Supplementary information for:

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

Betty Croft[1], Randall V. Martin[2,1], Richard H. Moore[3], Luke D. Ziemba[3], Ewan C. Crosbie[3,4], Hongyu Liu[5], Lynn M. Russell[6], Georges Saliba[6], Armin Wisthaler[7,8], Markus Müller[7], Arne Schiller[7], Martí Galí[9], Rachel Y.-W. Chang[1], Erin E. McDuffie[1,2], Kelsey R. Bilsback[10], and Jeffrey R. Pierce[10]

[1]Department of Physics and Atmospheric Science, Dalhousie University, Halifax, NS, Canada
[2]McKelvey School of Engineering, Washington University in St. Louis, St. Louis, MO, USA
[3]NASA Langley Research Center, Hampton, VA, USA
[4]Science Systems and Applications, Inc., Hampton, VA, USA
[5]National Institute of Aerospace, Hampton, VA, USA
[6]Scripps Institute of Oceanography, University of California, San Diego, La Jolla, CA, USA
[7]Institute for Ion Physics and Applied Physics, University of Innsbruck, Technikerstrasse 25, 6020 Innsbruck, Austria
[8]Department of Chemistry, University of Oslo, P.O. 1033 – Blindern, 0315 Oslo, Norway
[9]Barcelona Supercomputing Center (BSC)
[10]Department of Atmospheric Science, Colorado State University, Fort Collins, CO, USA
Correspondence to: Betty Croft (betty.croft@dal.ca)

S1. The simulated marine organic vapor source flux was tuned to yield an acceptable annual-mean MFE (<0.50) between observations and simulations for the NAAMES campaign-median ship-track marine-influenced aerosol size distributions (as described in Sect. 2.2). The aerosol size distributions for a set of sources fluxes are shown in Fig. S1. Table S1 shows the MFEs for this set of source fluxes. We found the lowest annual mean MFE for the source flux of 70T-350, where T is in ^{o}C and the flux is in kg m^{-2} d^{-1}. We caution that this tuning was specific for the NAAMES region and for a certain GEOS-Chem-TOMAS model configuration. As a result, this source flux may not perform as well in other models, other GEOS-Chem versions and other regions. Further work is needed to better constrain the flux of marine organic vapors.

The selected parameterization yielded agreement within the 25th to 75th percentiles for the campaign-median vertical profiles in the lowest 1 km for total aerosol number (N3, N10 and N3-N10) and integrated SMPS number, and near-surface OM concentrations (Figs. S2-S4). Figure S2 shows slight overprediction outside of these percentiles for the integrated SMPS surface area and volume below 2 km.

S2. We found enhancements in benzene (Fig. S5) relative to other tracers, such as acetone (Fig. S6), which have anthropogenic sources but not associated with ship emissions. These findings are supportive of the study region being influenced by ship emissions.

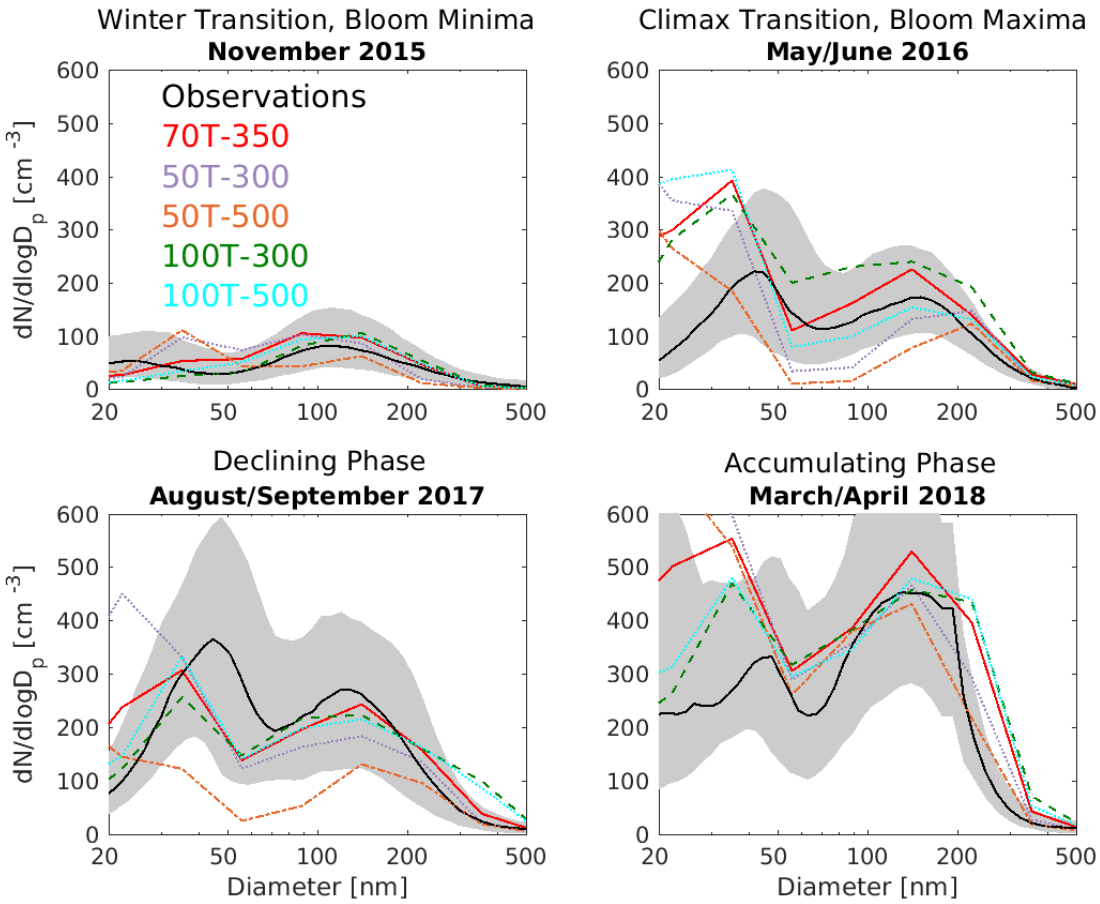

Figure S1: NAAMES cruise-track campaign-median marine boundary layer aerosol size distributions from marine-influenced SEMS observations (black, with 20th to 80th percentiles in grey) and for five GEOS-Chem-TOMAS simulations with different assumptions for the temperature dependence of the flux of condensable organic vapors (color-coded as shown in legend, flux in μg m^{-2} d^{-1} and T in ºC). Linestyles: Solid: Observations, 70T-350; Dotted: 50T-300, 100T-500; Dash-dot: 50T-500; Dashed: 100T-300.

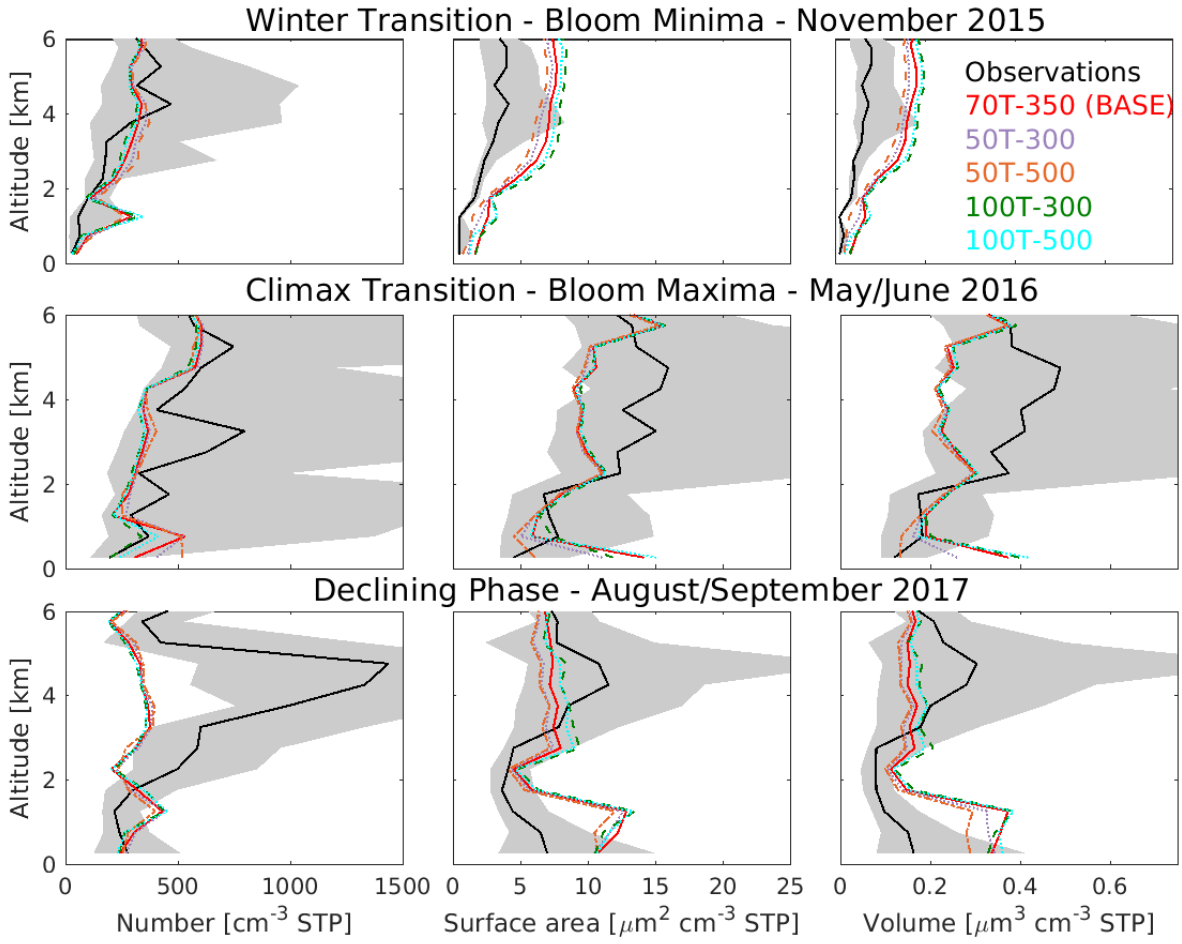

Figure S2: Vertical profiles of NAAMES campaign-median integrated SMPS observations at standard temperature and pressure (STP) for particles with diameters of 10 to 282 nm (black, with 25th-75th percentiles in grey) and at STP for five GEOS-Chem-TOMAS simulations with different assumptions for the temperature dependence of the flux of condensable marine organic vapors (color-coded as shown in legend, flux in μg m^{-2} d^{-1} and T in ^{o}C). Linestyles: Solid: Observations, 70T-350; Dotted: 50T-300 100T-500; Dash-dot: 50T-500; Dashed: 100T-300.

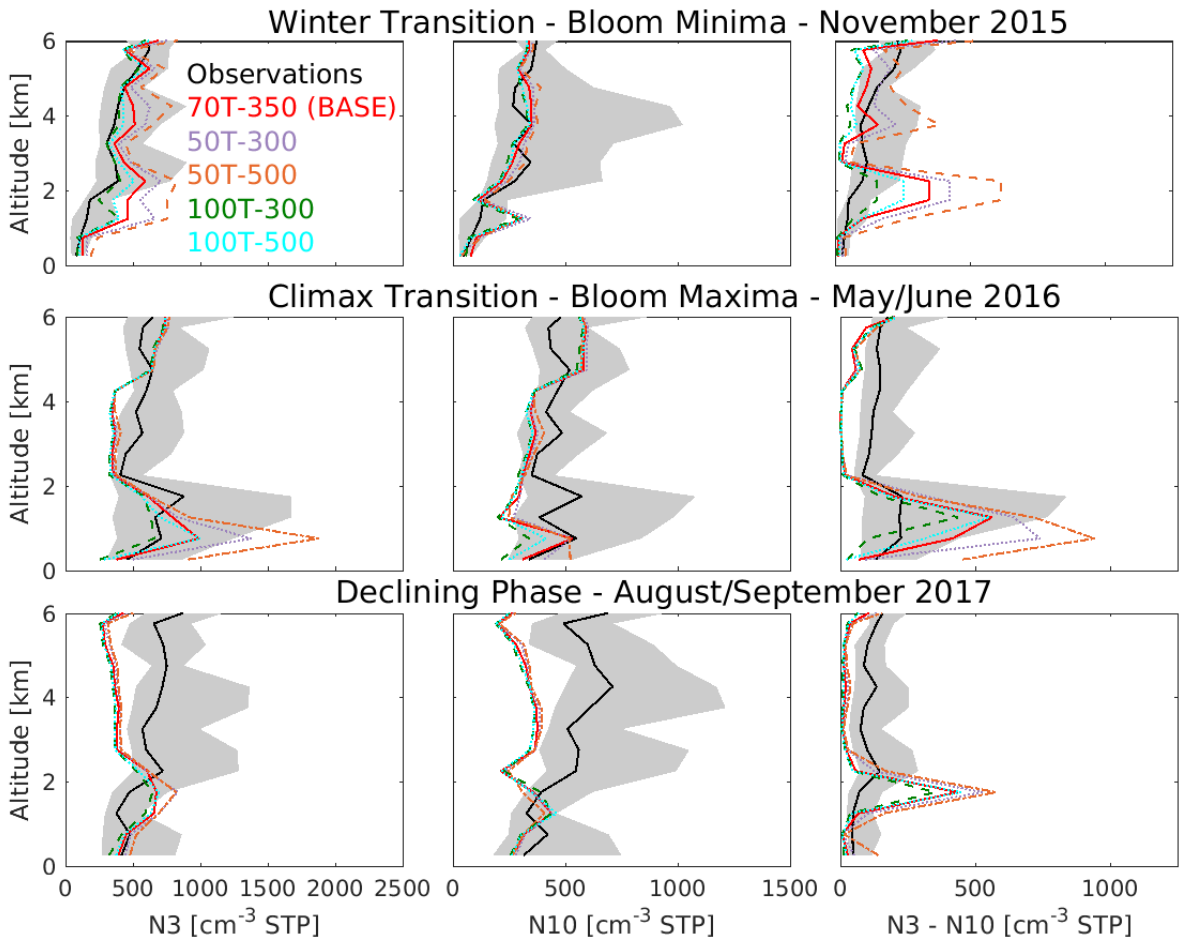

Figure S3: Vertical profiles of NAAMES campaign-median total number concentrations for particles with diameters larger than 3 nm (N3), 10 nm (N10) and between 3 to 10 nm (N3-N10) from CPC observations at standard temperature and pressure (STP) (black, with 25^{th}-75^{th} percentiles in grey) and at STP for five GEOS-Chem-TOMAS simulations with different assumptions for the temperature dependence of the flux of condensable marine organic vapors (color-coded as shown in legend, flux in μg m^{-2} d^{-1} and T in °C). Linestyles: Solid: Observations, 70T-350; Dotted: 50T-300 100T-500; Dash-dot: 50T-500; Dashed: 100T-300.

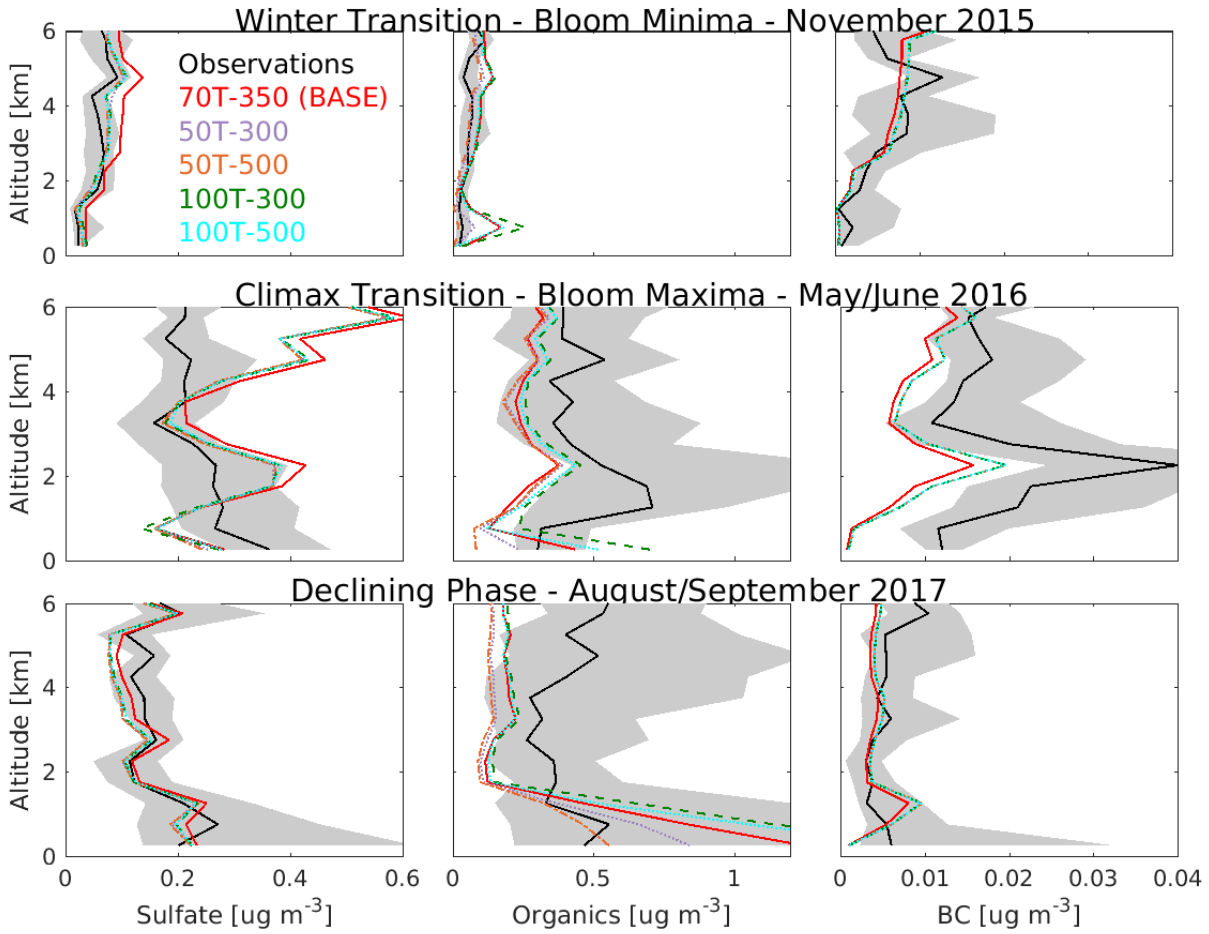

Figure S4: Vertical profiles of NAAMES campaign-median aerosol non-refractory sulfate and organic mass concentrations in μg m^{-3} from Aerosol Mass Spectrometer and refractory black carbon from SP2 observations at standard temperature and pressure (STP) (black, with 25^{th}-75^{th} percentiles in grey) and at STP for five GEOS-Chem-TOMAS simulations with different assumptions for the temperature dependence of the flux of marine condensable organic vapors (color-coded as shown in legend, flux in μg m^{-2} d^{-1} and T in °C). Simulated sulfate shown is non-sea-salt-sulfate. Linestyles: Solid: Observations, 70T-350; Dotted: 50T-300 100T-500; Dash-dot: 50T-500; Dashed: 100T-300.

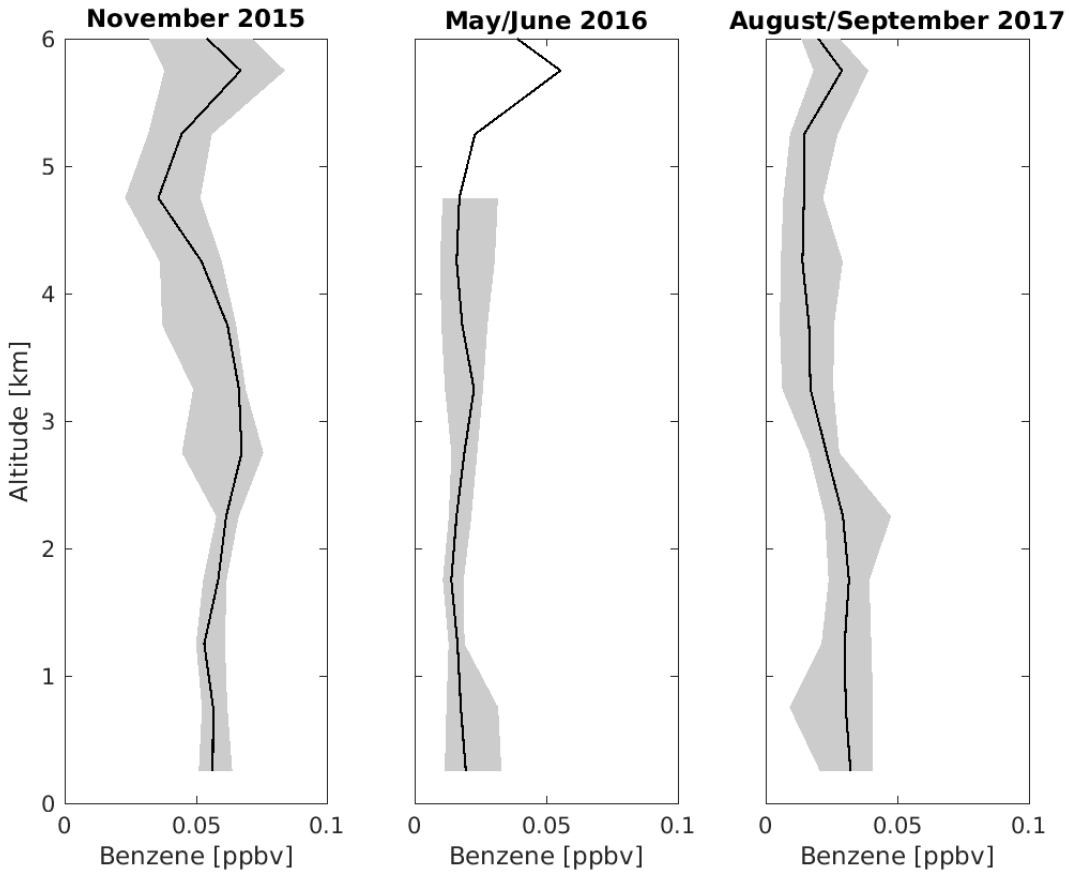

Figure S5: Vertical profiles of NAAMES campaign-median benzene mixing ratios (ppbv) obtained from a Proton-Transfer-Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS) aboard the NASA C130 aircraft (black, with 25^{th}-75^{th} percentiles in grey).

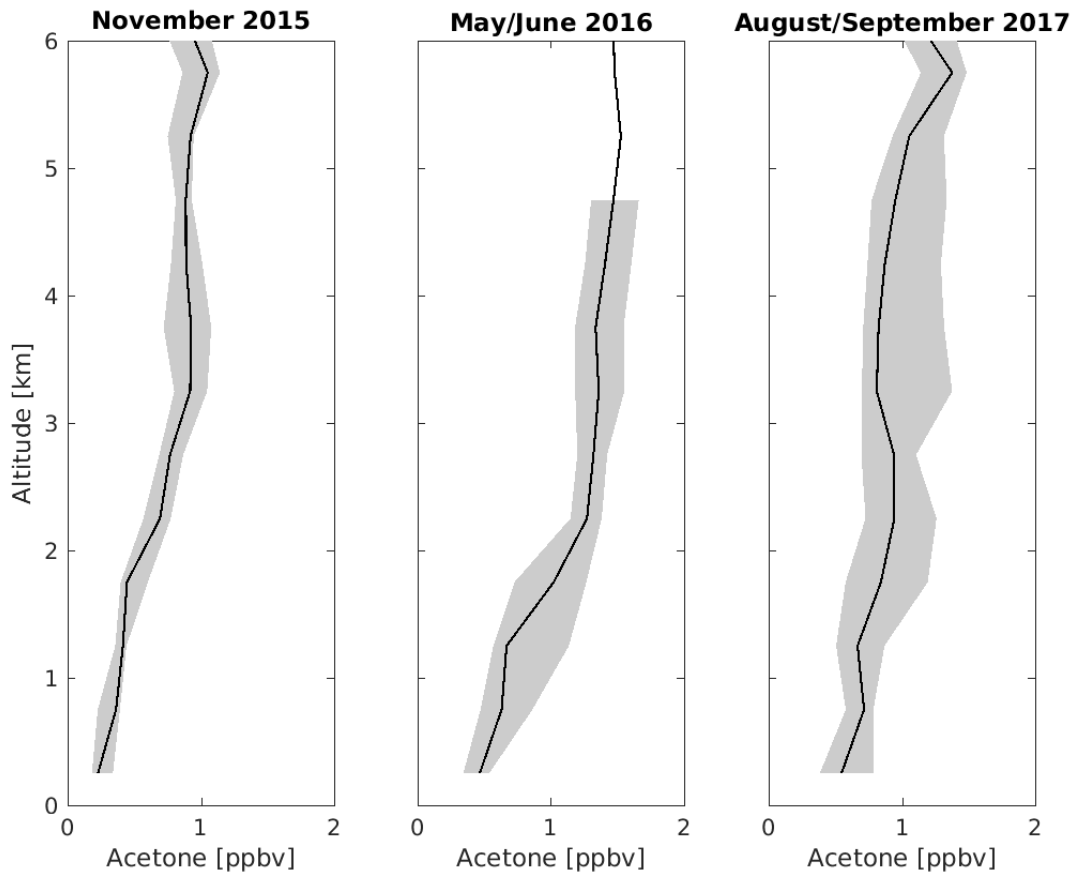

Figure S6: Vertical profiles of NAAMES campaign-median acetone mixing ratios (ppbv) obtained from a Proton-Transfer-Reaction Time-of-Flight Mass Spectrometer (PTR-ToF-MS) aboard the NASA C130 aircraft (black, with 25^{th}-75^{th} percentiles in grey).

Marine organic vapor source	Nov 2015 Bloom Minima	May/June 2016 Bloom Maxima	Aug/Sept 2017 Declining Phase	Mar/Apr 2018 Accumulating	Annual Mean
70T-350 (BASE)	0.17	0.40	0.02	0.31	0.23
50T-300	0.34	0.22	0.22	0.21	0.25
50T-500	0.55	0.20	0.56	0.23	0.38
100T-300	0.11	0.54	0.26	0.33	0.31
100T-500	0.13	0.30	0.19	0.31	0.27

144
145 **Table S1:** Mean fractional error between observations and five GEOS-Chem-TOMAS
146 simulations for the ship-track campaign-median aerosol size distributions shown in Fig. S1. An
147 MFE of 0.50 or less indicates acceptable model-measurement agreement. T in ^{o}C and source flux
148 in $\mu g\ m^{-2}\ d^{-1}$.
149
150
151
152
153