

Factors controlling marine aerosol size distributions and their climate effects over the Northwest Atlantic Ocean region

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Abstract.

Aerosols over Earth's remote and spatially extensive ocean surfaces have important influences on planetary climate. However, these aerosols and their effects remain poorly understood, in part due to the remoteness and limited observations over these regions. In this study, we seek to understand factors that shape marine aerosol size distributions and composition in the Northwest Atlantic Ocean region. We use the GEOS-Chem-TOMAS model to interpret measurements collected from ship and aircraft during the four seasonal campaigns of the North Atlantic Aerosols and Marine Ecosystems Study (NAAMES) conducted between 2015 and 2018. Observations from the NAAMES campaigns show enhancements in the campaign-median number of aerosols with

32 diameters larger than 3 nm in the lower troposphere (below 6 km), most pronounced during the
33 phytoplankton bloom maxima (May/June) below 2 km in the free troposphere. Our simulations,
34 combined with NAAMES ship and aircraft measurements, suggest several key factors that
35 contribute to aerosol number and size in the Northwest Atlantic lower troposphere, with significant
36 regional-mean (40-60 °N, 20-50 °W) aerosol-cloud albedo indirect effects (AIE) and direct
37 radiative effects (DRE) during the phytoplankton bloom. These key factors and their associated
38 simulated radiative effects in the region include: (1) particle formation near and above the marine
39 boundary layer (MBL) top (AIE: -3.37 W m^{-2} , DRE: -0.62 W m^{-2}), (2) particle growth due to
40 marine secondary organic aerosol (MSOA) as the nascent particles subside into the MBL, enabling
41 them to become cloud-condensation-nuclei-size particles (AIE: -2.27 W m^{-2} , DRE: -0.10 W m^{-2}),
42 (3) particle formation/growth due to the products of dimethyl sulfide, above/within the MBL ($-$
43 1.29 W m^{-2} , DRE: -0.06 W m^{-2}), (4) ship emissions (AIE: -0.62 W m^{-2} , DRE: -0.05 W m^{-2}) and
44 (5) primary sea spray emissions (AIE: $+0.04 \text{ W m}^{-2}$, DRE: -0.79 W m^{-2}). Our results suggest that
45 a synergy of particle formation in the lower troposphere (particularly near and above the MBL top)
46 and growth by MSOA contributes strongly to cloud-condensation-nuclei-sized particles with
47 significant regional radiative effects in the Northwest Atlantic. To gain confidence in radiative
48 effect magnitudes, future work is needed to understand 1) the sources and temperature-dependence
49 of condensable marine vapors forming MSOA, 2) primary sea spray emissions, and 3) the species
50 that can form new particles in the lower troposphere and grow these particles as they descend into
51 the marine boundary layer.

52

53 **1. Introduction**

54

55 Marine atmospheric particles have important roles in Earth's climate system. Similar to particles
56 in other regions, marine aerosols scatter and absorb solar radiation (Charlson et al., 1992), and
57 modify cloud properties by acting as the seeds for cloud droplet formation (Boucher and Haywood,
58 2000; Lohmann and Feichter, 2005). Aerosols in the atmosphere's marine boundary layer (MBL)
59 strongly influence the highly prevalent, low-altitude marine clouds, which have key climate
60 cooling effects due to their reflection of incoming solar radiation (Wood, 2012; Chen et al., 2014).
61 However, there remains high uncertainty about the magnitude of these aerosol effects (IPCC,
62 2013), due in part to limited understanding about the processes that control aerosols over Earth's
63 expansive and remote ocean surfaces (Willis et al., 2018). Marine aerosols are strongly influenced

64 by natural, but poorly understood sources, making a large contribution to uncertainty in aerosol-
65 climate effects (Carslaw et al., 2010; Carslaw et al., 2013). Limited observations of aerosols and
66 their precursors over Earth's remote marine regions contribute to these knowledge gaps. In this
67 study, we focus on investigation of several factors controlling the seasonal cycle of aerosol size
68 and number and their resultant climate effects over the Northwest Atlantic Ocean.

69
70 Aerosol particles in the remote MBL have several seasonally varying sources (O'Dowd et al.,
71 2004; Leck and Bigg, 2005; de Leeuw et al., 2011; Karl et al., 2012). Primary particles are emitted
72 through wave breaking and bubble bursting processes that eject sea spray aerosols (SSA) of sea
73 salt and organic composition (Russell et al., 2010; de Leeuw et al., 2011; Ovadnevaite et al., 2011;
74 Gantt and Meskhidze, 2013; Prather et al., 2013; Hamacher-Barth et al., 2016; Brooks and
75 Thornton, 2018). SSA have a not-yet-well-understood dependence on wind speed (Monahan et al.,
76 1983; O'Dowd et al., 1997; Ovadnevaite et al., 2012; Grassian et al., 2015; Brooks and Thornton,
77 2018; Saliba et al., 2019) and sea surface temperature (Mårtensson et al., 2003; Jaeglé et al., 2011;
78 Kasparian et al., 2017; Saliba et al., 2019). For the North Atlantic, observations indicate that
79 primary SSA make a limited (less than 30%) contribution to cloud condensation nuclei (CCN)
80 (Quinn et al., 2017, Zheng et al., 2018; Quinn et al., 2019) with no direct connection between SSA
81 emissions and plankton ecosystems because the organic SSA appears to arise from the ocean's
82 large pool of dissolved organic carbon (Quinn et al., 2014; Bates et al., 2020). SSA, however,
83 could modify the CCN number that activate to form cloud droplets (Fossum et al., 2020), act as
84 ice nuclei (Wilson et al., 2015; DeMott et al., 2016; Irish et al., 2017), and be more closely linked
85 with biogenic activity in other regions (Ault et al., 2013; Cravigan et al., 2015; O'Dowd et al.,
86 2015; Quinn et al., 2015; Wang et al., 2015; Schiffer et al., 2018; Christiansen et al., 2019;
87 Cravigan et al., 2019). Recent studies have highlighted knowledge gaps related to sea spray
88 emissions, particularly as related to the submicron sizes (e.g., Bian et al., 2019; Regayre et al.,
89 2020). Measurement and modeling studies are needed to better understand and simulate the size-
90 resolved contribution of sea spray to the Northwest Atlantic MBL.

91
92 For the North Atlantic, secondary aerosol of biogenic origin is observed to be an important
93 seasonally varying contributor to marine particles and their growth to yield CCN (Sanchez et al.,
94 2018). Marine secondary aerosol can arise from the condensation of a variety of marine-vapor-

95 oxidation products, which form and grow particles (Ceburnis et al., 2008; Rinaldi et al., 2010;
96 Decesari et al., 2011). Formation of new aerosol particles in the marine environment is observed
97 to be favored in clean atmospheric layers just below the marine inversion and also above the MBL
98 top (Kazil et al., 2011; Takegawa et al., 2020). Newly formed particles, including those from the
99 free troposphere can grow to CCN sizes (diameters larger than about 50 nm) through the
100 condensation of available organic and sulfur-containing vapors on descent into the MBL
101 (Korhonen et al., 2008). Once the particles reach CCN sizes, cloud processing (including aqueous
102 phase aerosol production, and cloud droplet coagulation with other droplets and interstitial
103 aerosols) also contributes to shaping the size distribution (Hoppel et al., 1986; Hoose et al., 2008;
104 Pierce et al., 2015). For the North Atlantic MBL, entrainment of growing new particles formed in
105 the relatively cleaner free troposphere is an important contributor to MBL particle number (Quinn
106 et al., 2017; Sanchez et al., 2018; Zheng et al., 2018). In the pristine conditions of the summertime
107 Arctic, both new particle formation (NPF) and growth (by condensation of organic and sulfur-
108 containing vapors) are frequently observed within the boundary layer itself (Leaitch et al., 2013;
109 Croft et al., 2016a; Willis et al., 2016; Collins et al., 2017; Burkart et al., 2017b). In addition to
110 sulfuric acid, other vapors including amines, methane sulfonic acid (MSA), ammonia, and iodine
111 all contribute to NPF in marine regions (O'Dowd, 2002; Facchini et al., 2008; Allan et al., 2015,
112 Chen et al., 2016; Croft et al., 2016a; Dall'Osto et al., 2018). Interpretation of a combination of
113 aircraft and ship-board observations with a size-resolved aerosol microphysics model is needed to
114 develop understanding of the relative importance of near and above MBL top NPF as a contributor
115 to aerosol size distributions in the Northwest Atlantic MBL.

116

117 Dimethyl sulfide (DMS) is one of the key contributors to secondary particle formation and growth
118 that is released from the oceans as a result of marine biogenic activity (Lana et al., 2012a; Galí and
119 Simó, 2015; Sanchez et al., 2018). The oxidation products of DMS include sulfuric acid and MSA
120 (Barnes et al., 2006), which can form new particles and grow existing particles to sizes that can
121 act as CCN (Hoffman et al., 2016; Hodshire et al., 2019). As well, hydroperoxymethyl thioformate
122 (HPMTF) is a recently discovered DMS-oxidation product, which could also contribute to NPF
123 and growth (Veres et al., 2020). The role of DMS in the climate system has undergone much debate
124 since 1987 when the CLAW hypothesis proposed that DMS could act as a regulator in a warming

125 climate (Charlson et al., 1987). For the North Atlantic and Arctic, observations have linked DMS
126 to the formation of aerosols during the times of phytoplankton blooms (Rempillo et al., 2011;
127 Chang et al., 2011; Park et al., 2017; Sanchez et al., 2018; Abbatt et al., 2019; Quinn et al., 2019).
128 As well, modelling studies have supported a role for DMS, linked to phytoplankton blooms, as a
129 contributor to CCN number concentrations in the North Atlantic and Arctic MBLs (Woodhouse et
130 al., 2013; Zheng et al., 2018; Ghahremaninezhad et al., 2019; Mahmood et al., 2019) and Southern
131 Ocean MBL (Korhonen et al., 2008; McCoy et al., 2015; Revell et al., 2019). However, the extent
132 to which DMS can act as a climate regulator remains unclear (Schwinger et al., 2017; Fiddes et
133 al., 2018), and this role has been refuted (Quinn and Bates, 2011). Analysis of in situ observations
134 of DMS and its products across the seasonal cycle of marine biogenic activity and in various ocean
135 regions is needed to improve understanding related to the role of DMS in Earth's climate system.

136
137 Marine secondary organic aerosol (SOA) is another important contributor to sub-micron diameter
138 marine aerosols, but is not well characterized (Rinaldi et al., 2010). The oceans are a source of a
139 variety of organic vapors that could lead to SOA formation (O'Dowd and de Leeuw, 2007; Yassaa
140 et al., 2008; Carpenter et al., 2012; Lana et al. 2012b; Hu et al., 2013; Carpenter and Nightingale,
141 2015; Kim et al., 2017; Rodríguez-Ros et al., 2020a). Oxygenated volatile organic compounds
142 (OVOCs) recently linked to photochemical oxidative processes at the sea surface microlayer are
143 possible contributors to marine SOA (Mungall et al., 2017). Isoprene and monoterpenes appear to
144 make relatively minor contributions to marine SOA by mass, less than 1% for particles with
145 diameters smaller than 10 μm at Cape Grim (Cui et al., 2019). The global, annual source of organic
146 vapors from the oceans is highly uncertain, but current estimates are about 23 to 92 Tg C yr⁻¹
147 (Brüggemann et al., 2018). Laboratory studies indicate that emissions of marine organic vapors
148 increase with both temperature and incident radiation for temperatures up to about 26 °C
149 (Meskhidze et al., 2015). Recent observations and modeling studies support a role for Arctic
150 marine secondary organic aerosol (AMSOA) as a contributor to particle growth to CCN sizes
151 (Burkart et al., 2017a; Collins et al., 2017; Willis et al., 2017; Willis et al., 2018; Tremblay et al.,
152 2018; Leaitch et al., 2018; Croft et al., 2019; Abbatt et al., 2019). For the North Atlantic, organics
153 are also found to make a large contribution to particle growth to CCN sizes (Sanchez et al., 2018;
154 Zheng et al., 2020a). The result of the above-noted processes is a large and complex pool of organic

155 aerosol in the marine environment with sources that vary seasonally and regionally (Cavalli et al.,
156 2004; Decesari et al., 2011; Cravigan et al., 2015; Liu et al., 2018; Leaitch et al., 2018).

157
158 Anthropogenic activity is also an important source of aerosols over the portions of the Earth's
159 oceans. For the North Atlantic, several previous studies (e.g., Savoie et al., 2002; Stohl et al., 2003;
160 Huntrieser et al., 2005; Fast et al., 2016) found a key role for synoptic scale motions in lifting
161 aerosols arising from North American continental emissions and transporting them in layers over
162 the North Atlantic with intrusions into the MBL. As well, ship traffic is an important source of
163 both particles and oxidants in the MBL (Corbett et al., 2007; Zanatta et al., 2019; Bilsback et al.,
164 in press). Ship emissions of nitrogen oxides have a significant control on levels of oxidants such
165 as ozone, the hydroxyl radical (OH) and NO₃ in the MBL (Vinken et al., 2011; Holmes et al.,
166 2014). In the remote MBL, both OH and NO₃ are key oxidants of DMS, along with natural-source
167 halogens such as BrO, with an important role for multiphase chemistry (Chen et al., 2018).
168 Interpretation of aerosol observations across several seasons is needed to better understand the
169 relative contribution of ship emissions to marine particles in the Northwest Atlantic region.

170
171 In this study, as part of the Ocean Frontier Institute (www.oceanfrontierinstitute.com), we address
172 the knowledge gaps that were identified above, concerning several key factors shaping Northwest
173 Atlantic MBL aerosol size distributions and their seasonal cycle. We consider the role of (1) new
174 particle formation in clean atmospheric layers near and above the MBL top, (2) particle growth by
175 marine SOA (MSOA) on descent into the MBL, (3) DMS contributions, (4) ship traffic emissions
176 and (5) primary sea spray emissions. Aerosol measurements from the North Atlantic Aerosols and
177 Marine Ecosystems Study (NAAMES) (Behrenfeld et al., 2019) provide an excellent basis for
178 addressing the role of these five factors in the Northwest Atlantic Ocean region. The NAAMES
179 aircraft and ship campaigns were conducted during four phases of the Northwest Atlantic annual
180 plankton cycle from 2015-2018. We interpret the NAAMES aerosol measurements using a state-
181 of-the-science size-resolved global aerosol microphysics model, GEOS-Chem-TOMAS
182 (www.geos-chem.org). Our synergistic approach in bringing together NAAMES measurements
183 and size-resolved aerosol process modeling enables a unique consideration of several key factors
184 shaping Northwest Atlantic MBL aerosol size distributions and their annual cycle. We also
185 quantify the impact of these factors on aerosol radiative effects over the North Atlantic.

186

187 The second section provides an overview of our measurement and modeling methodology. The
188 third section presents results using the GEOS-Chem-TOMAS model to interpret NAAMES
189 aerosol measurements and their seasonal cycle with a focus on the roles of near and above MBL
190 top NPF, MSOA, DMS, sea spray, and ship emissions. We also quantify the direct and cloud-
191 albedo indirect aerosol radiative effects attributed to each of these factors during the seasonal
192 cycle. The final section gives our summary and outlook.

193

194 **2. Methodology**

195

196 **2.1 Aerosol measurements during the NAAMES campaigns**

197

198 NAAMES campaigns were conducted during four key periods in the annual cycle of marine
199 biogenic activity, namely: the winter transition (November 2015), the accumulating phase
200 (March/April 2018), the climax transition (May/June 2016), and the declining phase
201 (August/September 2017) (Behrenfeld et al., 2019). These periods are defined by shifts in net
202 phytoplankton growth rates and span a wide range in phytoplankton biomass, here estimated from
203 chlorophyll-*a* concentrations (Chl-*a*). The winter transition is characterized by the annual
204 minimum in Chl-*a* concentrations (generally $< 1 \text{ mg m}^{-3}$) and a shift to favor phytoplankton growth
205 over loss as the increasing ocean mixed-layer depth leads to fewer encounters between
206 phytoplankton and their grazers. The accumulation phase occurs in early springtime when
207 increasing sunlight and decreasing ocean mixed layer depths promote increasing phytoplankton
208 growth rates and concentrations (Chl-*a* between 1 and 2 mg m^{-3}). The climax transition is the time
209 of the annual maximum in phytoplankton biomass (Chl-*a* between 2 and 9 mg m^{-3}) and marks the
210 shift from positive to negative growth rates owing to high grazing rates and depletion of nutrients.
211 The declining phase (Chl-*a* between 1 and 2 mg m^{-3}) occurs later in the summertime when the
212 ocean mixed layer depth increases and incident sunlight decreases, leading to further declines in
213 phytoplankton growth and concentrations. Behrenfeld et al. (2019) provide an overview of the four
214 measurement campaigns, and further details about Chl-*a* during NAAMES. The R/V Atlantis
215 cruise tracks and NASA C130 flight paths are shown in Figure 1. Due to aircraft mechanical
216 problems, there were no flights in 2018 during the accumulating phase.

217

218 In this study, we examine the NAAMES size-resolved aerosol measurements (particle diameters
219 20 to 500 nm) from the Scanning Electrical Mobility Sizer (SEMS, Model 138, 2002, BMI,
220 Hayward, CA) aboard the R/V Atlantis ship. Aerosol particles were isokinetically drawn through
221 an inlet positioned 18 m above sea level (Bates et al. 2002) and were subsequently dried below
222 20% relative humidity using silica diffusion driers prior to sampling by the SEMS. Clean marine
223 periods were identified with criteria of relative wind directions within 90° of the bow, condensation
224 nuclei number concentrations less than 2000 cm⁻³, ammonium and organic aerosol not covarying,
225 ammonium < 100 ng m⁻³ and having back trajectories primarily over the ocean surface. We also
226 consider aerosol size-resolved measurements (particle diameters 10 to 282 nm) from the Scanning
227 Mobility Particle Sizer (SMPS, TSI Inc., Shoreview, MN) aboard the C130 aircraft. As well, we
228 give attention to measurements of total particle number concentration from the Condensation
229 Particle Counters (CPCs) with differing nominal lower detection diameters: 3 nm for the CPC
230 3025 (yielding N3 measurements) and 10 nm for the CPC 3772 (TSI Inc., St. Paul, MN) (yielding
231 N10 measurements) aboard the C130 aircraft. We also consider submicron, non-refractory sulfate
232 (SO₄⁻) and organic mass (OM) concentrations from an Aerodyne High Resolution Time-of-Flight
233 Aerosol Mass Spectrometer (HR-ToF-AMS, DeCarlo et al., 2006) and refractory black carbon
234 from the Single Particle Soot Photometer (SP2, Schwarz et al., 2006) aboard the aircraft. HR-ToF-
235 AMS and SP2 measurements are restricted to accumulation-mode aerosol (60-600 nm and 105-
236 600 nm diameter, respectively). All aircraft observations are made behind a forward-facing,
237 shrouded, solid diffuser inlet that efficiently transmits particles with aerodynamic diameter less
238 than 5.0 μm to cabin-mounted instrumentation (McNaughton et al., 2007). Cloud-contaminated
239 aerosol observations have been removed using a combination of wing-mounted cloud probe and
240 relative humidity measurements. This filtering may possibly obscure some NPF events in
241 proximity to clouds and remove some cloud-processed samples from the vertical profiles. Aerosol
242 number and mass concentrations are reported at standard temperature and pressure. A Proton-
243 Transfer-Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS) (Müller et al, 2014;
244 Schiller, 2018) was used aboard the NASA C-130 to measure volatile organic compounds
245 including DMS and acetonitrile. Both observational and model data for periods where acetonitrile
246 concentrations exceed 200 ppt are filtered out following Singh et al. (2012) to remove significant
247 biomass burning contributions that are not the focus of this study.

248

249 2.2 GEOS-Chem-TOMAS model description

250

251 We use the GEOS-Chem model (v12.1.1) (<http://www.geos-chem.org>) coupled to the Two
252 Moment Aerosol Sectional (TOMAS) microphysics scheme (Adams and Seinfeld, 2002; Lee and
253 Adams, 2012; Kodros and Pierce, 2017), with 15 sections, representing particle sizes from 3 nm
254 to 10 μm . All simulations are at a $4^\circ \times 5^\circ$ resolution with 47 vertical levels extending to 0.01 hPa.
255 The meteorological fields are from the GEOS Forward Processing off-line fields (GEOS-FP;
256 https://gmao.gsfc.nasa.gov/GMAO_products/). Our size-resolved aerosol simulations
257 parameterize the processes of particle nucleation, coagulation, condensation, along with wet and
258 dry deposition and include the in-cloud aerosol coagulation scheme of Pierce et al. (2015). Sulfate,
259 organic and black carbon, sea salt, dust and aerosol water are simulated. TOMAS is coupled to the
260 full tropospheric aerosol/chemistry scheme of GEOS-Chem. Wet deposition follows Liu et al.
261 (2001), Wang et al. (2011) and Wang et al. (2014). To represent efficient wet removal by North
262 Atlantic drizzle in October and November, we implement a fixed in-cloud removal efficiency of
263 0.001 s^{-1} in the lowest 2 km of the model atmosphere over the ice-free ocean and enable wet
264 removal of sulfate and organic aerosol in clouds with temperatures between 237 K and 258 K. In
265 all seasons, we use the GEOS-FP cloud fraction as the precipitation fraction in the model layers
266 where precipitation occurs for a closer connection with the meteorological fields (Croft et al.,
267 2016b; Luo et al., 2019; Luo et al., 2020). Dry deposition uses the resistance in series approach of
268 Wesley (1989). Simulated gas-phase species are also removed by dry and wet deposition as
269 described in Amos et al. (2012).

270 For emissions, we use the GEOS-Chem v 12.1.1 default setup for gas-phase and primary aerosol
271 emissions. We use emissions from the Community Emissions Data System (CEDS) for global
272 anthropogenic sources of NO_x , CO, SO_2 , NH_3 , non-methane VOCs, black carbon, and organic
273 carbon, including from international shipping as a source of both primary and secondary particles.
274 Primary particles are emitted with a lognormal distribution (Lee et al., 2013). The most recent
275 CEDS emissions dataset extends to the year 2017, as described in McDuffie et al. (2020). In this
276 work, monthly CEDS emission totals for each compound are spatially gridded by source sector,
277 according to the $0.1^\circ \times 0.1^\circ$ gridded EDGAR v4.2 emissions inventory (EC-JRC/PBL, 2012) and
278 population, as described in Hoesly et al. (2018). To account for in-plume chemical processing of

279 ship emissions, we use the PARANOX scheme of Holmes et al. (2014). CEDS emissions are
280 overwritten over the United States by the National Emissions Inventory (NEI11) with updated
281 scale factors for our simulation years (2015-2018). We calculated these factors based on emission
282 data for these years from the United States Environmental Protection Agency. Over Canada, we
283 use the Air Pollutant Emissions Inventory (APEI). The Global Fire Emissions Database (GFED4s)
284 is used for biomass burning emissions (van der Werf et al., 2017) for the years 2015-2016, with
285 GFED4s climatological values for 2017 and 2018 since exact-year emissions were not available
286 when we conducted our simulations. Dust emissions are from the scheme of Zender et al. (2003).
287 Sea salt emissions follow Jaeglé et al. (2011). This temperature-dependent parameterization
288 decreases global emissions relative to the Gong (2003) parameterization. A coupled
289 parameterization for primary organic aerosol from sea spray was not available for our aerosol size-
290 resolved GEOS-Chem-TOMAS simulations, such that some sea spray organics could be
291 misrepresented as sea salt, since all sea spray in our simulations is considered sea salt. Such
292 primary organic emissions are expected to have no seasonal cycle when averaged over the
293 NAAMES region (Bates et al., 2020).

294
295 Exchange of DMS between the ocean and atmosphere is parameterized using the default GEOS-
296 Chem parameterization, which follows Johnson (2010), largely based on Nightingale et al. (2000a;
297 2000b). We use the 8-day mean satellite-retrieval seawater DMS dataset of Galí et al. (2019)
298 developed using the methodology of Galí et al. (2018), for available years (2015 and 2016) for the
299 region north of about 40 °N. The Lana et al. (2011) DMS climatology is used elsewhere. Terrestrial
300 biogenic emissions are from MEGAN2.1 as described in Guenther et al. (2012). Following Croft
301 et al. (2019), we add a source of MSOA coupled to the simple SOA scheme described in Pai et al.
302 (2020). Emissions of MSOA-precursor vapors have been found to increase with temperature
303 (Meskhidze et al., 2015; Rodríguez-Ros et al., 2020a; Rodríguez-Ros et al., 2020b). Here, we use
304 a temperature-dependent simulated source of MSOA-precursor emissions (S_{MSOA}), $S_{\text{MSOA}} = 70T$
305 $+ 350 \mu\text{g m}^{-2} \text{d}^{-1}$, where T is atmospheric temperature (°C) at 2 m altitude. The values of 70 and
306 350 are found to yield acceptable model-measurement agreement for NAAMES campaign-median
307 ship-track and aircraft measurements (Supplementary Figs. S1-S4 and Supplementary Tables S1
308 and S2). This simulated source of condensable vapors is emitted with a 50/50 split between vapors
309 that are immediately available to form MSOA and vapors with 1-day aging prior to availability

310 (and not susceptible to wet removal). MSOA contributes to particle growth in our simulations (in
311 agreement with observational-based studies e.g., Sanchez et al., 2018; Zheng et al., 2020a), along
312 with sulfuric acid, but since the particle nucleating abilities of MSOA are unclear, it does not
313 contribute to new-particle formation.

314

315 All simulations include particle nucleation in the boundary layer that is parameterized with the
316 ternary ($\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$) scheme of Napari et al. (2002), which was scaled by 10^{-5} to better match
317 continental boundary-layer measurements (Westervelt et al., 2013). The binary ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}$)
318 scheme of Vehkamäki et al. (2002) is employed in the free troposphere at low NH_3 concentrations.
319 Growth and loss of particles smaller than 3 nm are approximated following Kerminen et al. (2004).
320 In our simulations, as a surrogate for unparameterized processes in the lower free troposphere and
321 near the MBL top, we also employ an activation-type nucleation parameterization from the MBL
322 top to about 2 km altitude. This activation-type scheme parameterizes nucleation rates as a linear
323 function of sulfuric acid concentrations, using an empirical factor ($A = 2 \times 10^{-6} \text{ s}^{-1}$) (Kulmala et
324 al., 2006; Sihto et al., 2006), and serves as a proxy representing several unknown/unparameterized
325 mechanisms related to NPF. Pockets of very clean air with low condensation sink near MBL
326 clouds, which favor new particle formation (Kazil et al., 2011), are not resolved by large-scale
327 models such as ours, with grid boxes on the scale of 100s km^2 . Efficient wet removal by drizzling
328 MBL clouds contributes to these pristine conditions (Wood et al., 2017). As well, MBL clouds
329 reflect ultraviolet (UV) radiation and create pockets of enhanced UV, which favors photochemical
330 production of aerosol precursor vapors (Weber et al., 2001; Wehner et al., 2015), that are not
331 resolved by our model. Additionally, the particle nucleating capacity of MSOA is unclear and
332 particle formation parameterizations are not yet developed to represent NPF when several gas-
333 phase precursors interact. These precursors include, but are not limited to, MSA (Chen et al.,
334 2016), HPMTF (Veres et al., 2020), amines (Facchini et al., 2008), iodine (Allan et al., 2015), and
335 other extremely low-volatility organic compounds (ELVOCs) (Riccobono et al., 2014). The extra
336 nucleation in the lower troposphere with the activation-type parameterization represents particle
337 precursors that could have the same source as sulfuric acid. This approach may not capture the
338 timing and magnitude of the variability in NPF correctly because the vapors participating in this
339 nucleation are likely not just sulfuric acid. Future work is needed to better understand the nature
340 of the nucleating species in the lower troposphere over the oceans.

341
342 We also conduct off-line radiative transfer calculations using the Rapid Radiative Transfer Model
343 for Global Climate Models (RRTMG) (Iacono et al., 2008) to assess the direct radiative effect
344 (DRE) and cloud-albedo aerosol indirect effect (AIE). The aerosol optical properties are calculated
345 using the Mie code of Bohren and Hoffman (1983) to find the extinction efficiency, single
346 scattering albedo, and asymmetry factor. Then, these optical properties, along with the monthly
347 mean cloud fraction and surface albedo from the GEOS-FP meteorology fields, are input to the
348 RRTMG to determine the change in top-of-the-atmosphere solar flux (DRE) between two
349 simulations (our control simulation and one of the sensitivity simulations, Sect. 2.3). Our DRE
350 calculations follow Kodros et al. (2016), with updates to include ammonium nitrate as described
351 in Bilsback et al. (in press). All particles except black carbon are treated as internally mixed within
352 each size section. We also calculate the cloud-albedo aerosol indirect effect (AIE) as described in
353 Kodros et al. (2016), Croft et al. (2016a) and Ramnarine et al. (2019). The Abdul-Razzak and
354 Ghan (2002) parameterization is used to calculate offline cloud droplet number concentrations
355 (CDNC) using the aerosol mass and number concentrations from our simulations. We assume an
356 updraft velocity of 0.5 m s^{-1} and the hygroscopicity parameters used by Kodros et al. (2016) and
357 Kodros and Pierce (2017), assuming aerosol internal mixture, including ammonium nitrate
358 following Bilsback et al. (in press). For each model grid box, we assume cloud droplet radii of 10
359 μm and perturb this value with the ratio of the monthly mean CDNC between two simulations (our
360 control simulation and one of the sensitivity simulations, Sect. 2.3), assuming constant cloud liquid
361 water content. The RRTMG is used to calculate the change in the top-of-the-atmosphere solar flux
362 (AIE) due to changes in cloud droplet radii.

363
364 As one evaluation of simulation performance, we calculate the mean fractional error (MFE) of the
365 0th to 3rd moments between the simulated and observed MBL aerosol size distributions, following
366 Boylan and Russell (2006) and using the same methodology as Hodshire et al. (2019) and Croft et
367 al., (2019). The MFE is defined as a mean over the N aerosol size distribution moments,

$$369 \text{ MFE} = \frac{1}{N} \sum_{i=0}^{N-1} \frac{\text{abs}|C_m(i) - C_o(i)|}{(C_m(i) + C_o(i))/2} \quad (1)$$

370

371 where $C_m(i)$ is the integrated value of the i^{th} moment of the simulated aerosol size distribution and
372 $C_o(i)$ is the integrated value of the i^{th} moment of the observed aerosol size distribution. The MFE
373 can range from 0 to +2. We adopt the convention of Boylan and Russell (2006) to consider a MFE
374 of 0.5 or less as acceptable.

375
376 For consideration of vertical profiles, we binned the measurement and simulation values using a
377 500 m height resolution, starting from the surface to 500 m as the first bin. Campaign-median
378 values are calculated within each bin and plotted at the mid-point of the bin, starting at 250 m.
379 During NAAMES, the lowest aircraft flight level altitude was around 150-200 m GPS altitude. We
380 use a plane-flight diagnostic in the model to sample the simulation interpolated between grid-cell
381 centers to the aircraft-flight-track position, during the times when measurement data was available
382 for each respective instrument. We find consistent results with bin resolutions of 250, 500 and
383 1000 m, giving support for our selected binning resolution. The vertical profiles show
384 measurements and model output along the aircraft flight tracks only and do not include any
385 measurements or model output for the ship track. Vertical profile MFEs (Eq. 1) are calculated by
386 summation over the altitude bins.

387

388 **2.3 Summary of GEOS-Chem-TOMAS simulations**

389

390 Table 1 summarizes the simulations conducted. Simulation BASE is our control simulation and
391 includes all emissions and process parameterizations described above. We conduct five sensitivity
392 simulations to examine the role of several key factors involved in shaping the aerosol distributions
393 within the NAAMES study region. Simulation noABLNUC is the same as BASE, except without
394 the sulfuric acid-dependent activation-type surrogate nucleation parameterization, which we
395 implemented from the MBL top to about 2 km. Simulation noMSOA is the same as BASE, but
396 without the source of temperature-dependent condensable marine organic vapors, forming MSOA.
397 Simulation noDMS is the same as BASE, but without DMS. Simulation noSHIPS is the same as
398 BASE, but without any ship emissions. Finally, simulation noSS is the same as BASE, but without
399 any primary sea spray emissions. All simulations are sampled coincidentally with the
400 measurements using hourly output along the NAAMES aircraft and ship tracks within the
401 respective model grid boxes, using the NAAMES campaigns' 1-minute-resolution navigation data.

402 To manage computational expense, the simulations are necessarily at a coarse resolution, which
403 can bias model-measurement comparisons. However, these biases will be lower for remote marine
404 regions such as the NAAMES study region than over land regions, which generally have greater
405 spatial inhomogeneity. Representativeness errors were also reduced by limiting our model-
406 measurement comparisons to campaign-median values.

407

408 **3. Results and Discussion**

409

410 **3.1 Key features of aerosols observed during NAAMES**

411

412 Aerosol observations made during the NAAMES campaigns were in four seasons, capturing
413 different stages of the annual cycle of Northwest Atlantic marine biogenic activity (Behrenfeld et
414 al., 2019). Figure 2 shows the campaign-median marine-influenced aerosol size distributions from
415 SEMS (particle diameters 20-500 nm) for the four R/V Atlantis cruises. November 2015 (winter
416 transition, bloom minima) is characterized by the lowest aerosol number concentrations. The peak
417 of the Northwest Atlantic drizzle season occurs at this time, with efficient wet removal of
418 accumulation-sized aerosol (diameters larger than about 50 to 100 nm) (Browse et al., 2012). As
419 well, relative to other the seasons, marine biogenic emissions are low at this time of minimal
420 phytoplankton biomass. The summertime observations during both May/June 2016 (climax
421 transition, phytoplankton bloom maxima) and August/September 2017 (declining phase) are
422 characterized by a weakly dominant Aitken mode (particle diameters < 100 nm). The winter
423 transition (November 2015) and early spring accumulation phase observations (March/April 2018)
424 are characterized by the dominance of accumulation-mode aerosols (particle diameters > 100 nm).

425

426 The vertical profiles of campaign-median integrated-SMPS (particle diameters of 10 to 282 nm)
427 observations are shown in Fig. 3. There are several key features of the observed aerosol vertical
428 profiles for the three NAAMES flight campaigns. These profiles exhibit several particle number
429 maxima in the lower free troposphere below 6 km, including below 2 km during the May/June
430 climax transition period. As shown in Fig. 3, aerosol surface area and volume are less at altitudes
431 below about 3 km relative to altitudes above 3 km. This lower particle surface area at these altitudes
432 favors NPF over growth of pre-existing particles as available vapors condense in these relatively

433 cleaner atmospheric layers (Kazil et al., 2011). Transport of aerosols (in part associated with
434 continental emissions) contributes to particles in all seasons. Fast et al. (2016) characterized
435 summertime North Atlantic transport layers in the free troposphere associated with synoptic-scale
436 lifting. The late fall (November 2015, Fig.3) is characterized by the lowest aerosol number, surface
437 and volume concentrations, similar to the findings shown in Fig. 2.

438
439 Figure 4 shows the vertical-profile campaign-median total particle number concentrations from
440 CPCs, for aerosols with diameters larger than 3 nm (N3), larger than 10 nm (N10), and the
441 difference between the two (N3-N10). For the May/June 2016 climax transition (phytoplankton
442 bloom maximum), there are enhancements in observed number concentration (N3, N10 and N3-
443 N10) below about 2 km in the free troposphere, indicating NPF at these altitudes (Fig. 4). The
444 MBL top ranged from about 0.5 to 2 km for the NAAMES cruises (Behrenfeld et al., 2019). The
445 lower free tropospheric region near and above the MBL top is an important region for marine
446 NPF. These altitudes above the MBL clouds are generally very clean, which favors NPF, and
447 strongly sunlit, which favors the photochemical oxidative production of particle precursors for
448 NPF. Previous studies based on observations from other marine regions have also found a cloud-
449 processed ultra-clean layer with weak condensation/coagulation sinks at about 1 km altitude,
450 where NPF is favored (Kazil et al., 2011; Takegawa et al., 2020). Figure 4 also shows
451 enhancements in the observed N3 and N10 concentrations below 6 km during the declining
452 phase and winter transition (bloom minima). However, the total number concentration
453 enhancements below 2 km are most pronounced during the phytoplankton bloom maximum,
454 suggesting a connection between particle number and the level of marine biogenic activity.

455
456 SO_4^- and OM are dominant non-refractory components of the submicron-diameter aerosols, and
457 vertical profiles of campaign-median observations are shown on Fig. 5. During the summertime
458 (May/June 2016, climax transition and August/September 2017, declining phase), the OM
459 contribution exceeds that of SO_4^- at most altitudes up to 6 km. Non-refractory SO_4^- has its peak
460 contribution during the climax transition season. This May/June phytoplankton bloom maxima
461 period is the time of peak observed near-surface atmospheric DMS mixing ratios, as shown in Fig.
462 6. During the climax transition (bloom maxima), non-refractory SO_4^- concentrations increase
463 towards the surface, suggesting a marine surface source, similar to summertime Arctic marine

464 profile observations (Willis et al., 2017). Black carbon (BC) concentrations are also shown in Fig.
465 5 and have several peaks in the free troposphere in all seasons, consistent with a long-range
466 transport source. Maximum BC concentrations are in May/June, likely associated with greater
467 transport of anthropogenic continental pollution and biomass burning during this time, relative to
468 other seasons. Springtime has also been associated with peak BC concentrations in the Arctic due
469 to long-range transport (Sharma et al., 2004; Sharma et al., 2006; Fisher et al., 2010; Wang et al.,
470 2011; Xu et al., 2017). All aerosol mass concentrations in the lowest 2 km of the atmosphere (Fig.
471 5) are lowest in the November 2015 winter transition, which is a time of efficient wet removal by
472 drizzle (Browse et al., 2012; Wood et al., 2017), diminishing marine emissions due to diminishing
473 phytoplankton biomass, and outbreaks of relatively less polluted polar air advected down the
474 Labrador Strait (Behrenfeld et al., 2019). For the Arctic, the fall season has also been associated
475 with a relative minimum in aerosol number concentrations (Tunved et al., 2013; Croft et al.,
476 2016b).

477
478 The GEOS-Chem-TOMAS model (described in Sect. 2.2 and 2.3) is generally able to simulate the
479 above-noted features of the aerosols over the Northwest Atlantic. Simulation BASE captures key
480 aspects of the MBL size distributions including the minimum in aerosol number during the
481 November winter transition, the weakly dominant Aitken mode during the May/June climax
482 transition and August/September declining phase and the maximum in number of accumulation-
483 mode particles (diameters greater than 100 nm) during the March/April accumulation phase,
484 despite errors such as between 20-50 nm (Fig. 2). As well, the BASE simulation captures several
485 lower tropospheric enhancements in particle number concentration, although the simulated altitude
486 for the maximum is sometimes displaced and there are errors in the magnitude (Figs. 3 and 4). In
487 the lowest 2 km of the atmosphere, SO_4^- , OM, and BC mass concentrations for simulation BASE
488 are generally within the 25th to 75th measurement percentiles, except for BC and OM
489 underpredictions in May/June 2016, and OM overprediction in November 2015. All simulated
490 SO_4^- presented in this study is non-sea-salt SO_4^- . Simulation BASE also captures that the near-
491 surface SO_4^- is greatest during the May/June climax transition and the near-surface OM has its
492 maximum value during the August/September declining phase. For each season the mean MFE
493 across the parameters considered in Figs. 2 to 5 (BASE versus measurements, Supplementary
494 Table S2) is satisfactory (MFE ranges 0.43 to 0.50). In the next four sub-sections, we use the

495 GEOS-Chem-TOMAS BASE simulation, relative to a set of sensitivity simulations, to examine
496 the potential of five key factors to shape aerosol size distributions in the Northwest Atlantic during
497 four stages of the annual cycle of marine biogenic activity.

498

499 **3.2 Role of new particle formation (NPF) in the lower troposphere**

500

501 Our simulations (BASE relative to noABLNUC, Fig. 4) suggest that NPF near and above the MBL
502 has a strong control on the development of the total particle number (N₃) maxima, with peak
503 magnitude during the phytoplankton bloom maxima in layers below 2 km. Without the surrogate
504 NPF scheme employed near and above the MBL top, the ternary NPF scheme in the MBL in
505 simulation noABLNUC fails to simulate sufficient particle number, although vertical-profile
506 campaign-median ammonium concentrations below 4 km altitude had acceptable agreement with
507 observations (MFE ranges from 0.12 to 0.48, not shown). Figure 4 shows about a one-order-of-
508 magnitude underprediction of N₃ below about 2 km for noABLNUC. NoABLNUC has an
509 unacceptable seasonal-mean model-measurement agreement across the measurement set (MFE
510 ranges from 0.66 to 0.78, Supplementary Table S2). Figure 3 also shows that NPF near and above
511 the MBL top makes a significant contribution to simulated particle number concentrations for
512 aerosol diameters of 10 to 282 nm in the lower troposphere, most strongly in the summertime
513 (BASE relative to noABLNUC). There is little impact on aerosol mass concentrations for
514 simulation noABLNUC relative to BASE (Fig. 5).

515

516 The simulated N₃-N₁₀ (Fig. 4) illustrates that representation of NPF is a challenge for models,
517 because there are difficulties capturing the magnitude and altitudes of the N₃-N₁₀ maxima. These
518 discrepancies reflect key knowledge gaps related to the species that can form new particles in the
519 marine environment (e.g., Veres et al. 2020). As well, the coefficient that we used for the surrogate
520 activation-style nucleation parameterization was derived for a continental environment. The
521 empirical ('A') value used by the parameterization appears to yield excessive NPF for the
522 NAAMES marine environment. Activation-style nucleation was added in our simulations as a
523 proxy for missing nucleation when the condensation sink is low, and conditions favor high
524 oxidation rates. We acknowledge that this approach will miss variability in the timing and rates
525 because it is a surrogate and not exactly the correct mechanism. As well in the summertime, the

526 simulations underpredict N3-N10 concentrations above 2 km, suggesting the need for future work
527 to better understand the NPF processes at these levels, where the binary scheme of Vehkamaki et
528 al. (2002) does not generate sufficient NPF.

529
530 NPF also makes a very strong contribution to the simulated aerosol size distributions within the
531 MBL near the ocean surface (BASE versus noABLNUC, Fig. 2). Although our simulations do
532 include NPF within the MBL, simulated NPF occurs more strongly near and above the MBL top
533 and the resultant particles grow by condensation of available vapors and cloud processing while
534 descending into the MBL. This role for NPF is in agreement with previous studies including those
535 of Clarke et al. (2013), Quinn et al. (2017), and Williamson et al. (2019). As a result, NPF from
536 several altitudes above the ocean surface contributes to the near-ocean-surface particles, with
537 diameters from 20 to 200 nm. NPF does occur in the MBL. However, those levels above the MBL
538 clouds favor oxidative chemistry that yields particle precursors, particularly from the wide-spread
539 and persistent DMS sources in the marine environment (Kazil et al., 2011). Table 2 shows that for
540 all seasons, the surrogate nucleation (simulation BASE, MFEs ranging from 0.04 to 0.33)
541 represents an improvement over simulation noABLNUC (without this surrogate NPF
542 parameterization, MFEs ranging from 0.50 to 0.95).

543
544 Extending the surrogate activation-style parameterization to the surface (Supplementary Figs. S5-
545 S8 and Supplementary Table 3), leads to overprediction of the number of particles with diameters
546 less than 50 nm in the MBL and yields higher MFEs (ranging from 0.20 to 0.56) than for simulation
547 BASE, although the errors were not as large as those for noABLNUC. For the vertical profiles,
548 this extra NPF extended into the MBL yields overprediction of N3, N10, and N3-N10 below 1 km
549 in all seasons. Aerosol surface area and volume (in the SMPS particle-diameter size range of 10
550 nm - 282 nm) were also over predicted during the August/September declining phase, when the
551 simulated temperature-dependent MSOA source was strongest, growing these extra new particles
552 to larger sizes. These challenges highlight the relevance of ongoing research to better understand
553 NPF in the marine environment.

554

555 **3.3 Role of particle growth by condensing marine organic vapors**

556

557 Condensing marine organic vapors forming MSOA are needed in our simulations (in addition to
558 H₂SO₄) for sufficient particle growth to yield satisfactory model-measurement agreement for MBL
559 size distributions (BASE versus noMSOA, Fig. 2). For simulation noMSOA, the model
560 overpredicts the number of particles with diameters smaller than about 30 nm in the MBL. Due to
561 insufficient particle growth of these sub-30 nm particles, the number of particles with diameters
562 between about 30 to 200 nm is underpredicted by more than 50% for simulation noMSOA.

563
564 In our simulations, MSOA enables particle growth to CCN sizes (diameters of about 50 nm or
565 larger). After particles reach CCN sizes, cloud processing can also contribute to simulated particle
566 growth towards accumulation-mode particles (diameters of 100-1000 nm) due to aqueous-phase
567 aerosol production. Other cloud processes include coagulation of cloud droplets with each other
568 and with interstitial aerosols (Hoose et al., 2008; Pierce et al., 2015). Our simulations include the
569 latter and aqueous-phase sulfate production. As clouds evaporate, cloud processing leads to
570 development of the ‘Hoppel minima’ of the MBL aerosol size distributions (Hoppel et al., 1987),
571 which is the minimum aerosol diameter that activates to form a cloud droplet (about 50-70 nm for
572 the observations in Fig. 2). This minimum diameter is smallest in the winter transition (November
573 2015), suggesting that smaller particles activated under the clean condition of this season relative
574 to the other seasons. As shown by Table 2, simulation noMSOA has an unacceptable annual-mean
575 MFE of 0.63, larger than the MFE of 0.23 for simulation BASE, which includes particle growth
576 due to MSOA.

577
578 The nature and flux of marine vapors forming MSOA are not well understood. As a result, we
579 developed a simplistic MSOA parameterization for use in this study, such that the MSOA
580 precursors vapor emissions are an increasing function of temperature. This approach yields a
581 seasonal cycle, and is in agreement with the temperature dependence trend found by previous
582 studies, including Meskhidze et al. (2015), Rodríguez-Ros et al. (2020a) and Rodríguez-Ros et al.,
583 2020b). We find that the simulated NAAMES cruise-track median aerosol size distributions are
584 sensitive to the coefficients used in the parameterization ($S_{\text{MSOA}} = 70T - 350 \mu\text{g m}^{-2} \text{d}^{-1}$)
585 (Supplemental Figs. S1 and Table S1). For example, varying the temperature sensitivity between
586 50-100 and the intercept between 300-500 change the simulated number concentration of particles
587 with diameters larger than 50 nm in the MBL by up to a factor of two, with the greatest sensitivity

588 during the summertime (Supplemental Fig. S1). For the NAAMES MBL size distributions, the
589 annual-mean model-measurement MFEs are acceptable (ranging from 0.23 – 0.38, lowest for
590 BASE) for all temperature-dependent parameterizations that we tested, except for the factor-of-
591 ten scaling up of the BASE MSOA parameterization (simulation 10x(70T-350), Supplementary
592 Table S1, MFE of 0.75) and with the MSOA parameterization removed (simulation noMSOA,
593 Supplementary Table S1, MFE of 0.63). While this source flux is reasonably constrained for our
594 simulations, future work is needed to better understand and parameterize this source.

595
596 The vertical profiles are also sensitive to the MSOA parameterization (Supplementary Figs. S2-
597 4). Between noMSOA and the various MSOA parameterizations that we tested, concentrations
598 vary by up to a factor of about 2 for aerosol number (N3, N10, and N3-N10), SMPS-size-range
599 (diameters 10 nm - 282 nm) number, surface area, volume and also OM. Simulation noMSOA has
600 relatively greater error in the mean across the entire measurement set for each season (MFE ranges
601 from 0.53-0.68) relative to BASE (MFE ranges from 0.42-0.50) (Supplementary Table S2).

602
603 Although the chosen MSOA parameterization reasonably represents the observations, major
604 knowledge gaps remain regarding MSOA precursor species and their chemical lifetimes. While
605 the nature of MSOA precursors is not well-understood, recent measurements suggest that these
606 precursors could include a variety of chemical compounds. For example, measurements from the
607 Arctic indicate that the organics in marine aerosols were not typical biogenic SOA but had a long-
608 hydrocarbon chain implying a fatty acid type precursor (Willis et al., 2017). In other marine
609 regions, isoprene (Ciuraru et al., 2015) and carboxylic acids (Chiu et al., 2017) may also be
610 important. Given the limitations of current knowledge and the indications for a variety of MSOA
611 precursors, the improved MFEs for BASE relative to noMSOA provide support for the employed
612 MSOA parameterization.

613
614 The near-surface campaign-median climax transition and declining phase OM concentrations are
615 within the 25th to 75th measurement percentiles for simulation BASE, and below the 25th percentile
616 of the observations for simulation noMSOA (Fig. 5). On average over the lowest 2 km of the
617 atmosphere during the May/June climax transition and August/September declining phase,
618 simulation BASE relative to noMSOA indicates that MSOA contributes about 200-400 ng m⁻³ to

619 simulated OM. Saliba et al. (2020) suggest that MBL-measurement non-refractory OM during
620 NAAMES clean marine periods provides a good estimate of MSOA. Their seasonal-average non-
621 refractory OM of about 300-400 ng m⁻³ for the 2016 May/June climax transition (phytoplankton
622 bloom maxima) and 2017 August/September declining phase is similar to our model result. This
623 contribution is about 3- to 4-fold greater than the contribution upwards of 100 ng m⁻³ from previous
624 studies, noted in Kim et al. (2017). The model-measurement agreement for OM for 2017 is
625 influenced by significant biomass burning with high altitude emission injections during this time
626 (Zheng et al., 2020b; Saliba et al., 2020). Errors in the simulated emissions due to use of a GFED
627 climatological-year emissions and injection-height errors could account for some of the model-
628 measurement bias at high altitudes. As well, despite our implementation of a filter to remove
629 measurement and model samples with strong in-plume aerosol enhancements during times of high
630 acetonitrile concentrations, some biomass burning influence still affects the presented vertical
631 profiles. Below 500 m altitude, condensing organic vapors yielding MSOA also increase the
632 simulated aerosol surface area and volume by a factor of about 2-3 in all seasons (noMSOA versus
633 BASE, Fig. 3), to be slightly over the 75th percentile of the observations (Fig. 3). Surface area and
634 volume results from the simulation are very sensitive to the size-distribution simulation near the
635 282 nm diameter cut-off that contributes to differences between these simulations.

636
637 Figure 4 demonstrates that MSOA has a feedback on NPF. With lower aerosol surface area and
638 lower condensation sink (noMSOA), the N3 and N3-N10 below 2 km altitude are strongly
639 overpredicted because NPF increases and a lack of growth to larger sizes impacts N3-N10. During
640 November, the N3 and N3-N10 overprediction also occurs at altitudes above 2 km because MSOA
641 has a relatively greater influence on aerosol surface area at those altitudes in this season (Fig. 3).
642 In this less-polluted late fall season, the influence of MSOA is relatively stronger at higher altitudes
643 than in other seasons. Model-measurement agreement improves for N3 and N3-N10 with the
644 addition of MSOA (simulation BASE relative to noMSOA, Fig. 4). Kazil et al. (2011) also found
645 that condensing vapors generate a condensation sink that moderates the level of NPF in the marine
646 environment. As well, recent studies from the Arctic indicate a key contribution to particles from
647 condensing marine organic vapors (Burkart et al., 2017a; Willis et al., 2017; Croft et al., 2019).
648 The impact of MSOA on the simulated N10 vertical profiles is small. The cloud filtering, which

649 we applied to the model and measurement aerosol samples along the flight track, preferentially
650 removes some of the cloud-processed samples, and contributes to this result.

651

652 **3.4 Role of DMS**

653

654 Figure 2 shows that DMS also has a control on the simulated MBL aerosol size distributions
655 (BASE versus noDMS) for the four seasons of the NAAMES campaigns. The total simulated
656 number of particles attributed to DMS is lowest during the phytoplankton bloom minima (winter,
657 November 2015) and greater in other seasons. For example, for particle diameters at 40 nm, the
658 DMS-related contribution to the size distribution (Fig. 2) is about 200-300 cm⁻³ in all seasons,
659 except less than 50 cm⁻³ during the bloom minima. Sulfuric acid from the oxidation of DMS has a
660 two-fold role in both NPF and in growing particles. However, as indicated by simulations
661 noABLNUC and noMSOA relative to BASE (Fig. 2), the DMS contribution is in concert with
662 both (1) a source of condensable marine organic vapors and (2) NPF near and above the MBL top.
663 The contribution of DMS to MBL particles is consistent with the findings of many previous
664 studies, including Chang et al. (2011), Ghahremaninezhad et al. (2016), Park et al. (2018), Sanchez
665 et al. (2018), Mahmood et al. (2019), Quinn et al. (2019) and Veres et al. (2020).

666

667 Simulation noABLNUC relative to noDMS for the marine-influenced MBL size distributions (Fig.
668 2) suggests that anthropogenic influences make a contribution as a source of particle-precursor
669 vapors for NPF, in addition to DMS. This relative contribution is particularly strong during the
670 accumulation phase (March/April 2018). In our simulations, anthropogenic SO₂ is oxidized to
671 H₂SO₄ and contributes to the particle precursors for NPF near and above the MBL top (in addition
672 to DMS oxidation products), followed by particle growth on descent into the MBL. As a result,
673 Fig. 2 shows a greater underprediction of aerosol number for simulation noABLNUC versus
674 noDMS.

675

676 Figure 6 indicates that the simulated DMS is generally consistent (MFEs ranging from 0.12 to
677 0.26, Supplementary Table S2) with the observed DMS mixing ratio vertical profiles and their
678 seasonal cycle for the NAAMES campaigns. DMS makes the strongest contribution to simulated
679 sulfate mass concentrations in the lowest 2 km during the May/June climax transition, reducing

680 model-measurement bias from about 40% to 10% (Fig. 5). Figures 3 and 4 suggest that in the
681 lowest 2 km of the atmosphere, DMS contributes to both NPF and particle growth as there are
682 increases in N3, N10, N3-N10, particle surface area and volume for simulations BASE versus
683 noDMS. However, this behavior is co-dependent on conditions favorable to NPF near the MBL
684 top and a source of MSOA.

685

686 **3.5 Role of ship traffic emissions**

687

688 Ship emissions are a source of primary and secondary particles, as well as a control on oxidants
689 (Corbett et al., 2010; Vinken et al., 2011; Holmes et al., 2014). Our simulations suggest that ship
690 emissions are also a control on the NAAMES-region MBL marine-influenced aerosol size
691 distributions (Fig. 2, noSHIPS versus BASE). For example, for the simulated summertime MBL
692 size distribution at particle diameters at 40 nm, about 100-200 particles cm^{-3} are attributed to ship
693 emissions (Fig. 2). Table 2 shows that during the phytoplankton bloom and March/April
694 accumulating phase, the noSHIPS simulation agrees more closely with the measurements than the
695 BASE simulation, although both are within acceptable agreement ($\text{MFE} < 0.5$). These simulation
696 challenges highlight the importance of future work to better understand the role of oxidants from
697 ship emissions on particle production in the marine environment and to understand the size
698 distribution of primary marine emissions.

699

700 Ship emissions make about a 50% contribution to the simulated sulfate campaign-median near-
701 surface mass concentration in vertical profiles over the NAAMES study region (Fig. 5). For our
702 simulations the impact of ship emissions on particle number is mostly limited to the lowest 2 km.
703 Simulation BASE relative to noSHIPS suggest that about 10% of the N10 in the lowest 500 m of
704 the atmosphere is attributed to ship emissions (Fig. 4). Figure 4 (right column) indicates that among
705 the five factors considered by our sensitivity studies, ship emissions are among the smallest
706 influence on the NPF. Major trans-Atlantic ship traffic routes (Corbett et al. (2007) are included
707 in the NAAMES study region. Enhancements in observed benzene mixing ratios in the MBL
708 relative to other long-lived tracers of anthropogenic emissions such as acetone (not associated with
709 ship traffic) are observational evidence that ship emissions influence the study region
710 (Supplementary Fig. S9).

711
712 Figure 6 demonstrates that atmospheric DMS mixing ratios are also sensitive to ship emissions.
713 This effect occurs because ship emissions are a control on oxidants in the MBL, and enhance OH
714 and NO₃, which are chemical sinks of DMS. As a result, simulated DMS mixing ratios increase
715 when ship emissions are removed. As ship traffic is expected to change in future years with
716 changes to routes and regulations (Gilgen et al., 2018; Bilsback et al. (in press)), the relative
717 importance of ship emissions in the North Atlantic MBL will likely change.

718
719 **3.6 Role of sea spray**

720
721 Figure 2 shows that simulated sea spray acts as a condensation sink in the MBL. Without sea spray
722 emissions, there is an increase in the number of sub-200 nm diameter particles (simulation noSS
723 relative to BASE). However, this relative increase in simulated number is less than that attributed
724 to other factors considered in the previous sections. While not a strong contributor to particle
725 number in our simulations, sea spray is the dominant contributor to aerosol mass.

726
727 The simulated campaign-median MBL sea spray mass concentrations are within the measurement
728 range of 3-8 $\mu\text{g m}^{-3}$ found by Saliba et al. (2019) (Supplementary Fig. S10), despite the
729 considerable uncertainties related to size-resolved sea spray emissions (e.g., Bian et al., 2019;
730 Regayre et al. (2020)). Regayre et al. (2020) found that global sea spray emissions could be under
731 predicted by a factor of 3 by the Gong (2003) parameterization. We conducted a simulation with
732 factor-of-3 scaling of the sea spray emissions (Supplementary Figs. S11-S14, Supplementary
733 Table S4) and found a decrease in MBL number concentrations, rather than an increase. This
734 reduction occurred because the enhanced condensation sink from the additional sea spray
735 emissions suppressed NPF. Our simulations use the Gong (2003) parameterization with a sea-
736 surface-temperature-based scaling as described by Jaeglé et al. (2011), so are not directly
737 comparable to the Regayre et al. (2020) findings. Nonetheless, these findings highlight the
738 importance of ongoing work to improve size-resolved sea spray emissions parameterizations in
739 models. The direct radiative effect of this sea spray mass loading is examined in the following
740 section.

741

742 **3.7 Radiative effects attributed to NPF near MBL top, MSOA, DMS and ship emissions**

743

744 Figure 7 shows the simulated two-month mean direct radiative effect (DRE) attributed to the five
745 factors we consider, (1) NPF near and above the MBL top, (2) MSOA, (3) DMS, (4) ship emissions
746 and (5) primary sea spray emissions and magnitude of the regional-mean DREs over a region of
747 the North Atlantic (40-60 °N, 20-50 °W). We note that the radiative effects attributed to the
748 separate factors are not linearly additive because the factors impact each other non-linearly. For
749 example, turning off either MSOA or nucleation above the boundary layer would shut down the
750 majority of the production of accumulation-mode particles in the MBL (Fig. 2) since these particles
751 require both nucleation and growth. Hence, adding the radiative effects from these two factors
752 would result in double counting some radiative effects. Figure 7 indicates that the strongest
753 calculated DRE is attributed to sea spray, which dominates the aerosol mass loading in the MBL.
754 The sea spray DRE has a maximum during the 2018 March/April accumulating phase, which is a
755 time of frequent synoptic scale storms with strong winds. Stormy conditions prevented the R/V
756 Atlantis from travelling north of 45 °N during this final NAAMES campaign.

757

758 The strongest DRE values attributed to the above boundary layer NPF, MSOA, DMS and ship
759 emission factors are during the summer season (climax transition (bloom maxima) and declining
760 phase). This result highlights the link between the level of marine biogenic activity and aerosol
761 climate effects. The second strongest individual DRE is attributed to condensing marine organic
762 vapors, yielding MSOA. In our simulations, MSOA contributes significantly to particle growth to
763 diameters of about 100 to 200 nm, which can then interact directly with radiation (Fig. 2). This
764 effect is greatest in the declining phase because we used a temperature-dependent parameterization
765 and sea surface temperatures are warmest during the late summer. The DRE geographic
766 distribution suggests an increasing role for MSOA towards southern latitudes, again due to the
767 temperature-dependent parameterization. Further work is needed to examine the role of MSOA in
768 the more southerly latitudes as we cannot explicitly test this result across the annual cycle with the
769 NAAMES observations.

770

771 Among the factors considered, Figure 7 shows that during the time of the May/June phytoplankton
772 bloom, the aerosols produced and grown by the oxidation products of DMS have the third strongest

773 impact on the DRE, greatest over the regions where the bloom is located. The DRE is -0.10 Wm^{-2}
774 over the region between $40\text{-}60^\circ\text{N}$ and $20\text{-}50^\circ\text{W}$ during the bloom maxima and diminishes to -
775 0.005 Wm^{-2} during the bloom minima. This simulated impact of DMS relies in part on (1)
776 conditions favoring NPF processes near and above the MBL top, and (2) growth by MSOA as the
777 nascent DMS-related particles descend in the MBL. DMS (similar to MSOA) also contributes to
778 the DRE over the continents as these vapors have a lifetime of about a day in our simulations and
779 can be transported before their oxidation products are available for condensation. Once available
780 for condensation, DMS products and MSOA contribute to growing particles (of both marine and
781 continental origin) to sizes that can interact more strongly with radiation (diameters near $100\text{-}200$
782 nm). Particles arising from DMS grow during transport, and some particles may only reach sizes
783 large enough to interact with radiation when they are over the continents.

784

785 The DRE attributed to the near and above MBL top NPF factor (Fig. 7, top row, ABLNUC) is
786 strongest in summertime, during the May/June climax transition (bloom maxima) and
787 August/September declining phase. Summertime is the season of the greatest photochemical
788 production of particle precursors for NPF. In order to contribute to the DRE, this NPF factor acts
789 in synergy with the other factors, particularly DMS as a source of particle precursors and MSOA
790 for particle growth, such that during the May/June climax transition season the DREs for those
791 factors dominate over the NPF factor (ABLNUC, Fig. 7).

792

793 The DREs for ship emissions have a similar geographic distribution as those for DMS. In these
794 regions, major international ship traffic routes are coincident with regions of higher biogenic
795 activity, enabling an interaction of biogenic and anthropogenic emissions. Ships enhance oxidant
796 levels, which promote formation of biogenic aerosol precursors such as sulfuric acid and MSA
797 that arise from oxidation of DMS. Condensing vapors of marine origin (such as DMS products
798 and MSOA precursors) can also help to grow particles arising from ship emissions to sizes large
799 enough to interact directly with radiation. As a result, the largest DRE attributed to ship emissions
800 is during the phytoplankton bloom maxima. Figure 7 also suggests that ship emissions could
801 contribute to the DRE over the continents. This effect occurs because ship emissions include
802 particle precursors, oxidants, and primary particles that are transported and interact with
803 continental pollution to form and grow particles to sizes that can interact with radiation over the

804 continents as well as over the oceans. Figure 6 shows that there is a ship-emission-related control
805 on atmospheric DMS mixing ratios, which increase when the ship-source oxidants are removed.

806

807 Figure 8 shows the calculated two-month mean cloud-albedo aerosol indirect effect (AIE)
808 attributed to each of the same five factors that we considered for the DREs. The AIEs are about an
809 order-of-magnitude larger than the calculated DRE for each respective factor with the exception
810 of sea spray. The AIE is strongly controlled by changes to highly reflective MBL clouds, which
811 are in turn very sensitive to the aerosol number concentrations (diameters larger than about 50 to
812 70 nm that can act as CCN), which are controlled by the MBL-related factors examined here. On
813 the other hand, the DRE is relatively more sensitive to aerosol abundance in mid-tropospheric
814 layers, which are less influenced by the considered processes.

815

816 The strongest simulated AIEs for all considered factors are during the May/June climax transition
817 (Fig. 8). There is a strong synergy among all factors that reach their maxima during May/June
818 when the effective combination of sources, photochemistry and particle production/growth
819 processes peak. As well, during summertime, the magnitude of the AIE for all factors is greater in
820 the more northward regions of the North Atlantic relative to more southerly latitudes. These more
821 northerly regions are less influenced by continental pollution and have lower CCN concentrations,
822 coupled with persistent low cloud cover. These conditions make these regions quite sensitive to
823 the factors controlling MBL aerosol size distributions studied here.

824

825 In all seasons, we calculated a stronger AIE related to (1) NPF near and above the MBL top
826 (ABLNUC, top row, Fig. 8) and (2) MSOA (contributor to particle growth) than to (1) DMS (2)
827 ship emissions and (3) sea spray emissions. In our simulations, the major source of CCN-sized
828 particles in the North Atlantic MBL during the summer is particle nucleation near and above the
829 MBL top with growth by MSOA. Without either of these factors, the number concentration of
830 CCN-sized particles in the simulations drops dramatically (Fig. 2). Hence, it is unsurprising that
831 the largest simulated AIEs are due to these two factors during the summertime (climate transition
832 and declining phase). The stronger AIEs attributed to NPF near and above the MBL top (Fig. 8,
833 top row, ABLNUC) relative to DMS and ship emissions indicate that near and above MBL NPF
834 in our simulations is controlled not only by the sulfuric acid from the oxidation of DMS or ship

835 SO₂, but also arising from other sources, including SO₂ transported from continental sources.
836 However, the maximum North Atlantic regional-mean AIE attributed to ship emissions (-0.62 W
837 m⁻² for the May/June climax transition) still exceeds the global mean effect of -0.155 W m⁻²
838 attributed to international shipping calculated by Jin et al. (2018), showing the strong location-
839 dependence and seasonality of this factor. Ship emissions enhance the oxidation rate of DMS, such
840 that the largest AIE attributed to ships occurs during the phytoplankton bloom due to increased
841 particle formation/growth during this season.

842
843 In our simulations, sea spray has a lower contribution to aerosol number concentrations, among
844 the factors considered, and as a result has the smallest AIEs. However, recent studies have pointed
845 out that there are knowledge gaps related to the sea spray emissions parameterizations (e.g Bian et
846 al., 2019; Regayrre et al., 2020). Future work is needed to gain confidence in the magnitude of the
847 AIE attributed to sea spray.

848
849 We caution that both the DRE and AIE calculations represent a relative contribution of the
850 considered factors to climate effects in the North Atlantic. However, further work is needed to gain
851 confidence in the absolute magnitudes. The activation-style nucleation, which we used as a proxy
852 for the unknown nucleation mechanisms above the marine boundary layer, contributes uncertainty
853 to the climate effects of this nucleation. There is much more work that needs to be done regarding
854 the role of MSOA in this system. Certainly, if MSOA is contributing directly to NPF, it would
855 increase MSOA's climatic importance. However, we have little knowledge of the MSOA
856 precursor species, their chemical lifetimes, and their role in NPF, so we did not explore these
857 dimensions in the study. Like the DRE estimates, the separate AIEs are not linearly additive. Other
858 aerosol indirect effects related to changes in cloud lifetime and precipitation are the subject of
859 future work. In summary, these calculated DREs and AIEs suggest that aerosol-climate impacts
860 for North Atlantic regions are controlled by a combination of strong biogenic and anthropogenic
861 influences and that the nucleation near and above the MBL top contributes to important radiative
862 effects.

863
864 **4. Conclusions**
865

866 In this study, we examined aerosol size distribution and composition measurements from the
867 NAAMES campaigns. These ship and aircraft campaigns took place over four separate stages of
868 the annual cycle of marine biogenic activity in the Northwest Atlantic during 2015-2018. We used
869 the GEOS-Chem-TOMAS model with size-resolved aerosol microphysics to interpret these
870 NAAMES measurements. Observations in layers of the lower troposphere below 6 km showed
871 enhancements in the campaign-median number concentration of particles with diameters between
872 3-10 nm. These enhancements indicated new particle formation, and were most pronounced during
873 the May/June 2016 climax transition (phytoplankton bloom maxima) in the lowest 2 km of
874 atmosphere, particularly near and just above the boundary layer top. This lower tropospheric
875 region near and above the MBL top is a key region for marine NPF. This zone above the MBL
876 clouds is generally very clean, which favors both NPF and strongly sunlit, which favors the
877 photochemical oxidative production of particle precursors for NPF. The November 2015 winter
878 transition (phytoplankton bloom minima) was characterized by the lowest particle number
879 concentrations. During the summer months, OM, followed by sulfate mass concentrations made
880 strong contributions the total aerosol loading in the lowest 2 km. However, sea spray dominated
881 the MBL aerosol mass loading. Peak near-surface sulfate concentrations occurred in May/June
882 during the phytoplankton bloom, whereas peak near-surface OM concentrations were in
883 August/September. Campaign-median MBL aerosol size distributions were dominated by Aitken
884 mode particles (diameters 10-100 nm) during the summertime (May/June climax transition and
885 August/September declining phase). The larger accumulation mode particles were dominant in the
886 November winter transition and March/April accumulation phase.

887
888 Our simulations suggested that a synergy of key factors contributed to Northwest Atlantic MBL
889 aerosol size distributions, including (1) new particle formation near and above the MBL top; (2)
890 growth of the newly formed particles by condensation of marine organic vapors, forming marine
891 secondary organic aerosol (MSOA), which yields more abundant CCN-sized particles that descend
892 into the MBL while continuing to grow and being subject to cloud processing (e.g., aqueous-phase
893 aerosol production, which does not add to particle number); (3) DMS-oxidation products that
894 contribute to particle formation and growth; (4) ship emissions, which are a source of primary and
895 secondary particles and also contribute to atmospheric oxidants and (5) sea spray emissions, which
896 also provide a condensation sink that suppresses particle formation. These findings are in

897 agreement with previous observational-based studies for the North Atlantic region (e.g., Sanchez
898 et al., 2018; Zheng et al., 2020)

899

900 We calculated the aerosol direct (DRE) and cloud-albedo indirect (AIE) radiative effects over the
901 North Atlantic attributed to five key factors controlling MBL aerosols. The cooling effects were
902 about a factor of 10 larger for the AIEs than the respective DREs except for sea spray, which
903 dominated the DRE. The strong AIE response was attributed to the strong sensitivity of the MBL
904 cloud reflectivity to the MBL-related factors that we examined. Mid-tropospheric aerosol (altitude
905 of transport of continental pollution) has a strong impact on the DRE and the factors that we
906 considered had less impact at these altitudes. The maximum regional-mean (40-60 °N, 20-50 °W)
907 DRE for our simulations was -1.37 W m^{-2} , attributed to sea spray during the March/April
908 accumulating phase, which is a time of strong synoptic-scale storms in the Northwest Atlantic,
909 enhancing wind-generated sea spray. This strong DRE attributed to sea spray highlights the
910 importance of work to better constrain parameterizations for models. The second strongest DRE
911 was connected to the temperature-dependent source of MSOA, which had a key role in growing
912 simulated particles to large enough (diameters of 100-200 nm) to strongly reflect incoming solar
913 radiation. The maximum AIE was -3.37 W m^{-2} , for the May/June climax transition phase (peak
914 phytoplankton bloom). This AIE was related to the role MSOA in growing new particles to CCN
915 sizes as they descend into the MBL and are subject to further growth in clouds after their formation
916 near the MBL top. The AIE attributed to the NPF factor was nearly as large (-2.27 W m^{-2}) during
917 May/June. The NPF and MSOA factors act in concert with each other and removal of either of
918 these factors contributed to shutdown the production of cloud-condensation-nuclei-size particles.
919 Our study demonstrated acceptable model-measurement agreement for our base simulation, such
920 that our simulations can be employed to examine the *potential* role and relative importance of the
921 considered factors in the DRE and AIE. However, we caution that further work is needed to gain
922 confidence in the absolute magnitudes. In particular, the activation-style nucleation, which we
923 used as a proxy for the unknown nucleation mechanism above the marine boundary layer, adds
924 uncertainty to the climate effects of this nucleation

925

926 This study highlighted the importance of processes connected to both marine biogenic activity and
927 anthropogenic activity in controlling aerosol size distributions in the Northwest Atlantic MBL. We

928 identified key factors, which could be the focus of future work. Particularly, work is needed to
929 better understand the nature, flux, and chemistry of marine organic vapors that can form MSOA.
930 As well, work is needed to better understand the contributors to NPF near and above the MBL top.
931 Further work is also needed to understand the interactions of the considered factors with cloud
932 processing of aerosols and its relative importance in particle growth. As the Earth's climate
933 changes and shipping traffic/regulations/routes change, work to understand the source strength of
934 DMS, MSOA, shipping and sea spray emissions is highly relevant. Such work will bridge the
935 knowledge gaps related to factors controlling aerosols in the marine MBL and their climate
936 impacts.

937
938 **Code and data availability.** The NAAMES project website is at <https://naames.larc.nasa.gov>. The
939 NAAMES airborne and ship datasets used in this paper are publicly available and permanently
940 archived in the NASA Atmospheric Science Data Center (ASDC;
941 <https://doi.org/10.5067/Suborbital/NAAMES/DATA001>) and the SeaWiFS Bio-Optical Archive
942 and Storage System (SeaBASS; <https://doi.org/10.5067/SeaBASS/NAAMES/DATA001>). The
943 ship datasets generated during and analyzed for NAAMES studies are also available in the UCSD
944 Library Digital Collection repository, <https://doi.org/10.6075/J04T6GJ6>. The GEOS-Chem model
945 is freely available for download at <https://github.com/geoschem/geos-chem> (last access 19 July
946 2020).

947
948 **Supplement link.**

949
950 **Author contributions.** BC, RVM and JRP designed the study. BC conducted the GEOS-Chem-
951 TOMAS simulations, led the related analysis, and wrote the manuscript with contributions from
952 all coauthors. RHM, ECC, and LDZ contributed the aerosol measurements from aboard the NASA
953 C130 aircraft. AW, MM and AS contributed the gas-phase measurements from aboard the NASA
954 C130 aircraft. LMR and GS contributed the aerosol measurements from aboard the R/V Atlantis.
955 RYWC and HL contributed to the interpretation of model-measurement comparisons. EEM
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957 contributed the satellite DMS data set.

958

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960

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978

979 **References.**

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1751

1752 **Tables and figure captions and tables**

1753

Simulation	Description
BASE	Control simulation with GEOS-Chem-TOMAS model (GCT12.1.1) as described in Sect. 2.2
noABLNUC	Same as BASE, excluding the surrogate activation-type particle nucleation parameterization above the marine boundary layer to about 2 km altitude, as described in Sect. 2.2
noMSOA	Same as BASE, excluding the temperature-dependent marine organic vapors, forming marine secondary organic aerosol (MSOA)
noDMS	Same as BASE, excluding all emissions of DMS
noSHIPS	Same as BASE, excluding all ship emissions
noSS	Same as BASE, excluding all sea spray emissions

1754

1755 **Table 1:** GEOS-Chem-TOMAS simulation acronyms. Simulations and methodology are
 1756 described in detail in Sect. 2.2 and 2.3.

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Simulation	Nov 2015 Bloom Minima	May/June 2016 Bloom Maxima	Aug/Sept 2017 Declining Phase	Mar/Apr 2018 Accumulating	Annual Mean
BASE	0.20	0.33	0.04	0.28	0.21
noABLNUC	0.95	0.51	0.89	0.50	0.71
noMSOA	0.76	0.31	0.84	0.59	0.63
noDMS	0.44	0.27	0.43	0.06	0.30
noSHIPS	0.31	0.13	0.23	0.21	0.22
noSS	0.31	0.24	0.12	0.28	0.24

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1760 **Table 2:** Mean fractional error (MFE) between observations and the six GEOS-Chem-TOMAS
 1761 simulations described in Sect. 2.2 and Table 1 for the ship-track campaign-median aerosol size
 1762 distributions shown in Fig. 2.

1763

1764 **Figure 1:** Cruise and aircraft tracks for the 2015-2018 NAAMES campaigns. Flight altitudes
1765 below 3 km are color-coded in medium blue and above 3 km in red. Ship tracks campaigns are
1766 color-coded for each year as shown by the legend, and as follows: Orange: November 2015
1767 winter transition (bloom minima); Cyan: May/June 2016 climax transition (bloom maxima);
1768 Purple: August/September 2017 declining phase; Green: March/April 2018 accumulation phase.
1769

1770 **Figure 2:** NAAMES cruise-track campaign-median marine boundary layer aerosol size
1771 distributions from marine-influenced SEMS (particle diameters 20-500 nm) observations (black,
1772 with 25th to 75th percentiles in grey) and for the six GEOS-Chem-TOMAS simulations as
1773 described in Table 1 (color-coded as shown in legend).
1774

1775 **Figure 3:** Vertical profiles of NAAMES campaign-median integrated SMPS observations aboard
1776 aircraft at standard temperature and pressure (STP) for particles with diameters of 10 to 282 nm
1777 (black, with 25th-75th percentiles in grey) and for the six GEOS-Chem-TOMAS simulations
1778 described in Table 1 (color-coded as shown in legend). All measurement and model output is
1779 binned at 500 m resolution and campaign-median values plotted at the mid-point of each bin
1780 starting at 250 m above the surface. Lines show linear interpolation between these values.

1781 **Figure 4:** Vertical profiles of NAAMES campaign-median total number concentrations for
1782 particles with diameters larger than 3 nm (N3), 10 nm (N10) and between 3 to 10 nm (N3-N10)
1783 from CPC observations aboard aircraft at standard temperature and pressure (STP) (black, with
1784 25th-75th percentiles in grey) and for the six GEOS-Chem-TOMAS simulations described in Table
1785 1 (color-coded as shown in legend). All measurement and model output is binned at 500 m
1786 resolution and campaign-median values are plotted at the mid-point of each bin starting at 250 m
1787 above the surface. Lines show linear interpolation between these values.
1788

1789 **Figure 5:** Vertical profiles of NAAMES campaign-median aerosol non-refractory sulfate and
1790 organic mass concentrations at standard temperature and pressure (STP) from Aerosol Mass
1791 Spectrometer and refractory black carbon from Single Particle Soot Photometer observations
1792 aboard aircraft (black, with 25th-75th percentiles in grey) and for the six GEOS-Chem-TOMAS
1793 simulations described in Table 1 (color-coded as shown in legend). Simulated sulfate shown is
1794

1795 non-sea-salt-sulfate. All measurement and model output is binned at 500 m resolution and
1796 campaign-median values are plotted at the mid-point of each bin starting at 250 m above the
1797 surface. Lines show linear interpolation between these values.

1798
1799 **Figure 6:** Vertical profiles of NAAMES cruise-track campaign-median observed dimethyl sulfide
1800 (DMS) mixing ratios (black, 25th-75th percentiles in grey) from aboard aircraft and for the six
1801 GEOS-Chem-TOMAS simulations described in Table 1 (color-coded as shown in legend).
1802 Simulations BASE, noABLNUC, noMSOA and noSS are nearly coincident. All measurement and
1803 model output is binned at 500 m resolution and campaign-median values plotted at the mid-point
1804 of each bin starting at 250 m above the surface. Lines show linear interpolation between these
1805 values. Note the horizontal scale change between panels.

1806
1807 **Figure 7:** GEOS-Chem-TOMAS-simulated two-monthly-mean aerosol direct radiative effect
1808 (DRE) attributed to five key factors. Top row: Above boundary layer particle nucleation
1809 (ABLNUC); Second row: Particle growth by marine secondary organic aerosol (MSOA); Third
1810 row: Particle formation/growth due to DMS-oxidation products (DMS); Fourth row: Shipping
1811 emissions contribution to particles (SHIPS); Bottom row: Sea spray (SS). DREs are in columns
1812 for the following time periods, March/April 2018 (Accumulating Phase), May/June 2016 (Climax
1813 Transition, Bloom Maxima), August/September 2017 (Declining Phase), and October/November
1814 2015 (Winter Transition, Bloom Minima). DREs for ABLNUC, MSOA, DMS, SHIPS, and SS are
1815 calculated using the differences in the top-of-the-atmosphere solar flux between simulation BASE
1816 and respective sensitivity simulations (noABLNUC, noMSOA, noDMS, noSHIPS, noSS). Values
1817 shown are area-weighted-mean DREs over the region bounded by 40-60 °N, 20-50 °W.

1818
1819 **Figure 8:** GEOS-Chem-TOMAS-simulated two-monthly-mean aerosol cloud-albedo indirect
1820 radiative effect (AIE) attributed to five key factors. Top row: Above boundary layer particle
1821 nucleation (ABLNUC); Second row: Particle growth by marine secondary organic aerosol
1822 (MSOA); Third row: Particle formation/growth due to DMS-oxidation products (DMS); Fourth
1823 row: Shipping emissions contribution to particles (SHIPS); Bottom row: Sea spray (SS). AIEs are
1824 in columns for the following time periods, March/April 2018 (Accumulating Phase), May/June
1825 2016 (Climax Transition, Bloom Maxima), August/September 2017 (Declining Phase), and

1826 October/November 2015 (Winter Transition, Bloom Minima). AIEs for ABLNUC, MSOA, DMS,
1827 SHIPS, and SS are calculated using the differences in the top-of-the-atmosphere solar flux between
1828 simulation BASE and respective sensitivity simulations (noABLNUC, noMSOA, noDMS,
1829 noSHIPS, noSS). Values shown are area-weighted-mean AIEs over the region bounded by 40-60
1830 °N, 20-50 °W.

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