times  
349 (Jimenez et al., 2019b). As detailed in Table 1, the accumulation mode for the bulk models  
350 discussed in this study overlaps with the size range of the AMS, and for the sectional  
351 models (CESM1-CARMA, GEOS-Chem-TOMAS, ECHAM6-HAM) only the bins that  
352 match the AMS size range were used. As expected based on the previous discussion,  
353 however, a comparison of the total OA calculated by these sectional models with the  
354 modeled OA inside the AMS size-range showed small differences (Slopes for ATom-1  
355 linear regressions: CESM1-CARMA: 0.91, GC10-TOMAS: 0.94, ECHAM6-HAM 1.00)  
356 mostly influenced by the high concentration points in the biomass plumes off Africa that  
357 have a large effect on the regression since they are about 10 times larger than the bulk of  
358 the dataset).

359 Refractory and non-refractory aerosol composition was also measured using the NOAA  
360 Particle Analysis by Laser Mass Spectrometry (PALMS) instrument. PALMS classifies  
361 individual aerosol particles into compositional classes including biomass burning (Hudson  
362 et al., 2004), sea salt (Murphy et al., 2019), mineral dust (Froyd et al., 2019), and

363 others. Mass concentrations for these particles types are derived by combining PALMS  
364 composition data with aerosol size distribution measurements (Froyd et al., 2019). Good  
365 agreement overall was found for OA, sulfate and seasalt between the two particle mass  
366 spectrometers during ATom once the AMS and PALMS instrument transmissions were  
367 accounted for (Jimenez et al., 2019b). For all PALMS data used in this work (biomass  
368 burning fraction and dust) the AMS transmission function was applied to ensure that both  
369 instruments were characterizing approximately the same particle range.

370 For a particular airmass, the mass fraction of biomass burning (BB) aerosol reported by  
371 the PALMS instrument  $f(\text{BB})_{\text{PALMS}}$  (Thompson and Murphy, 2000; Froyd et al., 2019) was  
372 then used to evaluate the degree of BB influence. This parameter correlates quite well  
373 with other gas-phase BB tracers (Figure S20), and is more useful as a particle tracer since  
374 its lifetime follows that of the particles. Importantly, it is not impacted by the long lifetimes  
375 of the gas-phase tracers (e.g. 9 months for  $\text{CH}_3\text{CN}$ ) and unrelated removal processes  
376 (e.g. ocean uptake for  $\text{CH}_3\text{CN}$  and  $\text{HCN}$ ) that result in highly variable backgrounds. Hence  
377  $f(\text{BB})_{\text{PALMS}}$  has a much higher contrast ratio and linearity for particle BB impacts, compared  
378 to the available gas-phase tracers in the ATom dataset. An airmass was classified as non-  
379 BB influenced when  $f(\text{BB})_{\text{PALMS}}$  was lower than 0.30 (Hudson et al, 2004) as shown in  
380 Figure 2b. For both ATom-1 and 2, about 74% of measurements were classified as not  
381 influenced by biomass burning.  $f(\text{BB})_{\text{PALMS}}$  was also used to assess the impact of POA on  
382 the total OA burden (next section); note that no thresholding was applied in that case.

### 383 **3.2 Estimation of the POA fraction for the ATom dataset**

384 For model evaluation purposes, it is important to know whether the source of OA is primary  
385 or secondary. For ground studies close to sources (e.g. Jimenez et al., 2009) Positive  
386 Matrix Factorization of AMS mass spectra (PMF, Ulbrich et al., 2009) can be used to  
387 estimate the contribution of primary sources (mostly from transportation, heating, cooking,  
388 and biomass burning) to total OA. This approach is not suitable for ATom. To accurately  
389 resolve a minor factor such as POA in an AMS dataset, there needs to be a combination  
390 of: (a) Sufficient OA mass concentration, so that the signal-to-noise of the spectra is  
391 sufficient; (b) Enough fractional mass for the factor to be resolved (>5% in urban areas  
392 per Ulbrich et al. (2009), probably a larger fraction at low concentrations such as in ATom);  
393 (c) Sufficient spatio-temporal variability (“contrast”) in the relative contributions of different  
394 factors, since that is part of what PMF uses to extract the factors; (d) Sufficient difference  
395 in the spectra of the different factors (for the same reason as (c)), and (e) relatively

396 invariant spectra for each factor across the dataset (as this is a key assumption of the  
397 PMF algorithm). As an example of a near ideal case, in Hodshire et al (2019) we extracted  
398 MSA by PMF from the ATom-1 data, and were able to match that factor with our  
399 independently calibrated MSA species. A very distinct and nearly invariant mass spectrum  
400 was measured repeatedly near sources (MBL) (and was mostly absent elsewhere, thus  
401 providing strong spatio-temporal contrast) and accounted for about 6% of the fractional  
402 mass and 15% of the variance in time. Thus all the conditions were met. For POA, on the  
403 other hand, the air sampled in ATom and coming from e.g. Asia has POA and SOA very  
404 well mixed, with little change on their relative mass fractions vs. time (as the aircraft flies  
405 through that airmass). POA is very low, as documented later in this paper. Atmospheric  
406 aging makes the spectra from all OA sources more and more similar as measured by AMS  
407 spectra (Jimenez et al., 2009). Thus most of the conditions above are not satisfied for  
408 extracting POA by PMF analysis of this dataset.

409 Instead, in this work we have estimated POA based on the fact that it is co-emitted with  
410 BC as part of the combustion processes releasing both species in source regions, and  
411 that BC is not impacted by chemical aging processes over the lifetime of the airmass. Note  
412 that BC can physically age but it is not lost in any significant amount to the gas-phase due  
413 to chemical processes in the atmosphere. We assume non-differential removal (and  
414 transport) of the BC fraction relative to the rest of the POA (the two are generally internally  
415 mixed, Lee et al., 2015). Table S1 summarizes recent POA/BC and POC/EC emission  
416 ratio determinations for urban background sites, which best represent real mixes of  
417 pollution sources, and for individual sources of POA (from mobile sources – commonly  
418 referred as HOA – and cooking aerosol – COA). Based on Table S1 data, we assume  
419 POA to be co-emitted with BC for anthropogenic fossil fuel / urban region POA (herein  
420 called  $FF_{\text{ratio}}$  for simplicity, even though much of it is non-fossil, Zotter et al., 2014; Hayes  
421 et al., 2015) at a ratio of  $1.5 \pm 0.82$  (average  $\pm 1\sigma$  of all urban ambient air studies that report  
422 POA and BC for best intercomparability to the ATom dataset; including all urban studies  
423 results in a very similar number,  $1.48 \pm 0.65$ , median: 1.41). Measurements where mobile  
424 source are the main contributor in general exhibit lower ratios (POA/OA ratio 0.5-1.5),  
425 while COA determination typically ranges from 2 to 3. Hence, the ratio used here is a good  
426 estimate for a diverse mix of urban sources as appropriate for ATom. The studies used to  
427 derive the emission ratio used ambient data in urban air, where all sources mix together  
428 and impact the POA/BC ratio, and thus the ratios include the impact of POA sources that  
429 may not emit BC. It should be noted that urban model ratios do not include emissions

430 associated with fugitive dust from road, tire and construction, as those are typically found  
431 in larger particles than those studied here (Zhao et al., 2017). For biomass burning  
432 sources, we use a value of POA/BC = 11.8 ( $BB_{ratio}$ ), based on the average of the recent  
433 review by Andreae (2019), which included over 200 previous determinations for a variety  
434 of fuels and burning conditions (since Andreae (2019) used and OA/OC ratio of 1.6 in his  
435 work, we have used that value to calculate POA/BC; we note that this is different from the  
436 1.8 OA/OC ratio used for other studies listed in Table S1). We note the measured total  
437 OA/BC of ~3.5 (conservatively assuming that all OA is POA) observed on both ATom  
438 missions for the large African-sourced BB plumes over the Equatorial Atlantic. We note  
439 that using the larger  $BB_{ratio}$  from Andreae (2019) leads to a POA fraction >> 100% in the  
440 ATom African plumes. We also perform sensitivity studies with values of both  $FF_{ratio}$  and  
441  $BB_{ratio}$  within the literature range.

442 The PALMS determined mass fraction of biomass impacted aerosol ( $f(BB)_{PALMS}$ ) can then  
443 be used to determine a total POA contribution from both types of sources:

$$444 \quad POA = [BC] * (FF_{ratio} + (BB_{ratio} - FF_{ratio}) * f(BB)_{PALMS}) \quad (Eq. 1)$$

445 Further detail is provided in Table S2, which summarizes the POA/BC ratios used in the  
446 emission inventories implemented in current models. Overall, there is reasonable  
447 agreement with the measurements in Table S1, with  $FF_{ratio}$  ranging from ~0.5 for diesel  
448 fuels, to >2 for energy production and ~5 for residential emissions (which include some  
449 BB). On the other hand, for biomass burning, the emission inventories ratios range from  
450 ~5 for crop, to ~15 for forest, and up to ~50 for peatland. While generally consistent with  
451 the values discussed by Andreae (2019), they are on the lower end of the ranges  
452 discussed in that work. The averages and ranges of the measurement and model ratios  
453 are similar, and thus no significant model bias on the ratios is apparent.

454 PALMS detection efficiency increases with size across the accumulation mode, and  
455 therefore the  $f(BB)$  number fraction is weighted to the larger size end of the accumulation  
456 mode. In very clean regions of the upper troposphere (typically <0.15  $\mu\text{g sm}^{-3}$  submicron  
457 mass) particles below the PALMS size range can contribute significantly to aerosol mass  
458 (Williamson et al., 2019; Jimenez et al., 2019b). If BB particles are not evenly distributed  
459 across the entire accumulation mode (due to preferential removal in convective updrafts  
460 of primary aerosol, cf. Yu et al., 2019 and Section 4.5; and preferential condensation of  
461 SOA on smaller particles), then the  $f(BB)$  reported by PALMS will be an overestimation.  
462 For the final analysis these periods were left in the dataset, and therefore for the LS the

463 reported POA is likely overestimated for these regions, although their impact on the mass-  
464 weighted campaign average is negligible.

465 The contribution of POA from sea spray is difficult to constrain. As an order-of-magnitude  
466 estimate, marine POA is roughly calculated based on preliminary calibrations of OA on  
467 mineral dust particles from the PALMS instrument (personal communication K. Froyd).  
468 Using this calibration, the average OA by mass on sea salt was <10% for the large majority  
469 of MBL sampling (>85%). Since sea salt contributed 4% (11%) of mass in the AMS size  
470 range for ATom-1(2) (Figure 2), we estimate that marine POA is on the order of ~1% of  
471 aerosol mass in the AMS size range, and possibly much lower. Thus we think that it is  
472 reasonable to neglect the contribution of marine POA to this dataset. Future studies will  
473 refine this estimate.

### 474 **3.3 Data processing for comparisons**

475 For the comparisons between the measurements and the various global models, data  
476 were averaged both vertically and zonally to minimize the impact of smaller plumes or  
477 vertical gradients in aerosol concentrations that might not be captured by coarse resolution  
478 models. For the same reason, all data near airports was removed from the datasets prior  
479 to analysis (up to about 3 km on the climb in/out). In order to restrict this analysis to the  
480 remote troposphere, the last leg of the ATom-1 mission (over the continental US) was  
481 taken out of the dataset as well. Data was binned into 5 large latitude regions as shown in  
482 Figure 2a including southern polar (55-80°S, "S.Polar"), southern mid-latitudes (25-55°S,  
483 "S.Mid"), equatorial (25°S-25°N, "Equatorial"), northern mid-latitudes (25-55°N, "N.Mid"),  
484 northern polar (55-80°N, "N.Polar") and analyzed separately for the Pacific and Atlantic  
485 basins. For data in each of these latitude regions, altitude profiles were calculated with a  
486 constant 600 m altitude resolution. According to both variability in the cleanest air and  
487 statistical analysis of the organic background subtraction (Drewnick et al. 2009), the 1 $\sigma$   
488 precision at low concentrations for one-minute data ranged between 20 and 50 ng sm<sup>-3</sup>,  
489 or a 3 $\sigma$  detection limit between 60 and 150 ng sm<sup>-3</sup> for the one-minute data (confirmed by  
490 frequent filter blanks). Per standard statistics, the precision of a measurement decreases  
491 (i.e., gets better) with the square root of the number of points (or time interval) sampled.  
492 I.e. the precision of an average can be approximated by the standard error of the mean  
493 ( $\sigma/\sqrt{n}$ ), where  $n$  is the number of measurements averaged), and it is better than the  
494 precision of the individual data points ( $\sigma$ ). This also applies to the detection limit, since it  
495 is just 3 times the precision. Note that a detection limit is not meaningful unless the

496 averaging time is specified. For example let's assume that the detection limit is  $20 \text{ ng m}^{-3}$   
497 (1-second), and the data points over 60 consecutive seconds are all  $10 \text{ ng m}^{-3}$ . All 1-  
498 second measurements are below the 1-second DL. However the average ( $10 \text{ ng m}^{-3}$ ) is  
499 now above the DL for 1-minute averages, which is  $20/\sqrt{60} = 2.6 \text{ ng m}^{-3}$ . On average,  
500 each individual point in the profiles represents the average of about 25 min of ATom flight  
501 data. At that time resolution, the OA  $1\sigma$  precision was about  $10 \text{ ng sm}^{-3}$ . Hence with very  
502 few exceptions (10 points for both missions combined), the OA concentrations in the  
503 averaged profiles reported are well above the instrumental detection limit in those regions.  
504 For model-measurement comparisons along flight tracks, model outputs and  
505 measurements were considered at 1-minute time resolution, which corresponds to ~0-700  
506 m vertical resolution and ~0.05-0.15 degrees horizontal resolution. Note that a large  
507 fraction of the 1-minute OA values in the remote free troposphere were below the local  $3\sigma$   
508 detection limit. The data of periods of zero concentration (sampling ambient air through a  
509 particle filter) do average to zero. Some negative measurements are present, and this is  
510 normal for measurements of very low concentrations in the presence of instrumental  
511 noise. Averaging of longer periods, as done for the figures in this paper, reduces the  
512 detection limit. We therefore caution future data users that the reported data should be  
513 averaged as needed, as replacing below-detection limit (or negative) values by other  
514 values introduces biases on averages. For fractional ratio analysis, measurements were  
515 averaged to 5-minute time resolution to reduce the noise in the ratios due to noise in the  
516 denominator. The results are not very sensitive to the 5-minute averaging (compared to  
517 1-minute) as shown in Figure S12 for OA to sulfate ratios. The same figure also illustrates  
518 that excluding ratios affected by negative concentrations (the non-bracketed case, overall  
519 these are about 15% of the dataset) does not really affect the fractional distribution, with  
520 the variance between the two cases diminishing as the averaging interval increases. To  
521 further confirm that there is no inherent bias in the fractional products regardless of the  
522 treatment of low concentration values, an additional sensitivity analysis was performed  
523 where data was filtered by an independent measurement proxy for aerosol mass, the  
524 aerosol volume measured in ATom (Brock et al., 2019). Using a range of value that  
525 encompasses the regime where the AMS calculated volume to aerosol measured volume  
526 exhibited increased noise (Jimenez et al., 2019b), no systematic bias was found (Figure  
527 S13), with variations of about 10% in fractional volume for different filtering conditions.

528 Some of the performed analysis required separating the dataset into vertical subsets. In  
529 this manuscript, we define the marine boundary layer (MBL) as the region below 1.5 times



530 the calculated boundary layer height in the NCEP global model reanalysis. The free  
531 troposphere (FT) includes all data points between the top of MBL and the NCEP  
532 tropopause height, and the LS region includes all points above the NCEP tropopause  
533 height. The tropopause height varied during ATom between 8 and 16.5 km; given the DC-  
534 8 ceiling (42 kft, 12.8 km) the stratosphere was only sampled at latitudes higher than 30  
535 degrees in both hemispheres. The MBL height varied between up to 1.5 km in the mid-  
536 latitudes, ~1 km in the tropics, and sometimes <150 m (lowest DC-8 altitude) for some of  
537 the sampling in the polar troposphere.

### 538 **3.4 Submicron aerosol composition**

539 Figure 2b shows that during both NH summer and winter ATom deployments, OA is one  
540 of the three dominant components of the measured submicron aerosol in the remote  
541 troposphere, together with sulfate and sea salt. During ATom-1, average submicron  
542 aerosol concentrations were close to  $0.8 \mu\text{g sm}^{-3}$  in the marine boundary layer and  
543 biomass burning outflow regions, and ~2 times lower in the free troposphere and lower  
544 stratosphere regions. ATom-2 had overall lower average concentrations below  $0.4 \mu\text{g sm}^{-3}$   
545 (vs.  $0.5 \mu\text{g sm}^{-3}$  for ATom-1). As expected, sulfate (sulfuric acid in the lower stratosphere)  
546 is the dominant constituent in the MBL (~50%) and LS (50-70%), while the OA contribution  
547 is generally below 10% and 40%, respectively in those regions. A large fraction of sea salt  
548 aerosol is found in the MBL especially during the NH winter deployment (~30%, see  
549 Murphy et al., 2019).

550 OA is found to be a major constituent (~50%) of submicron aerosol in the clean (non-BB  
551 influenced) free troposphere. The contribution of OA is 1.4 times larger than that of sulfate  
552 during the NH summer, and 1.2 times lower than that of sulfate during the NH winter,  
553 which is likely due to a large contribution of the NH sources to SOA production in the NH  
554 summer. Biomass-burning events increase the OA contribution relative to that of sulfate,  
555 and lead to a higher contribution of OA to total during the ATom-1 mission (stronger BB  
556 influence).

### 557 **3.5 Spatial and vertical distribution of OA**

558 Figure 2a (and Fig. S1) shows the spatial and vertical distribution of OA mass  
559 concentrations measured during ATom-1 (and ATom-2) campaigns. Most data were taken  
560 over remote oceanic regions (and a few remote continental regions, primarily over the  
561 Arctic). The measured OA varies between extremely clean conditions ( $< 0.1 \mu\text{g sm}^{-3}$ )

562 encountered mostly in the Pacific and Southern Ocean regions and moderately polluted  
563 conditions ( $> 2 \mu\text{g sm}^{-3}$ ) in the biomass burning outflow regions. During ATom-1 (August  
564 2016), a strong BB influence is observed in the lower troposphere (below 6 km) over the  
565 Atlantic basin off the African coast and over California with OA concentrations exceeding  
566  $10 \mu\text{g sm}^{-3}$ . OA associated with biomass burning is also present in the upper troposphere  
567 over equatorial regions and over Alaska, associated with the deep convective transport of  
568 biomass burning aerosols. The biomass burning contribution to carbonaceous aerosols in  
569 those regions during ATom-1 was also apparent in the black carbon measurements  
570 (Katich et al., 2019). ATom-2 was generally less polluted than ATom-1, likely due to a  
571 more limited global influence of biomass burning emissions during that period, and also to  
572 a less active photochemistry during winter months in the NH.

573 The measured OA is characterized by a strong latitudinal gradient. Figure 2c shows the  
574 average vertical profiles of measured OA over the selected latitudinal bands during August  
575 2016. The cleanest airmasses are observed over the remote oceanic regions of the  
576 Southern Hemisphere (SH, 25-80°S) with OA mass concentrations below  $0.06 \mu\text{g sm}^{-3}$ .  
577 These extremely low OA concentrations can be explained by the very low influence from  
578 continental emission sources, and presumably low marine POA and SOA precursor  
579 emissions. This is consistent with low concentrations of gas-phase pollutants (e.g. CO,  
580 ethane, propane). An enhancement can be noticed above 10 km in the lower stratosphere.  
581 In some cases, this could be related to the long-range transport of biomass burning  
582 aerosols from the tropics. By comparison, the Arctic region is more polluted with an order  
583 of magnitude higher OA levels compared to its analog of the SH (i.e. OA loadings ranging  
584 from  $0.1$  to  $0.5 \mu\text{g sm}^{-3}$ ). These concentrations are comparable to FT levels measured in  
585 the extratropical regions (25-55°N) of the NH. The equatorial marine regions (25°S-25°N)  
586 display the highest OA concentrations with a strong gradient between lower and upper  
587 troposphere. In the lower troposphere OA, concentrations are close to  $1 \mu\text{g sm}^{-3}$ , and  
588 decrease down to  $0.1 \mu\text{g sm}^{-3}$  at altitudes above 4km. The highest OA levels are  
589 associated with the African outflow over the southeastern Atlantic Ocean, which results  
590 from the transport of the biomass burning smoke from the sub-Saharan regions and  
591 increasing urban and industrial air pollution in southern West Africa (Flamant et al., 2018).  
592 Figure 2d shows that the Atlantic basin is often more polluted than the Pacific basin, not  
593 only because of the African biomass burning influence but also due to the contribution of  
594 anthropogenic pollution in the lower troposphere of the NH. It should be noted that Asian

595 pollution was likely an important contributor to the North Pacific Basin, especially between  
596 2 and 6 km, in both ATom deployments (see figures 2a and S1). Several-fold higher OA  
597 concentrations are found near the surface (below 1km) over the southern Pacific  
598 compared to that same location in the southern Atlantic, which could be indicative of the  
599 stronger emission of marine OA in the Pacific basin.

600 In addition to spatial gradients, a strong summer-to-winter contrast is observed in OA  
601 concentrations. Figure 2e shows the ratio between OA vertical profiles measured in the  
602 NH summer ATom-1 vs. in the NH winter ATom-2. The NH is more polluted during the NH  
603 summer due to the photochemical production of SOA, as well as biomass burning  
604 emissions, leading to the tripling of OA concentrations in the extratropical regions (25-  
605 80°N) on average regardless of altitude. The doubling of OA loading in the lower  
606 troposphere at the equator (25°S-25°N) in the NH summer (August, ATom-1) is strongly  
607 influenced by the biomass burning activity in the sub-Saharan African region as already  
608 mentioned above. Likewise, OA concentrations are found to be generally higher in the SH  
609 during the SH summer. These zonal trends are broadly similar to the ones described in  
610 Katich et al (2018) for BC.

## 611 **4 Model-measurement comparisons**

### 612 **4.1 Evaluation of predicted OA concentrations**

613 Prior to evaluating model performance in simulating OA, we have assessed the ATom  
614 models' ability to simulate sulfate aerosols. According to the model evaluation shown in  
615 Table S3, the predicted sulfate concentrations are generally within 40% of the measured  
616 values, which is comparable to the AMS measurement uncertainties. The only exception  
617 is found for the ECHAM6-HAM model, which overestimates sulfate aerosols by a factor of  
618 two. These results imply that most ATom models capture relatively well the overall sulfate  
619 burden. However, large root mean square error (RMSE > 0.4  $\mu\text{g sm}^{-3}$  for ATom-1 and >  
620 0.2  $\mu\text{g sm}^{-3}$  for ATom-2) is indicative of their limited skill in reproducing the observed  
621 variability in sulfate concentrations.

622 For OA, model evaluation metrics for the entire ATom-1 and ATom-2 campaigns are given  
623 in Table 2 for the eight ATom models and their ensemble, as well as the AeroCom-II  
624 ensemble. The results show that the normalized mean bias is substantially lower for the  
625 ATom model ensemble compared to AeroCom-II decreasing from 74% to 4% for ATom-1  
626 and from 137% to 23% to ATom-2, which is within the measurement uncertainty range.

627 The mean temporal correlations are substantially improved from 0.31 (0.38) for AeroCom-  
628 II to 0.66 (0.48) for ATom model ensemble during ATom-1 (ATom-2). However, results  
629 vary strongly among ATom models. Models using prescribed emissions of non-volatile  
630 SOA have the tendency to overestimate the OA concentrations during both NH summer  
631 and winter deployments (with ~35-60% overestimation for CESM2-SMP, ~70-100% for  
632 ECHAM6-HAM, and up to 150% for GC10-TOMAS during ATom-2), with the exception of  
633 the GEOS5 model that on the contrary underestimates OA concentrations by 5-25%.  
634 During the NH summer (ATom-1), models using the VBS parameterization from Pye et al.  
635 (2010) tend to underpredict the OA concentrations by 43% for GC12-REF and 33% for  
636 CESM1-CARMA for ATom-1, most likely due to the excessive evaporation of the formed  
637 SOA in remote regions and low yields for anthropogenic SOA (Schroder et al., 2018; Shah  
638 et al., 2019). Models using the VBS parameterization from Hodzic et al. (2016) (CESM2-  
639 DYN and GC12-DYN) where OA is less volatile and also OA yields are corrected for wall  
640 losses show an improved agreement with observations especially for CESM2-DYN (with  
641 NMB of ~5%), and to a lesser extent for GC12-DYN (NMB of ~33%). During the NH winter  
642 (ATom-2) characterized by a lower production of SOA, both VBS approaches lead to an  
643 overestimation of the predicted OA. This is likely caused by excessively high levels of  
644 primary emitted OA as discussed in section 4.4.

645 Figure 3 compares the average median ratios between modeled and observed OA  
646 concentrations for the ATom and AeroCom-II model ensembles for different regions (BB,  
647 MBL, FT, LS). The results show that the median ratio for the ATom model ensemble is  
648 close to unity in all regions. This is at least a factor of two improvement compared to  
649 AeroCom-II models, which were almost always biased high for the remote regions  
650 sampled in ATom. The model spread has also been reduced by a factor of 2-3 in all  
651 regions. This reduction in the ensemble spread may partially be explained by a smaller  
652 size of the ATom model ensemble (see Fig. S2), which also includes models with a more  
653 up-to-date OA representation. In order to explore this point further, results for a subset of  
654 AeroCom-II models (using earlier versions of models in the ATom ensemble) show only a  
655 slight reduction (~10%) in the model spread, with however some regional differences i.e.  
656 an improved agreement with observations in the MBL, but an increase in the model bias  
657 and spread in the LS (Figure S2). Thus, model improvement for the more recent models  
658 appears to be the main reason for the reduced spread.

#### 659 **4.2 Evaluation of predicted OA vertical distribution**

660 Figure 4 compares the mean vertical profiles of OA measured during ATom-1 and -2 with  
661 the predictions of the model ensemble average based on the eight ATom models (Table  
662 1) and 28 AeroCom-II models for the different latitudinal regions of the Pacific and Atlantic  
663 basins. Note that the use of a wide logarithmic scale (to be able to span all the  
664 observations) may make the observed differences appear small, although they often reach  
665 factors of 2-10 and larger (Figure S5 shows the results on a linear scale). For AeroCom-  
666 II, large latitudinal differences exist in the results with a better performance closer to source  
667 regions and large disagreement in the lower stratosphere and remote regions, as already  
668 suggested by the mission medians shown in Figure 3. The best AeroCom-II model  
669 performance is found over the equator in both basins, where the model ensemble captures  
670 within a factor of 2 the observed OA concentrations throughout the troposphere in the  
671 Pacific basin, and matches remarkably well the observations in the lower troposphere of  
672 the Atlantic basin that is heavily influenced by biomass burning emissions. Reasonable  
673 agreement is found for the OA vertical distribution over the NH Atlantic and Pacific oceans,  
674 especially in the lower troposphere (< 4 km). The largest model discrepancies (1-2 orders  
675 of magnitude) are found in the remote regions of the Southern Ocean and SH mid-latitudes  
676 during both seasons and basins. The model overestimation is also large over the NH mid-  
677 latitude Pacific basin in the upper troposphere. A spread of 2-3 orders of magnitude is  
678 observed around the ensemble average indicating a very large variability in individual  
679 model predictions. This evaluation of AeroCom-II models in remote regions is an extension  
680 of that performed at the surface for urban and remote stations by Tsigaridis et al. (2014)  
681 (as in that previous study, the data and model simulations compared are not synchronous  
682 in time). The tendency of the model ensemble to overpredict OA concentrations by a factor  
683 of 2 on average in the remote regions is consistent with the transition from the large  
684 underprediction in OA near the source region to a slight overprediction of OA in remote  
685 continental sites that was reported for most AeroCom-II models (Tsigaridis et al., 2014),  
686 and also observed for default parameterizations in other studies (Heald et al., 2011;  
687 Hodzic et al., 2016).

688 By comparison, the results of the ATom model ensemble show a much better agreement  
689 with observations. The model spread is still substantial, but mostly below a factor of 5.  
690 Figures S6 and S7 show OA vertical profiles for individual ATom models and the spread  
691 in their results. In most regions, the ATom model ensemble captures reasonably well both

692 the absolute concentrations as well as the shape of the vertical profiles. In the biomass  
693 burning outflow and NH mid-latitude regions, the ATom ensemble average better captures  
694 the higher OA concentrations in the boundary layer and lower OA concentrations in the  
695 lower stratosphere than the AeroCom-II ensemble. We note that using the ensemble  
696 median OA profiles instead of ensemble mean OA profiles (as shown in Figure 5 and S7)  
697 results in a slightly lower values of OA but does not change the conclusions of the model-  
698 measurement comparisons (Figure S18).

### 699 **4.3 Oxidation level of organic aerosols (OA/OC ratios)**

700 In addition to OA mass concentrations, we also evaluate the model's ability to simulate  
701 their degree of oxygenation, an indicator of their oxidation and aging (Aiken et al., 2008;  
702 Kroll et al., 2011). Ambient measurements of the oxidation level of organic particles are  
703 limited (Aiken et al., 2008, Canagaratna et al., 2015), and the ATom dataset provides the  
704 first global distribution of O/C and OA/OC ratios for the remote aerosol. The OA/OC ratio  
705 is an estimate of the average molecular weight of organic matter per carbon weight, and  
706 it mostly depends on the oxygen content (i.e. the O/C ratio), in the absence of significant  
707 concentrations of organonitrates and -sulfates. It is needed to compare measurements of  
708 organic aerosol mass (from e.g. AMS) with organic carbon measurements (from e.g.  
709 thermo-optical methods). It is also needed to compare the various types of measurements  
710 to model concentrations, which are sometimes carried internally as OA and sometimes as  
711 OC. A low OA/OC ratio is indicative of freshly emitted OA from fossil fuel combustion  
712 (typically  $\sim 1.4$ ), and its value increases with increased processing of organics in the  
713 atmosphere. Figure 5 shows that in the remote regions the bulk of measured OA/OC  
714 ratios during ATom-1 and -2 range between 2.2 and 2.5, and are larger than values of  $2.1$   
715  $\pm 0.2$  found in the polluted US continental outflow regions that were sampled during  
716 SEAC4RS, WINTER and DC3 field campaigns (Schroder et al., 2018). These values  
717 indicate that remote OA is highly oxidized and chemically processed.

718 Note that for organosulfates (R-O-SO<sub>2</sub>H and organonitrates (R-O-NO<sub>2</sub>, pRONO<sub>2</sub> in the  
719 following) only one oxygen is included in the reported OA/OC, as the fragments of these  
720 species are typically the same as for inorganic species in the AMS (Farmer et al., 2010).  
721 However in ATom organosulfates are estimated to account for  $\sim 1\%$  of the total sulfate  
722 (based on PALMS data, see Liao et al., 2015 for the methodology). Since sulfate and OA  
723 concentrations are comparable, organosulfates would only increase the OA/OC by  $\sim 1\%$   
724 on average. Organonitrates are reported from the AMS for ATom. Their impact on OA/OC

725 is not propagated for the default values, to maintain consistency with a large set of OA/OC  
726 measurements by AMS in the literature, and since they would increase OA/OC on average  
727 by only 4.5% (ATom-1) and 2.2% (ATom-2), which is smaller than the uncertainty of this  
728 measurement. However, we show the results with both methods in Fig. 5 to fully document  
729 this topic.

730 Importantly, this ratio is also used to calculate the total OA mass concentration for models  
731 that provided their outputs in terms of organic carbon concentrations ( $[OA]_i = [OC]_i \times$   
732  $OA/OC_{ratio}$ ). Most Models use a constant OA/OC ratio, but the value used varies  
733 substantially. OA/OC of 1.4 is used in ECHAM6-HAM, whereas 1.8 is used in GEOS5 and  
734 GC10-TOMAS simulations for both POA and SOA. Other models calculated directly SOA  
735 concentrations without applying this conversion (CESM1-CARMA, CESM2-SMP, CESM2-  
736 DYN, GC12-REF and GC12-DYN), but for POA used the ratio of 1.8 (CESM1-CARMA,  
737 CESM2-SMP, CESM2-DYN) and 2.1 (GC12-REF and GC12-DYN). Most of the AeroCom-  
738 II models used the ratio of 1.4 for all primary and secondary OA (Tsigaridis et al., 2014).  
739 The comparison with measurements shows that the measured values are ~40% larger  
740 than those assumed in some of the ATom models, and 60-80% larger than used in  
741 AeroCom-II models. The comparison between the observed and predicted OA/OC vertical  
742 profiles (Fig. S3) shows that AeroCom-II models tend to generally underpredict this ratio,  
743 and do not capture its increase in remote regions. As a result, this underestimation of  
744 OA/OC ratios and the use of a constant value could substantially impact the comparisons  
745 of OA mass concentrations for several models considered in this study (ECHAM6-HAM,  
746 GEOS5, CESM1-CARMA and GC10-TOMAS). If we correct for the underestimated  
747 OA/OC ratio using the ATom measured values of 2.2 (to be conservative) and compare  
748 to previously discussed biases in Table 2, the overprediction of the ECHAM6-HAM model  
749 is increased to ~110-160%, and that of GC10-TOMAS to 180% during ATom-2 while  
750 having ~15% bias in ATom-1, whereas GEOS5 results now overestimate up to 30% during  
751 ATom1, and perform much better during ATom-2.

752 These results demonstrate that current global chemistry-climate models use unrealistically  
753 low OA/OC ratios, which results in a large underestimate of the degree of oxidation of OA  
754 in remote regions. Inaccurate prediction of OA oxidation as it ages could impact not only  
755 the calculations of OA burden, but also its optical properties as the absorption of OA  
756 changes with its degree of oxidation (through the formation and destruction of brown

757 carbon, Laskin et al., 2015, Forrister et al., 2015). However, models used in this study did  
758 not include these effects.

#### 759 **4.4 Contribution of primary vs. secondary OA**

760 We further assess whether global models can adequately predict the relative contributions  
761 of primary and secondary OA. We strive to quantify these fractions with the most  
762 straightforward methods (with the fewest assumptions) for both models and  
763 measurements. POA concentrations were estimated from the BC measurements by using  
764 an emission ratio appropriate to the air mass origin (biomass burning vs. anthropogenic),  
765 as quantified by the  $f(\text{BB})$  mass fraction from the PALMS single particle instrument (see  
766 Section 3.2), with  $f(\text{BB})=1$  taken as a BC and OA being of pure BB air mass origin and  
767  $f(\text{BB})=0$  exclusively from a non-biomass burning source. By using the POA/BC ratio at the  
768 source regions after most evaporation, but before POA chemical degradation evaporation  
769 has taken place, we implicitly assume POA to be chemically inert, while in reality it can  
770 slowly be lost to the gas-phase by heterogeneous chemistry (e.g. George and Abbatt,  
771 2010; Palm et al., 2018). Thus, the observation-based method provides an upper limit to  
772 the fraction of POA. The model/measurement comparison is only shown for the CESM  
773 and GEOS-Chem model variants, as other participating models do not separate or did not  
774 report their POA and SOA fractions. In all simulations, POA was treated as a chemically  
775 inert directly emitted primary aerosol species that only undergoes transport,  
776 transformation from hydrophobic to hydrophilic state with ageing (1-2 days typically),  
777 coagulation, and dry and wet deposition. Importantly, the treatment of POA as non-volatile  
778 (rather than semi-volatile) in models is fully consistent with the assumptions for POA  
779 estimation from the measurements.

780 Figure 6 compares the vertical profiles of measurement-derived POA during ATom-1 and  
781 predicted by the CESM2-DYN model over clean remote regions of the Pacific basin and  
782 northern polar Atlantic that are not influenced by biomass burning. Comparisons for other  
783 models are similar (not shown). Observations show that POA is extremely small in remote  
784 regions, whereas the model predicts that about half of the OA is made of POA in those  
785 areas. Although the model reproduces quite well the measured total OA, it tends to  
786 severely overpredict the amount of POA and underpredict that of SOA over clean remote  
787 regions (with the two errors canceling each other when it comes to total OA). Over the  
788 biomass burning regions (not shown here) it can be difficult to directly quantify POA and  
789 SOA with this method, as total OA remains about constant, while POA decreases with



790 aging and SOA increases (Cubison et al., 2011; Jolleys et al., 2015; Hodshire et al.,  
791 2019b). However, given this evolution the method used here would lead to an  
792 overestimate of POA for this reason.

793 A more general comparison is made in Figure 7, using the frequency distributions of the  
794 measured and simulated fraction of POA/OA, for the free troposphere only (Figure S8  
795 shows the corresponding cumulative distributions). Observations indicate that most  
796 remote FT airmasses contain less than 10% POA, except for biomass burning plumes that  
797 are considered mostly primary. A slightly higher proportion of POA is seen in ATom-2,  
798 which is consistent with a slower photochemical production of SOA during NH winter.  
799 These results indicate that the remote OA is consistently dominated by SOA regardless  
800 of the season and location. The comparison with models reveals a very large discrepancy  
801 in the predicted vs. measured POA vs. SOA contributions. Models have a general  
802 tendency to severely overpredict the fraction of POA and underpredict that of SOA,  
803 displaying a much wider frequency distribution than the measurements (as also shown for  
804 POA and SOA vertical profiles for individual models on Figures S6 and S7). In GC12-REF,  
805 CESM2-DYN and CESM1-CARMA (without improved in-cloud removal) predictions for  
806 ATom-1, more than a half of the remote OA is POA, while that is very rarely observed in  
807 the free troposphere (possibly only during strong biomass burning events). Most models  
808 fail to reproduce the overwhelming dominance of SOA that is inferred from the  
809 measurements during ATom-1, while the discrepancies are less severe during NH winter  
810 (ATom-2). These seasonal differences suggest that model errors could be partially due to  
811 inefficient production of SOA and/or too high POA emissions, although removal errors also  
812 probably play a major role (see next section).

813 The differences are so large that they are pretty insensitive to details of the POA estimation  
814 method from the measurements, mostly because for the vast majority of the ATom track  
815 BC/OA ratios were extremely low and hence the exact magnitude of the multiplicative  
816 factor is secondary to the estimation of POA (Figure S11). As Figure S9 illustrates, the  
817 choice of  $FF_{\text{ratio}}$  has very little impact on the overall distribution of POA. On the other hand,  
818 while the  $BB_{\text{ratio}}$  does impact the overall distribution of POA, it mostly affects the points in  
819 the vicinity of the large Atlantic plumes. Since the POA/BC ratio in those plumes is fairly  
820 low, (see Section 3.2), using a very large  $BB_{\text{ratio}}$  mostly leads to an increase of the fraction  
821 of the points where POA > 100%. While the large range of published  $BB_{\text{ratio}}$  for different  
822 sources precludes a more accurate estimation by our method, for the purposes of the

823 comparison with the model results we emphasize that even using the largest  $BB_{ratio}$ ,  
824 fraction of SOA is still significantly larger in the ATom dataset than in any of the models.

825 Additional sensitivity tests were performed to investigate the impact of noisy data and  
826 uncertainties of  $f(BB)$  on the estimation of POA. Figure S11 clearly shows that the impact  
827 of a misattribution of the aerosol type by the stated PALMS uncertainty (Froyd et al., 2019)  
828 is completely negligible. Figure S10 details how the choice of averaging interval (with  
829 longer averaging times reducing both the fraction of OA measurements under the DL and  
830 below zero) impact the distribution of POA. Overall, no large changes are observed for  
831 averaging times  $>5$  min, and hence a 5 min averaging interval was used for the analysis  
832 in Figure 7. Figure S10 also illustrates how capping the histogram impacts the POA  
833 distribution. To capture the most realistic  $f_{(POA)}$  distribution, the data in Fig 7 was capped  
834 at the extremes (so  $f_{(POA)} < 0$  is taken as  $f_{(POA)} = 0$ , and  $f_{(POA)} > 1$  is taken as  $f_{(POA)} = 1$ ). As Figure  
835 S10 shows, data with  $f_{(POA)} < 0$  is almost exclusively due to very small (and always positive,  
836 since BC cannot go negative) POA values being divided by small, negative noise in total  
837 OA, and hence treating that fraction of the histogram as essentially  $f_{POA} \sim 0$  is justified.  
838 On the other end of the distribution, data where POA is larger than OA is mostly due to  
839 our average  $BB_{ratio}$  being larger than the one encountered in most of the BB plumes in  
840 ATom. Choosing a lower  $BB_{ratio}$ , as Figures S9b and S9d illustrate, leads to  $f_{(POA)} > 1$   
841 basically trending to zero, confirming our interpretation. This is a limitation of the dataset,  
842 and it does not seem appropriate to remove these points, since some fraction are likely  
843 dominated by POA. However, it shows that the POA estimation, especially for this part of  
844 the distribution likely overstates the importance of POA.

845 A comparison between simulations that have the same treatment of POA, and only differ  
846 in their chemistry and removal of SOA (e.g. CESM2-SMP vs. CESM2-DYN; GC12-REF  
847 vs. GC12-DYN) indicate that a more complex SOA treatment does not always result in a  
848 more accurate simulation of the primary/secondary character of OA, a result that was also  
849 found in the AeroCom-II multi-model intercomparison (Tsigaridis et al., 2014).

850 Finally, we have examined whether the non-volatile treatment of POA in models could  
851 lead to these unrealistically high POA fractions in the remote regions. Figure S16 shows  
852 a comparison of POA vertical profiles as predicted by the GC12-REF simulations that use  
853 non-volatile POA and a sensitivity simulation GC12-REF-SVPOA that uses semi-volatile  
854 POA similar to the standard treatment in GEOS-Chem as described in Pai et al. (2020).  
855 Note, however, that Pai et al. (2020) included marine POA emissions, used different

856 reanalysis meteorology, and a different model version (12.1.1 rather than 12.0.1 here), so  
857 their resulting comparisons to ATom measurements are somewhat different than found  
858 here for GC12-REF-SVPOA. The comparison indicates that the POA concentrations  
859 increase substantially in most regions when the semi-volatile POA parameterization is  
860 used. These results suggest that non-volatile treatment of POA is not responsible of the  
861 model bias.

#### 862 **4.5 Sensitivity to OA formation and removal**

863 In this section, we further investigate some of the possible reasons for the incorrect model  
864 predictions of the relative contributions of POA and SOA in remote regions. Given the  
865 tendency of models to underestimate OA close to anthropogenic source regions and  
866 overestimate OA downwind in past studies (e.g. Heald et al., 2011; Tsigaridis et al., 2014;  
867 Hodzic et al., 2016), in this section we investigate the sensitivity of OA to increasing  
868 sources and increasing removals. We have performed two additional model simulations  
869 to test the sensitivity of the POA/SOA fractions to uncertainties in the representation of (i)  
870 wet scavenging, based on the CESM1-CARMA simulations in which we have removed  
871 the improvements in the aerosol removal by the convective updrafts (Yu et al., 2019); and  
872 of (ii) SOA formation based on the GC12-REF simulations in which we have replaced the  
873 SOA formation VBS mechanism (Pye et al., 2010) by an updated VBS mechanism that  
874 uses chamber wall-loss corrected SOA yields (Hodzic et al., 2016, same formation  
875 scheme that is used in GC12-DYN and CESM2-DYN runs, but with removals kept identical  
876 to GC12-REF). The results of these two sensitivity simulations are displayed on Figure 8,  
877 which shows measured and predicted mass concentrations of OA, POA, SOA and sulfate  
878 for ATom-1 as a function of the number of days since the air mass was processed through  
879 convection. One should keep in mind that this is an averaged plot that included airmasses  
880 from various regions and altitudes, and not a Lagrangian plot following the same airmass.

881 **Sensitivity to in-cloud scavenging in convective clouds.** Inefficient wet removal of  
882 primary OA could contribute to the POA overprediction in global models, especially in the  
883 tropics. Previous global model studies have reported two to three orders of magnitude  
884 overestimation of primary carbonaceous species such as BC in the free troposphere when  
885 the removal in the convective updrafts was not included (e.g. Schwarz et al., 2013, Yu et  
886 al., 2019). A strong reduction due to convective removal is also expected for POA  
887 concentrations, as POA is a primary species co-emitted with BC at the surface and  
888 internally mixed with it (Lee et al., 2015), and that is typically coated by secondary

889 inorganics and organics over short timescales (Petters et al., 2006; Jiang et al., 2010;  
890 Wang et al., 2010). Figures 7a and 8 compare the simulations of CESM1-CARMA with  
891 and without improved convective in-cloud scavenging during ATom-1. The improved in-  
892 cloud scavenging scheme considers aerosol activation into cloud droplets from entrained  
893 air above the cloud base, which is more realistic and results in a more efficient removal of  
894 aerosols in the upper troposphere by convection. E.g. a two order of magnitude reduction  
895 in BC in the upper FT was reported by Yu et al. (2019), resulting in much improved  
896 agreement with observations. Similar results were observed for sea salt aerosols in  
897 Murphy et al. (2019). Figure 8 shows that all submicron aerosol species simulated in  
898 CESM1-CARMA are strongly impacted by the in-cloud removal above the cloud base.  
899 POA concentrations are reduced by an order of magnitude while sulfate is reduced by  
900 30% leading in both cases to a much-improved agreement with observations. SOA is  
901 reduced by ~30% as well, which leads to an underprediction of measured SOA  
902 concentrations. The overall impact on OA concentrations is a significant reduction, which  
903 leads to ~20% underestimation of OA in the aged remote air during ATom-1.

904 For the CESM2-DYN model that does not have improved in-cloud removal, the reasonable  
905 agreement (within 20%) with the observed OA concentrations thus results from  
906 coincidental error compensation between the overpredicted POA and underpredicted  
907 SOA. The prescribed SOA formation and the artificial 50% adjustment of SOA emissions  
908 based on Liu et al. (2012) in CESM2-SMP leads to an overestimation of observed SOA in  
909 aged remote airmasses.

910 **Sensitivity to SOA formation.** In addition, we have also tested the sensitivity of the OA  
911 composition to the choice of the SOA formation mechanism. Figure 8 compares the results  
912 of the GC12-REF model that uses SOA formation yields derived from traditional chamber  
913 experiments (Pye et al., 2010) and those corrected for losses of organic vapors onto  
914 chamber walls as proposed in Hodzic et al. (2016). Previous studies have reported that  
915 chamber wall losses could lead up to a factor of 4 underprediction of formed SOA (Zhang  
916 et al., 2014; Krechmer et al., 2016). It should be noted that in both cases, isoprene-SOA  
917 is formed in aqueous aerosols following Marais et al. (2016). The comparison shows a  
918 factor of 3 increase in SOA concentrations when the updated SOA formation is considered  
919 leading to a much better agreement with the observed SOA as well as the observed total  
920 OA. GC12-REF predicts well the amount of POA and overpredicts somewhat the amount  
921 of sulfate aerosols, which is expected as it already includes the improved aerosol removal

922 in convective updrafts (Wang et al., 2014). Figure S6 also shows that POA vertical  
923 distribution is well captured in GEOS-Chem in most regions, except over the polar north  
924 Pacific. It should be noted that these results are consistent with the POA/OA frequency  
925 distribution shown in Figure 7 (the POA/OA ratio predicted by GC12-REF is larger than  
926 the measured ratio, which is consistent with the fact that POA is about the right amount,  
927 and OA is underpredicted in Figure 8).

928 These sensitivity simulations suggest that a stronger convective removal of POA and a  
929 stronger production of SOA might be needed to correctly represent not only the total OA  
930 concentrations but also its primary and secondary nature in remote free troposphere and  
931 remote ocean regions. Accurate predictions of the OA concentration, composition, and  
932 source contributions for the right reasons are key for accurately predicting their lifecycle  
933 and radiative impacts. Only when there is confidence that the sources are accurately  
934 predicted, we can have confidence in OA predictions for pre-industrial and future  
935 conditions, as well as to evaluate PM mitigation strategies.

#### 936 **4.6 OA and sulfate relative contributions in FT**

937 Finally, we assess the model ability to predict relative amounts of OA and sulfate in the  
938 free troposphere where they are the two major constituents of the submicron aerosol  
939 (Figure 2b). Accurate predictions of their relative contributions are crucial to determine the  
940 hygroscopicity of the submicron aerosol, and its ability to serve as a cloud condensation  
941 nuclei (CCN) in the remote free troposphere (Carslaw et al., 2013; Brock et al., 2016).

942 Figure 9a compares the average measured relative fractions of sulfate (36%) and  
943 carbonaceous aerosols (OA=59% and BC=5%) in the FT with those predicted by  
944 individual models during ATom-1. The CESM2 models best reproduce the observed  
945 relative contributions, with a slight underestimation of OA (57% instead of 59%) for  
946 CESM2-DYN, and a slight overestimation of OA (63% instead of 59%) for CESM2-SMP.  
947 GEOS5 has 15% more OA relative to sulfate than observed. All other models  
948 underestimate both OA and BC relative fractions. For instance, in GC12-REF and -DYN,  
949 both the BC and OA fractions are ~40% (relative) lower than observed.

950 Figure 9b shows the frequency distribution of observed and predicted fractions of OA  
951 relative to sulfate during ATom-1 and -2 in the free troposphere. Most models fail to  
952 reproduce the relatively uniform nature of the observed distributions during ATom-1, with  
953 typically narrower model shapes around a preferred ratio. The NH summer measurements

954 indicate that OA > sulfate in ~55% of the samples (consistent with Fig. 2b), while models  
955 generally tend to underestimate the relative OA contribution. In particular, GEOS-Chem  
956 and ECHAM6-HAM tend to overestimate the relative contribution of sulfate. A better  
957 agreement is found for GEOS5, CESM1-CARMA and CESM2-DYN, which follow more  
958 closely the shape of the observed distribution. The comparisons also suggest that the  
959 more complex SOA treatment of SOA formation and removal proposed by Hodzic et al.  
960 (2016) in the same host model leads to an improved agreement with observations (e.g.  
961 CESM2-DYN vs. CESM2-SMP; GC12-DYN vs. GC12-REF). It should be noted that  
962 CESM2-SMP uses fixed SOA yields that were increased by 50% as suggested by Liu et  
963 al. (2012), leading to an overestimation of the relative contribution of OA compared to that  
964 of sulfate in the free troposphere. During the NH winter (ATom-2), measurements show a  
965 somewhat higher proportion of sulfate aerosols (vs. ATom-1), which is consistent with a  
966 slower production of SOA in the NH during winter and a reduced influence of biomass  
967 burning. Similar conclusions are found for the evaluation of different models. It is worth  
968 mentioning that the comparison performed for the whole ATom-1 and 2 dataset (not  
969 shown) leads to similar results with even slightly stronger overestimation of the sulfate  
970 relative contribution compared to OA.

971 The discrepancies between the observed and predicted composition of submicron aerosol  
972 over remote regions can be quite large for other constituents as well. Figure 10 shows the  
973 comparison of measured and predicted composition of the submicron aerosol over the  
974 Southern Ocean (during the NH winter) where the disagreement in simulated sea salt,  
975 nitrates, ammonium, and MSA often exceeds the contribution of OA. While the  
976 observations show a more uniform distribution of non-marine aerosol with higher values  
977 in the mid and upper troposphere, respectively, most models tend to simulate highest  
978 fractions of OA (and sulfate) towards the tropopause. This may also be explained by the  
979 uncertainties in modeled wet removal of aerosol that has been discussed above. Specific  
980 studies have discussed and continue to investigate the ATom measurements and  
981 simulations of different components in more detail, including particle number (Williamson  
982 et al., 2019), black carbon (Katich et al., 2018; Ditas et al., 2019), MSA (Hodshire et al.,  
983 2019), sulfate-nitrate-ammonium (Nault et al., 2019), and sea salt (Yu et al, 2019; Bian et  
984 al., 2019; Murphy et al., 2019).

985

## 986 **5 Conclusions and implications**

987 Our understanding and representation in global models of the lifecycle of the OA remain  
988 highly uncertain, especially in remote regions where constraints from measurements have  
989 been very sparse. We have performed a systematic evaluation of the performance of eight  
990 global chemistry climate models and of 28 AeroCom-II models in simulating the latitudinal  
991 and vertical distribution of OA and its composition in the remote regions of the Atlantic and  
992 Pacific marine boundary layer, free troposphere and lower stratosphere, using the unique  
993 measurements from the ATom campaign. Our simulations are conducted for both ATom-  
994 1 and ATom-2 deployments that took place in August 2016 and February 2017,  
995 respectively. The main conclusions of the comparison are as follows:

- 996 • The AeroCom-II ensemble average tends to be biased high by a factor of 2-5 in  
997 comparison to measured vertical OA profiles in the remote atmosphere during both  
998 NH summer and NH winter. The ensemble spread increases from a factor of 40 in the  
999 NH source regions to a factor of 1000 in remote regions of the Southern Ocean. The  
1000 evaluation of AeroCom-II models in the remote regions provides an extension of the  
1001 previous evaluation with continental ground data by Tsigaridis et al. (2014). We note  
1002 that the data from the AeroCom-II models were based on monthly mean values from  
1003 a different simulated year than the ATom campaigns; however, the consistent model  
1004 biases are strong enough that we would not expect our conclusions to change for a  
1005 different modeled year.
- 1006 • The results of the ATom model ensemble used in this work show a much better  
1007 agreement with the OA observations in all regions and reduced model variability.  
1008 However, some of the agreement is for the wrong reasons, as most models severely  
1009 overestimate the contribution of POA and underestimate the contribution of SOA to  
1010 total OA. Sensitivity simulations indicate that the POA overestimate in CESM could be  
1011 due to an inadequate representation of primary aerosol removal by convective clouds,  
1012 (additional convective removal per Yu et al. (2019) in CESM1-CARMA led to a better  
1013 agreement with observations). Most models have insufficient production of SOA, and  
1014 sensitivity studies indicate that a stronger production of SOA is needed to capture the  
1015 measured concentrations. The photochemical ageing of POA which was not  
1016 considered here (unlike for SOA) could also contribute to the model overestimation.  
1017 The non-volatile POA treatment in models is consistent with the assumption of inert  
1018 POA particles used to estimate POA from measurements, and cannot explain the

1019 model bias. Indeed, sensitivity simulations with semi-volatile POA lead to a much  
1020 larger model bias for OA in the upper troposphere and remote regions. The  
1021 compensation between errors in POA and SOA in remote regions is however a  
1022 recurring issue in OA modeling (de Gouw and Jimenez, 2009). For instance, it was  
1023 found in the urban outflow regions such as Mexico City during MILAGRO 2006 field  
1024 campaign (Fast et al., 2009; Hodzic et al., 2009); Paris during MEGAPOLI 2009  
1025 (Zhang et al., 2013); the Los Angeles area during CalNex-2010 (Baker et al., 2015;  
1026 Woody et al., 2016); the NE US outflow during WINTER 2015 (Schroder et al., 2018;  
1027 Shah et al., 2019).

1028 • Additional errors in simulated OA concentrations can arise from the use of too low  
1029 OA/OC ratios when model results (often calculated as OC) are converted to OA for  
1030 comparison with measurements. We note that OA is the most atmospherically-relevant  
1031 quantity, while OC is an operational quantity, partially a relic from a period in which  
1032 only OC could be separately quantified (although also of some use for carbon budget  
1033 studies). It should also be noted that most emission inventories still use OC as the  
1034 primary variable, which is why the use of accurate OA/OC ratios is still key for all  
1035 models. We show that the OA/OC ratio used in most models is too low compared to  
1036 measured values that range mostly from 2.2 to 2.5, resulting in errors in OA mass of  
1037 ~70% for AeroCom-II models and ~30% for current models that use organic carbon to  
1038 track OA mass. Remote OA is thus highly oxidized and chemically processed. These  
1039 results demonstrate that current global chemistry-climate models underestimate the  
1040 degree of oxidation of OA in remote regions and need to consider further chemical  
1041 ageing of OA, which could impact the calculations of its burden, and optical and  
1042 hygroscopic properties.

1043 • The results also show that in most models (except CESM2) the predicted OA  
1044 contribution to the total submicron aerosol is underestimated relative to sulfate in the  
1045 remote FT where OA and sulfate are the dominant submicron aerosols (important for  
1046 climate). Accurate predictions of composition of submicron particles remains  
1047 challenging in remote regions and should be the topic of future studies.

1048 Key implications of our results are: (i) Model errors on the relative contribution of POA and  
1049 SOA to OA reduce our confidence on the ability to simulate radiative forcing over time or  
1050 OA health impacts; (ii) Model errors for the relative contributions of sulfate and organics  
1051 to the submicron aerosol in the free troposphere could lead to errors in the predicted CCN  
1052 or radiative forcing of aerosols as inorganics are more hygroscopic than OA; (iii) the OA



1053 system seems to be more dynamic with a need for an enhanced removal of primary OA,  
1054 and a stronger production of secondary OA in global models to provide a better agreement  
1055 with observations.

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1600

1601 Tables

1602 Table 1: ATom global model configurations and their treatment of the most important processes affecting organic aerosols.

Models & horizontal res. & met. fields & config. reference	Aerosol module	Submicron size <sup>(6)</sup> OA (dust/sea salt)	SOA precursors <sup>(1)</sup>					SOA production	Emission	POA/POC (SOA/SOC)	Removal		
			ISO	MT	SQ	ANT	C <sub>&gt;12</sub>				Standard <sup>(2)</sup>	Improved	Photolytic
CESM1-CARMA (1.9°lon x 2.5°lat) MERRA-2 (Yu et al. 2019)	20 bins	< 500 nm (< 800 nm)	x	x		x		Semi-volatile using VBS (Pye et al. 2010)	GAIS and GFED v3	1.8 (N/A)	x	For convective updrafts (Yu et al. 2019) <sup>(3)</sup>	
CESM2-DYN (0.9° lon x 1.25° lat) GEOS5 (Tilmes et al. 2019)	4 modes	< 270 nm (< 800 nm)	x	x	x	x	x	Semi-volatile using VBS (Hodzic et al. 2016)	CMIP6 and QFED v2.4	1.8 (N/A)	x	Water solubility of organic gases per Hodzic et al. (2014)	For SOA (Hodzic et al. 2016)
CESM2-SMP GEOS5 (0.9° lon x 1.25° lat) (Tilmes et al. 2019)	4 modes	< 270 nm (< 800 nm)	x	x		x		Non-volatile with prescribed mass yields for all precursors <sup>(4)</sup>	CMIP6 and QFED v2.4	1.8 (N/A)	x		
ECHAM6-HAM ECHAM6 (1.87°lon x 1.87°lat) (Tegen et al. 2019)	7 modes	< 500 nm (< 500 nm)		x				Non-volatile with 15% prescribed mass yields (Dentener et al. 2006)	ECLIPSE <sup>(5)</sup> and GFAS	1.4 (1.4)	x		
GC12-REF (2° lon x 2.5° lat) GEOS-FP (Bey et al. 2001)	Bulk	Bulk (< 500 nm)	x	x	x	x		Semi-volatile using VBS (Pye et al. 2010); non-volatile isoprene-	CMIP6 and GFED v4	2.1 (N/A)	x	For convective updrafts per Wang et al. 2014	

								SOA (Marais et al. 2016)					
GC12-DYN (2° lon x 2.5° lat) GEOS-FP (Bey et al. 2001)	Bulk	Bulk (< 500 nm)	x	x	x	x	x	Semi-volatile using VBS (Hodzic et al. 2016); non-volatile isoprene-SOA (Marais et al. 2016)	CMIP6 and GFED v4	2.1 (N/A)	x	For convective updrafts (Wang et al. 2014); Water solubility of organic gases (Hodzic et al. 2014)	For SOA (Hodzic et al. 2016)
GC10-TOMAS (5° lon x 4° lat) GEOS-FP (Kodros et al. 2016)	15 bins	< 316 nm (< 316 nm)		x			x	Non-Volatile using 10% mass yields for MT, 0.2 Tg SOA per Tg CO for anthropogenic emissions	EDGAR v4 and GFED v3	1.8 (1.8)	x	For convective updrafts (Wang et al. 2014)	
GEOS5 (0.5°lon x 0.625°lat) MERRA-2 (Bian et al. 2019)	Bulk	bulk (< 1 µm for dust, 500 nm for seasalt)	x	x			x	Non-Volatile, 10% mass yields for all precursors	HTAP and QFED v2.54	1.8 (1.8)	x		

- 1603 (1) SOA precursors include isoprene (ISO), monoterpenes (MT), sesquiterpenes (SQ), anthropogenics (ANT) including aromatics such as  
1604 benzene, toluene and xylene, as well as lumped shorter chain alkanes and alkenes; and higher molecular weight n-alkanes and n-alkenes  
1605 (C>12).  
1606 (2) Standard removal includes dry deposition and sedimentation, as well as convective and large-scale scavenging of soluble organic gases  
1607 and aerosols, and below-cloud scavenging of aerosols.  
1608 (3) A sensitivity simulation is performed with CESM1-CARMA without the improved scavenging in convective updrafts.  
1609 (4) 5% for lumped C<12 alkanes, 5% for lumped C<12 alkenes, 15% for aromatics, 4% for isoprene, 25% for monoterpenes.  
1610 (5) Anthropogenic BC emission are replaced in Russia with the dataset of Huang et al. (2015).  
1611 (6) Submicron size range (diameter) used in various models for comparison with the AMS data.

1612 Table 2: Comparison of observed and simulated OA concentrations along ATom-1 and  
 1613 ATom-2 flights for eight global model simulations and their ensemble. The results of the  
 1614 model ensemble are also indicated. The statistical indicators are calculated as normalized  
 1615 mean bias  $NMB(\%) = 100 \times \sum_i (M_i - O_i) / \sum_i O_i$ ; normalized mean error  $NME(\%) =$   
 1616  $100 \times \sum_i |M_i - O_i| / \sum_i O_i$ ; root mean square error  $RMSE(\mu g m^{-3}) =$   
 1617  $\sqrt{(1/N) \sum_i (M_i - O_i)^2}$  and correlation coefficient ( $R^2$ ) between modeled ( $M_i$ ) and observed  
 1618 ( $O_i$ ) data points. The mean of ATom-1 observations is  $\sim 0.23 \mu g m^{-3}$  and for ATom-2 is  
 1619  $0.11 \mu g m^{-3}$ . Figure S4 shows the normalized mean bias for all individual ATom model  
 1620 simulations for various latitudinal regions and for both the Atlantic and Pacific basins.

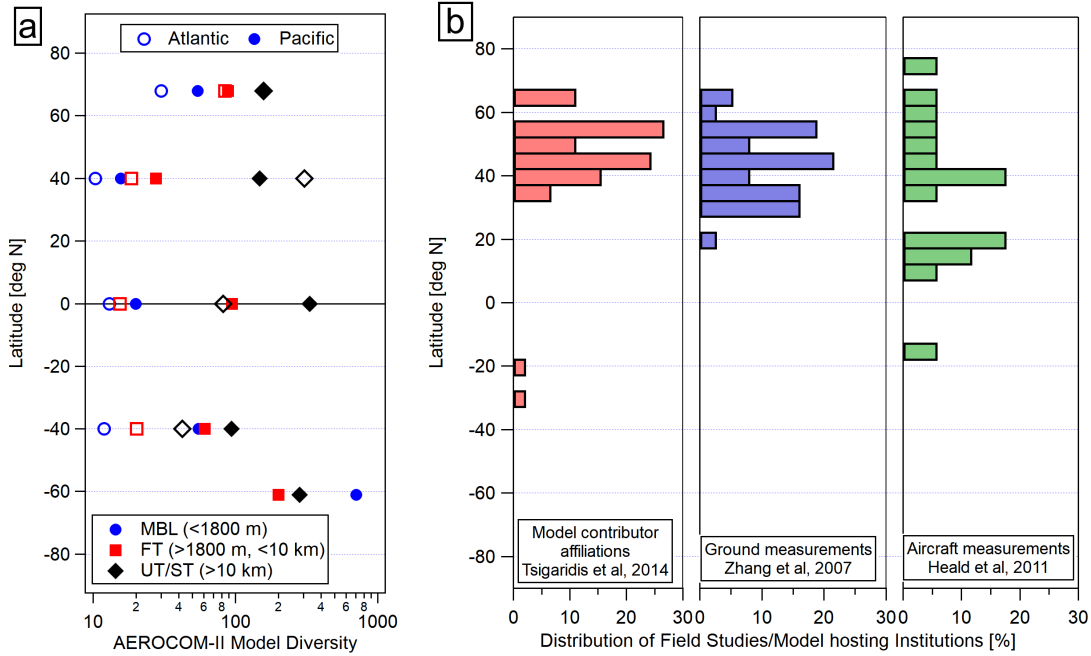
<b>Organic aerosols</b>	Avg.Mod. ( $\mu g m^{-3}$ )	NMB (%)	NME (%)	RMSE ( $\mu g m^{-3}$ )	$R^2$	Avg.Mod. ( $\mu g m^{-3}$ )	NMB (%)	NME (%)	RMSE ( $\mu g m^{-3}$ )	$R^2$
Model	<i>ATom-1 scores (August 2016)</i>					<i>ATom-2 scores (February 2017)</i>				
AeroCom-II Ens.	0.400	74.2	127.3	0.560	0.31	0.254	137	175	0.278	0.38
AeroCom-II Sub. <sup>(1)</sup>	0.335	47.0	111	0.557	0.28	0.242	127	178	0.290	0.27
ATom Ensemble	0.239	-4.5	64.6	0.372	0.66	0.139	23	92.6	0.224	0.48
CESM2-DYN	0.268	4.6	83.7	0.867	0.47	0.140	25.6	111.7	0.317	0.36
CESM2-SMP	0.349	36.3	94.3	0.556	0.51	0.175	57.2	125.4	0.299	0.31
CESM1-CARMA	0.155	-33.2	93.8	0.603	0.12	0.131	22.6	119.6	0.244	0.31
ECHAM6-HAM	0.400	73.6	143.6	0.714	0.24	0.214	100	184.0	0.363	0.23
GC12-DYN	0.142	-32.6	79.4	0.560	0.16	0.174	14.7	96.6	0.312	0.39
GC12-REF	0.122	-43.0	76.5	0.536	0.18	0.147	3.6	96.3	0.292	0.35
GC10-TOMAS	0.218	-14.4	86.5	0.644	0.16	0.313	150.0	223.7	0.537	0.12
GEOS5	0.242	-5.4	86.6	0.975	0.38	0.084	-24.9	86.4	0.268	0.29

1621 (1) This is the subset of AeroCom-II model ensemble that includes only seven  
 1622 models that are similar to those that are included in the ATom ensemble (either  
 1623 the same model, or an older model version, or the same aerosol module).  
 1624 AeroCom-II Sub. includes CAM5-MAM3, CCSM4-hem, ECHAM5-HAM2,

1625  
1626

GEOSChem-APM 8.2, GEOSChem 9, GISS-TOMAS and GMI (see Tsigaridis et al., 2014 for their description).

1627 Figures:



1628

1629 Figure 1: (a, left) The ratio between the average OA concentrations of the highest and the  
1630 lowest models (for each region) as predicted among 28 global chemistry transport models  
1631 participating in the AeroCom phase II intercomparison study (Tsigaridis et al. 2014); (b,  
1632 right) Geographical distribution of institutions at which the AeroCom-II models were  
1633 ran/developed (based on author affiliations) and of the field measurements included in two  
1634 major literature overview studies (Zhang et al., 2007; Heald et al., 2011) for the OA ground  
1635 and aircraft AMS as a function of latitude. For the aircraft campaigns, the average latitude  
1636 for the full deployment was taken.

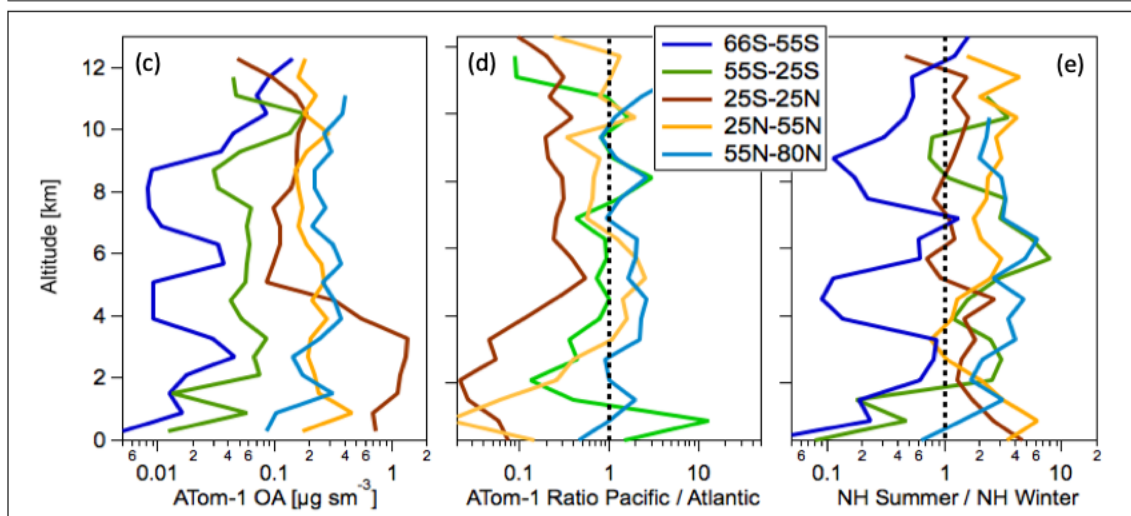
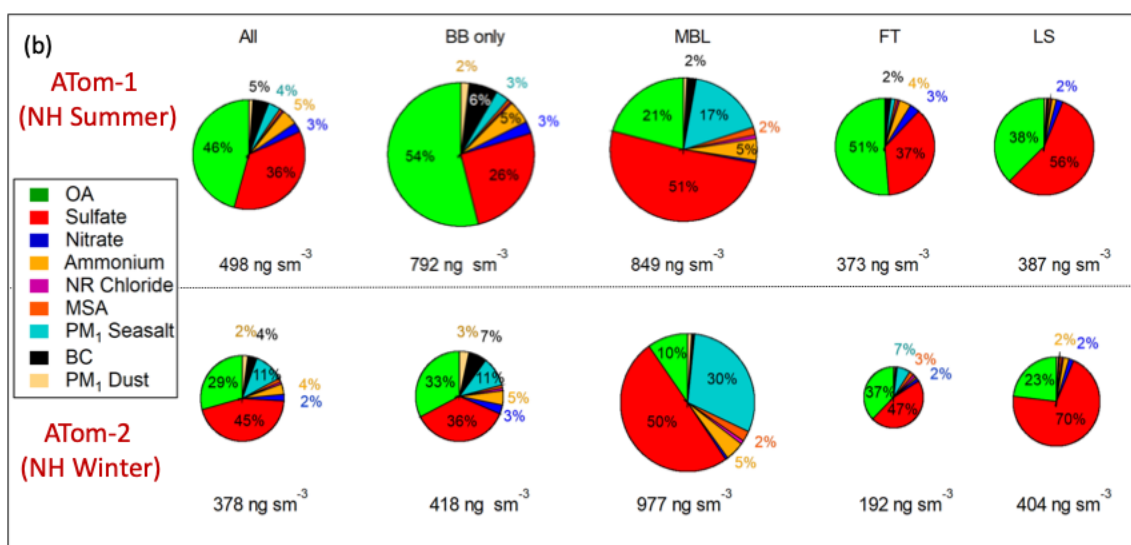
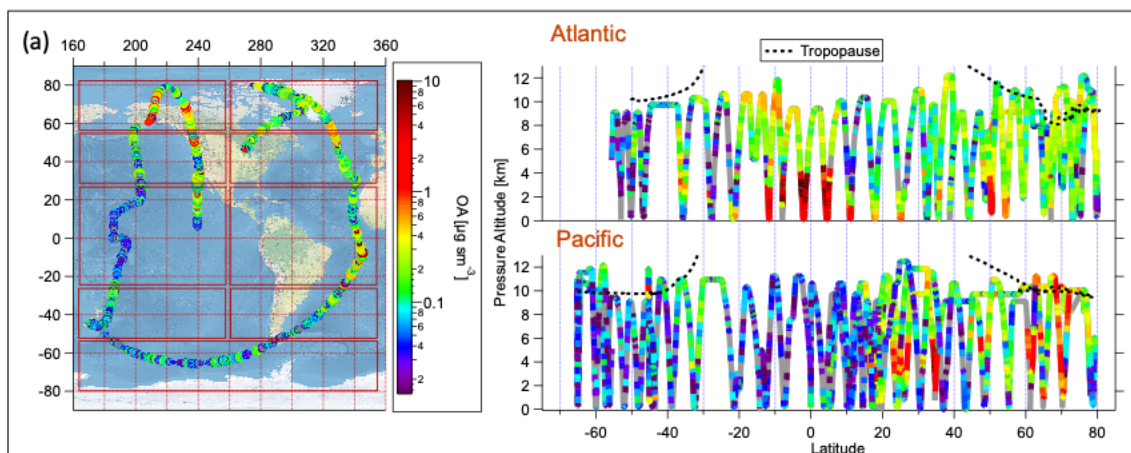
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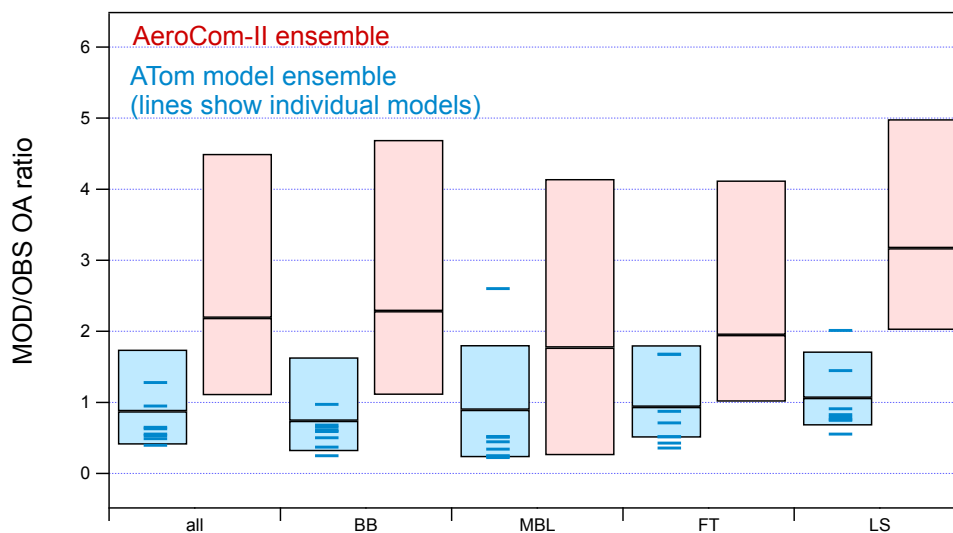
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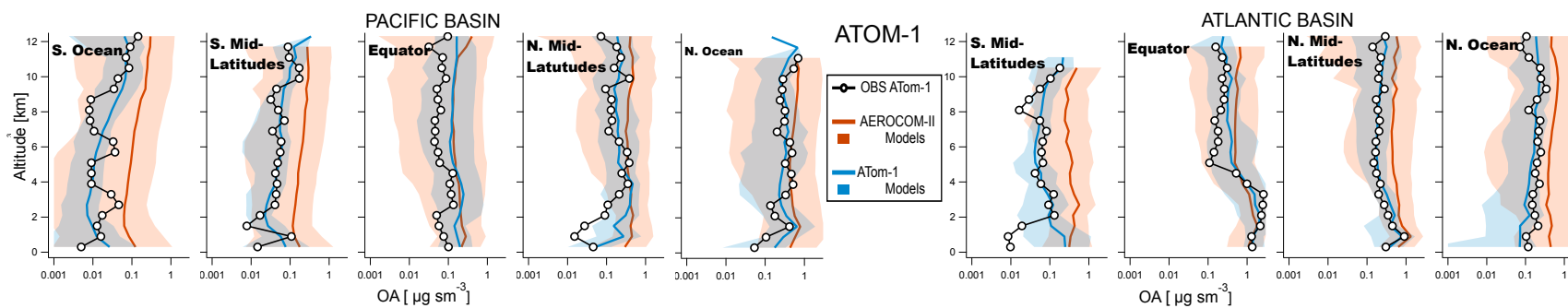
1643 Figure 2: (a, left) ATom-1 DC-8 flights during the August 2016 deployment. Red boxes  
 1644 indicate regions used for the latitude averaging of the model results. (a, right) Vertical  
 1645 distribution of OA concentrations ( $\mu\text{g sm}^{-3}$ ) along ATom-1 flight tracks (b) Average  
 1646 submicron aerosol composition as measured in the biomass-burning influenced regions  
 1647 (BB only), and the non-BB influenced regions including the marine boundary layer (MBL),  
 1648 free troposphere (FT), and lower stratosphere (LS) for ATom-1 (upper plots) and ATom-2  
 1649 (lower plots). The BB influenced airmasses were filtered using the PALMS data (see  
 1650 section 3.1). Contributions below 2% are shown but not labeled on the pie chart graph. In  
 1651 ATom-1, BB-only represents 24% of the data, clean MBL 8%, clean FT 57% and clean  
 1652 UT 12%, whereas in ATom-2 BB-only represents 3%, clean MBL 8%, clean FT 74%, clean  
 1653 UT 16%. (c) The average OA vertical profiles are shown for each latitude region as well  
 1654 as (d) the ratios between the Pacific and Atlantic Oceans in each region. (e) The seasonal  
 1655 contrast in OA concentrations as calculated as the ratio in OA concentrations between the  
 1656 NH summer (ATom-1) and NH winter (ATom-2) campaigns. The corresponding plots for  
 1657 ATom-2 can be found in Fig. S1.



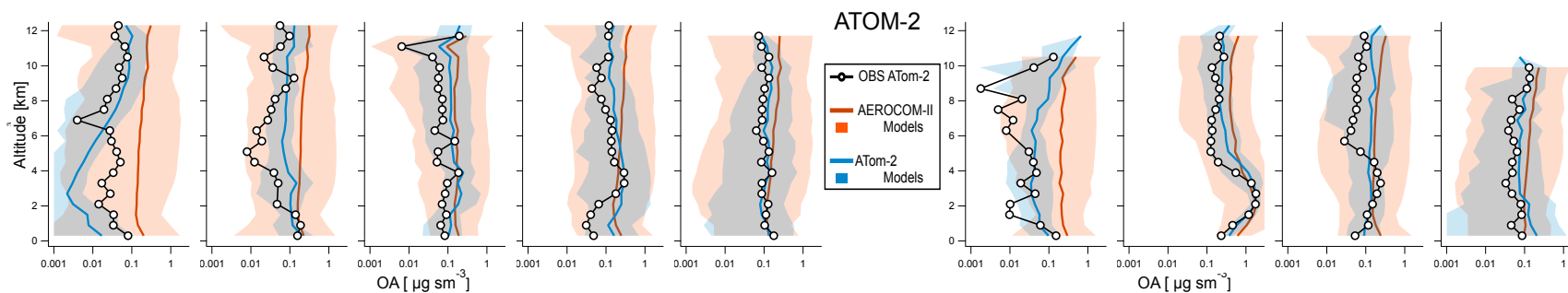
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1659 Figure 3: Ratios between predicted and observed OA concentrations for all ATom-1 flights  
 1660 as calculated for the ATom and AeroCom-II model ensembles in different regions (“BB”  
 1661 biomass burning influenced regions; “MBL” clean marine boundary layer; “FT” clean free  
 1662 troposphere’ and “LS” lower stratosphere). Median of the ensemble ratio is shown as a  
 1663 horizontal line, while the boxes indicate 25th and 75th percentiles. Medians for the  
 1664 individual models included in the current ATom model ensemble are also shown as blue  
 1665 lines.

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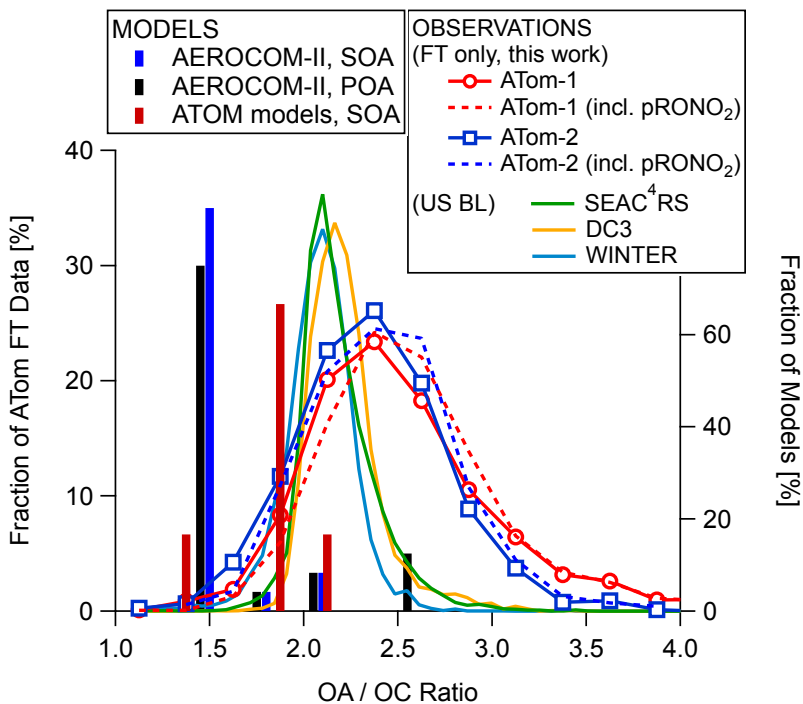


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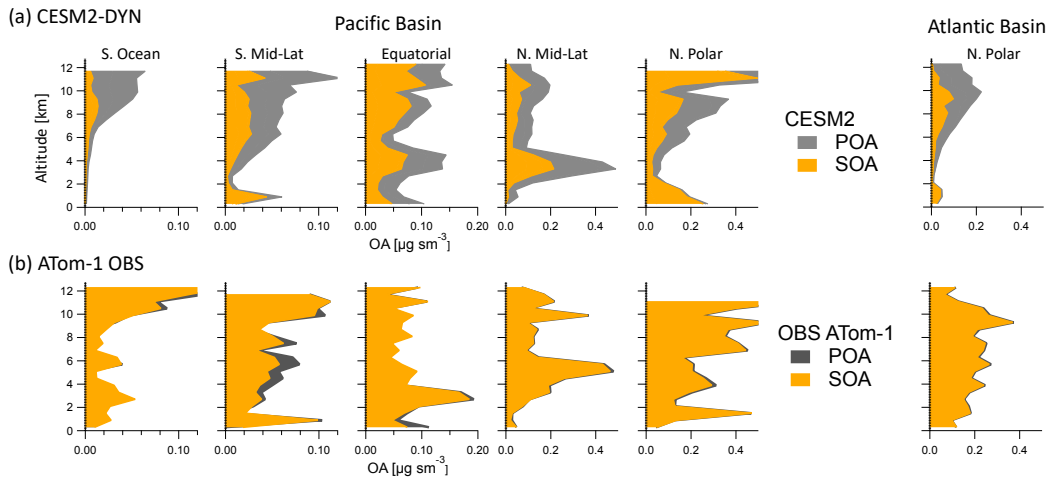
1668 Figure 4: Comparison of latitude-averaged predicted OA vertical profiles with ATom-1 and -2 measurements taken over the Pacific (left  
1669 side) and Atlantic (right side) basins. Results of the AeroCom-II model ensemble average are shown in red while those of the ATom  
1670 model ensemble are shown in blue. Shaded areas indicate the variability (two standard deviations) within each model ensemble.

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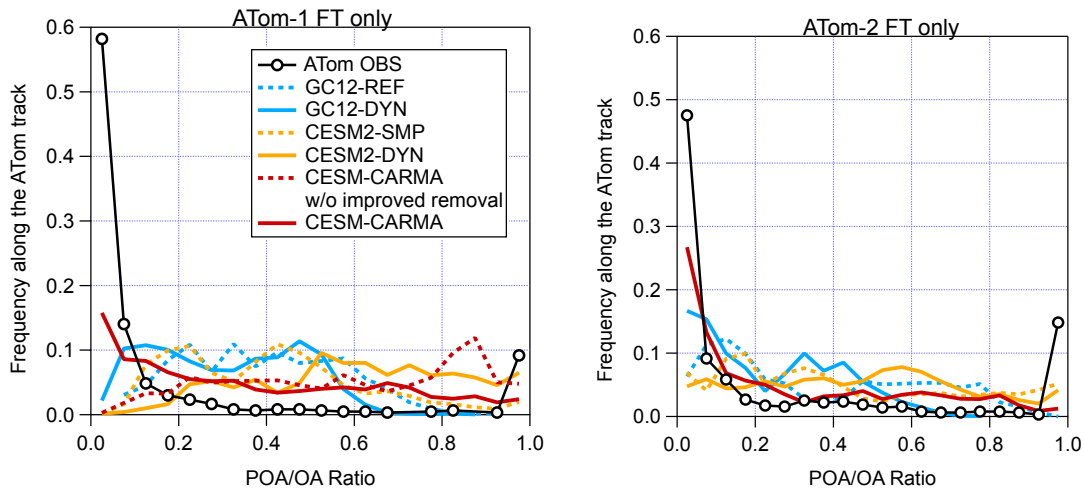
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1673 Figure 5: Distribution of the OA / OC ratio as measured during ATom-1 and -2. Values for  
1674 the recent aircraft campaigns (SEAC<sup>4</sup>RS, DC3 and WINTER) that took place over  
1675 continental US regions closer to continental source regions are also shown (Schroder et  
1676 al., 2018). The bars (right axis) show the OA/OC used for SOA and POA by the models  
1677 included in the AeroCom and ATom ensemble, with OA/OC=1.4 being the modal value for  
1678 the former and 1.8 for the latter.



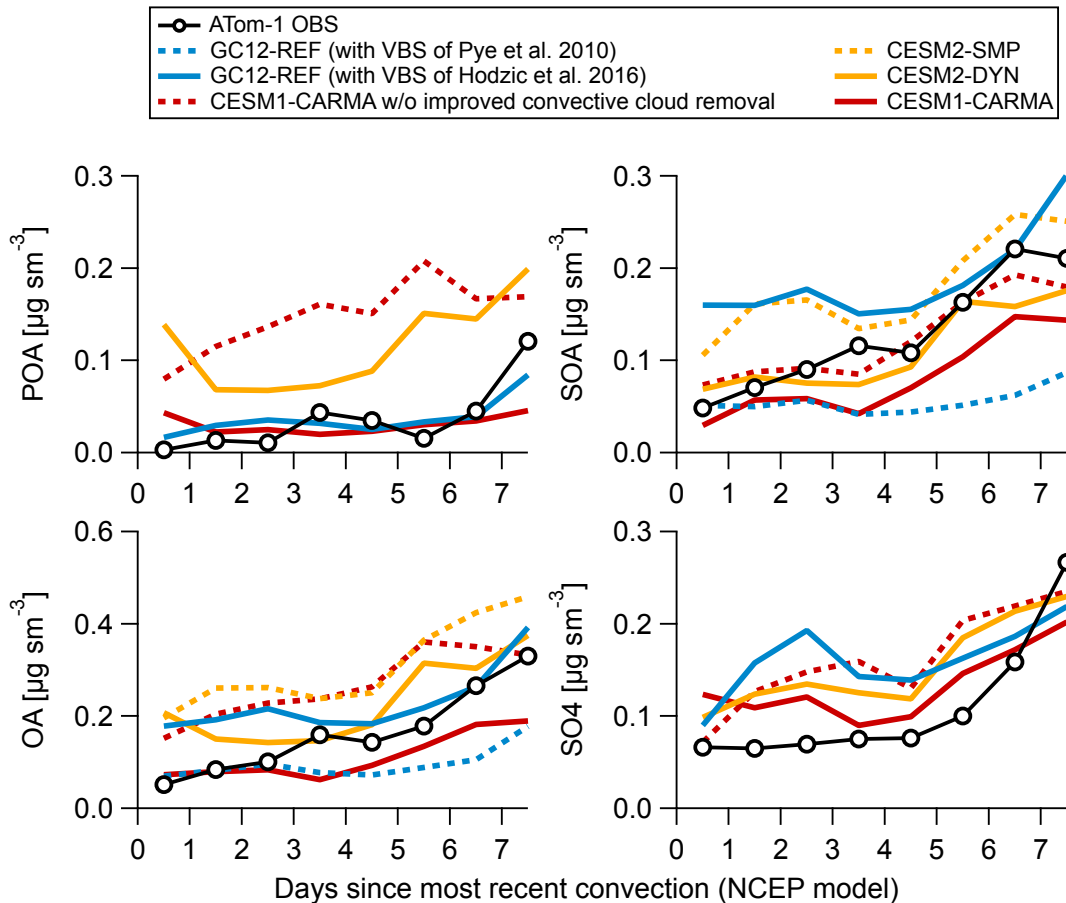
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1680 Figure 6: Comparison of averaged POA and SOA vertical profiles as observed during  
 1681 ATom and as predicted by the CSM2-DYN model over the non-BB influenced Pacific  
 1682 and Atlantic basins. The comparison is not shown for the strongly biomass burning  
 1683 influenced regions as all the OA is conservatively allocated to POA in those regions.



1684

1685 Figure 7: Frequency distribution of observed and simulated ratio of POA to total OA in the  
 1686 free troposphere during ATom-1 and ATom-2 as computed by the GC12-, CSM2-, and  
 1687 CSM1-CARMA models.



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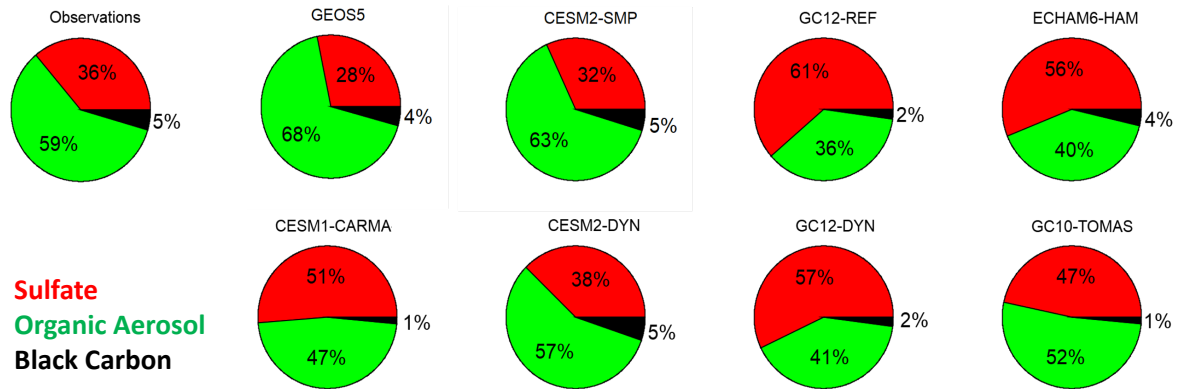
1689 Figure 8: Measured and predicted mass concentrations of POA, SOA, OA and sulfate  
 1690 aerosols during ATom-1 as a function of the number of days since the air mass was  
 1691 processed through convection (based on a trajectory model from Bowman, 1993, and  
 1692 satellite cloud data from NASA Langley, <https://clouds.larc.nasa.gov/>). CESM2-SMP and  
 1693 CESM2-DYN have the same emissions and processing of POA and sulfate, and thus  
 1694 similar concentrations. The same is true for the two versions of GC12.

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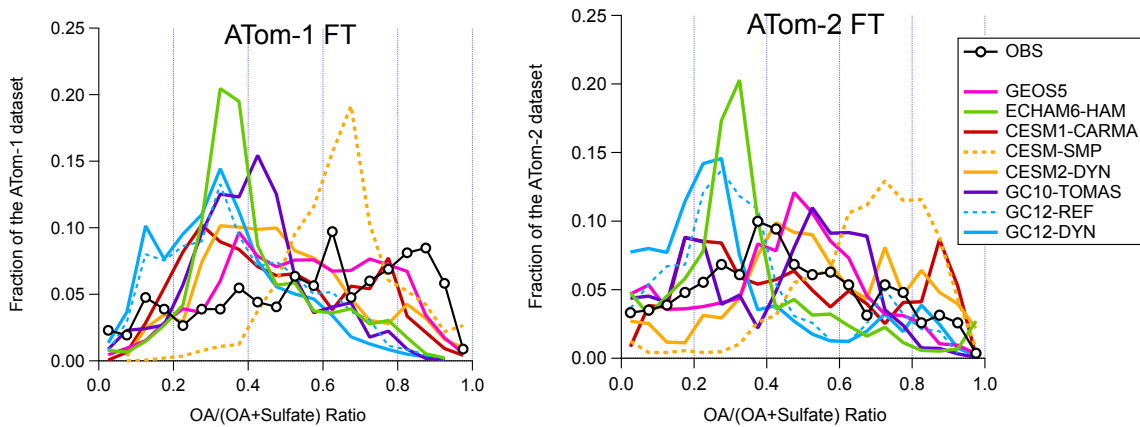
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1698 (a)



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1700 (b)



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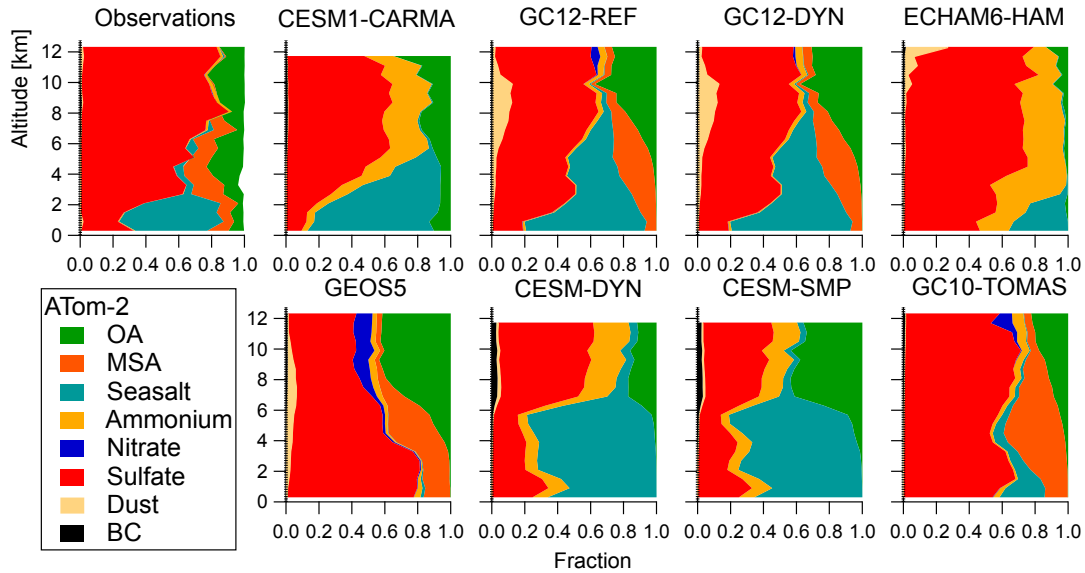
1702 Figure 9: (a) Predicted and measured composition of submicron aerosols in the free  
1703 troposphere as a function of the submicron aerosol mass concentrations during ATom-1.

1704 (b) Frequency distribution of observed and simulated ratio of organic to organic plus  
1705 sulfate aerosols in the free troposphere during ATom-1 and -2.

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1710 Figure 10: Comparison of measured and predicted composition of submicron aerosols as  
1711 a function of altitude over the remote Southern Ocean region during NH Winter (ATom-2).  
1712 For models that do not calculate ammonium in the aerosol (such as CESM1-CARMA,  
1713 CESM2-SMP, CESM2-DYN and ECHAM6-HAM), ammonium was estimated from the  
1714 sulfate mass assuming the formation of ammonium sulfate. Note that while the modeled  
1715 and measured submicron sea salt size ranges agree fairly well (Table 1), this is not quite  
1716 the case for dust. Given that the accumulation mode dust in the models presented  
1717 contains larger sizes than the AMS range ( $< 500$  nm), it is expected for the modeled dust  
1718 concentration to be larger than measured.

1719