1	Supporting Information
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3	Cloud Drop Number Concentrations over the Western North Atlantic Ocean: Seasonal
4	Cycle, Aerosol Interrelationships, and Other Influential Factors
5	
6	Hossein Dadashazar <sup>1</sup> , David Painemal <sup>2,3</sup> , Majid Alipanah <sup>4</sup> , Michael Brunke <sup>5</sup> , Seethala
7	Chellappan <sup>6</sup> , Andrea F. Corral <sup>1</sup> , Ewan Crosbie <sup>2,3</sup> , Simon Kirschler <sup>7</sup> , Hongyu Liu <sup>8</sup> , Richard
8	Moore <sup>2</sup> , Claire Robinson <sup>2,3</sup> , Amy Jo Scarino <sup>2,3</sup> , Michael Shook <sup>2</sup> , Kenneth Sinclair <sup>9,10</sup> , K. Lee
9	Thornhill <sup>2</sup> , Christiane Voigt <sup>7</sup> , Hailong Wang <sup>11</sup> , Edward Winstead <sup>2,3</sup> , Xubin Zeng <sup>5</sup> , Luke
10	Ziemba <sup>2</sup> , Paquita Zuidema <sup>6</sup> , Armin Sorooshian <sup>1,5</sup>
11	
12	<sup>1</sup> Department of Chemical and Environmental Engineering, University of Arizona, Tucson, AZ,
13	USA
14	<sup>2</sup> NASA Langley Research Center, Hampton, VA, USA
15	<sup>3</sup> Science Systems and Applications, Inc., Hampton, VA, USA
16	<sup>4</sup> Department of Systems and Industrial Engineering, University of Arizona, Tucson, AZ, USA
17	<sup>5</sup> Department of Hydrology and Atmospheric Sciences, University of Arizona, Tucson, AZ, USA
18	<sup>6</sup> Rosenstiel School of Marine and Atmospheric Science, University of Miami, Miami, FL, USA
19	<sup>7</sup> Institute of Atmospheric Physics, German Aerospace Center
20	<sup>8</sup> National Institute of Aerospace, Hampton, VA, USA
21	<sup>9</sup> NASA Goddard Institute for Space Studies, New York, NY, USA
22	<sup>10</sup> Universities Space Research Association, Columbia, MD, USA
23	<sup>11</sup> Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory,
24	Richland, WA, USA
25	
26	<sup>*</sup> Correspondence to: Hossein Dadashazar ( <u>hosseind@arizona.edu</u> )

27 Table S1: Mean seasonal values of speciated AOD and surface mass concentration for black

28 carbon, dust, organic carbon, sulfate, and sea-salt for the six sub-domains in Figures S1-S2.

Speciated AOD/Surface mass concentration	n (µg m¯)	)
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	S	C-S	С	C-N	Ν	Bermuda				
Sulfate										
DJF	0.03/0.91	0.05/1.37	0.05/1.49	0.06/1.99	0.06/1.50	0.03/0.59				
MAM	0.05/0.99	0.06/1.37	0.07/1.54	0.08/2.38	0.07/1.84	0.04/0.72				
JJA	0.05/0.75	0.07/1.10	0.07/1.25	0.09/2.26	0.08/1.45	0.04/0.50				
SON	0.04/0.73	0.04/1.00	0.04/1.08	0.05/1.56	0.05/1.09	0.02/0.41				
Sea-salt										
DJF	0.04/38.10	0.04/39.12	0.05/45.89	0.03/29.55	0.03/27.45	0.05/49.51				
MAM	0.04/36.72	0.04/38.04	0.04/44.28	0.03/28.52	0.02/23.22	0.04/41.76				
JJA	0.05/46.12	0.05/48.83	0.05/56.56	0.03/30.91	0.02/17.99	0.03/35.95				
SON	0.06/48.44	0.05/43.76	0.06/54.24	0.04/36.20	0.03/27.24	0.06/54.60				
Dust										
DJF	< 0.01/0.74	< 0.01/1.06	0.01/1.64	0.01/2.64	0.01/1.24	<0.01/0.98				
MAM	0.02/3.90	0.02/3.86	0.02/4.03	0.03/4.47	0.02/2.90	0.02/2.81				
JJA	0.03/12.05	0.02/8.22	0.02/6.08	0.02/5.02	0.02/2.41	0.02/6.58				
SON	0.01/2.56	0.01/1.79	0.01/2.01	0.01/2.51	0.01/1.20	0.01/2.01				
			Organic car	bon						
DJF	0.01/0.69	0.02/0.92	0.02/0.67	0.02/1.07	0.02/0.58	0.01/0.25				
MAM	0.03/1.00	0.04/1.37	0.04/1.02	0.04/1.53	0.03/0.92	0.03/0.42				
JJA	0.01/0.55	0.03/1.16	0.03/1.25	0.05/2.49	0.05/2.43	0.02/0.35				
SON	0.01/0.67	0.02/1.03	0.02/0.87	0.02/1.43	0.02/0.98	0.01/0.34				
Black carbon										
DJF	< 0.01/0.10	< 0.01/0.15	0.01/0.15	0.01/0.37	0.01/0.20	<0.01/0.08				
MAM	0.01/0.12	0.01/0.16	0.01/0.17	0.01/0.38	0.01/0.21	0.01/0.09				
JJA	< 0.01/0.08	0.01/0.12	0.01/0.15	0.01/0.41	0.01/0.31	<0.01/0.06				
SON	<0.01/0.09	< 0.01/0.12	< 0.01/0.14	0.01/0.32	0.01/0.20	< 0.01/0.06				



- 32
- Figure S1: Seasonal maps of MERRA-2 speciated AOD based on data between January 2013 and December 2017. The boxes in top left panel represent sub-domains examined in
- more detail throughout the study, with the blue star denoting Bermuda.





85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W

- 40
- 41 Figure S3: Seasonal maps of the coefficient of determination (R<sup>2</sup>) and number of points for
- 42 the analysis of the aerosol-cloud interaction (ACI) parameters over the WNAO using daily
- 43 N<sub>d</sub> and four different aerosol proxy parameter values (AI, AOD, Sulfate<sub>AOD</sub>, Sulfate<sub>sf-mass</sub>)
- 44 from CERES-MODIS and MERRA-2, respectively. ACI statistics associated with the six
- 45 sub-domains shown are summarized in Table 4.
- 46





- 48 Figure S4: Seasonal climatology of N<sub>d</sub> (middle column) and anomalies from seasonal
- 49 averages for low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The red box
- 50 represents sub-domain C-N for which the analysis was conducted, as explained in Section
- 51 4.1.





- 54 seasonal averages for low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The
- 55 reference wind vector is shown on the top left panel. The red box represents sub-domain C-
- 56 N for which the analysis was conducted.



58 Figure S6: Seasonal averages of low-level liquid cloud-top effective height (middle column)

- and associated anomalies on low- $N_d$  days (left column) and high- $N_d$  days (right column). The
- 60 red box represents sub-domain C-N for which the analysis was conducted.



62 Figure S7: Seasonal averages of PERSIANN-CDR precipitation rate (middle column) and

- associated anomalies on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The
- red box represents sub-domain C-N for which the analysis was conducted.





Figure S8: Seasonal averages of planetary boundary layer height (middle column) and

- associated anomalies on low-Nd days (left column) and high-Nd days (right column). The
- red box represents sub-domain C-N for which the analysis was conducted.





73 Figure S9: Seasonal averages of vertical pressure velocity at 800 hPa (middle column) and

- 74 associated anomalies on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The 75 and her moments such domain C N for which the analysis may conducted
- 75 red box represents sub-domain C-N for which the analysis was conducted.
- 76





associated anomalies on low-Nd days (left column) and high-Nd days (right column). The red box represents sub-domain C-N for which the analysis was conducted.





Figure S11: Seasonal averages of relative humidity at 800 hPa (middle column) and

85 associated anomalies on low-Nd days (left column) and high-Nd days (right column). The 86 red box represents sub-domain C-N for which the analysis was conducted.





Figure S12: Seasonal averages of sulfate AOD (middle column) and associated anomalies

 $r_{1}$  on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The red box represents sub-

- 90 domain C-N for which the analysis was conducted.
- 91





65 W 60 W 75 W 70 W 65 W 60 W 85 W 80 W 75 W 70 W 65 W 60 W 85 W 80 W 75 W 70 W 65 W 60 W

Figure S13: Seasonal averages of sea-salt AOD (middle column) and associated anomalies
on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The red box represents sub domain C-N for which the analysis was conducted.





85 W 80 W 75 W 70 W 85 W 80 W

98 Figure S14: Seasonal averages of dust AOD (middle column) and associated anomalies on

99 low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The red box represents sub-

100 domain C-N for which the analysis was conducted.





Figure S15: Seasonal averages of organic carbon AOD (middle column) and associated anomalies on low-Nd days (left column) and high-Nd days (right column). The red box represents sub-domain C-N for which the analysis was conducted. 





05 W 00 W 75 W 70 W 05 W 00 W 85 W 80 W 75 W 70 W 85 W 80 W 75 W 70 W 85 W 80

108Figure S16: Seasonal averages of black carbon AOD (middle column) and associated109anomalies on low-Nd days (left column) and high-Nd days (right column). The red box

110 represents sub-domain C-N for which the analysis was conducted.





. 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W 85<sup>°</sup>W 80<sup>°</sup>W 75<sup>°</sup>W 70<sup>°</sup>W 65<sup>°</sup>W 60<sup>°</sup>W

- Figure S17: Seasonal averages of sulfate surface mass concentration (middle column) and 113
- associated anomalies on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The 114
- red box represents sub-domain C-N for which the analysis was conducted. 115
- 116





118 Figure S18: Seasonal averages of sea-salt surface mass concentration (middle column) and

- associated anomalies on low- $N_d$  days (left column) and high- $N_d$  days (right column). The
- 120 red box represents sub-domain C-N for which the analysis was conducted.





. 85°W 80°W 75°W 70°W 65°W 60°W 85°W 80°W 75°W 70°W 65°W 60°W 85°W 80°W 75°W 70°W 65°W 60°W

Figure S19: Seasonal averages of dust surface mass concentration (middle column) and 122

- associated anomalies on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right column). The 123
- red box represents sub-domain C-N for which the analysis was conducted. 124
- 125



Figure S20: Seasonal averages of organic carbon (OC) surface mass concentration (middle
column) and associated anomalies on low-N<sub>d</sub> days (left column) and high-N<sub>d</sub> days (right
column). The red box represents sub-domain C-N for which the analysis was conducted.



Figure S21: Seasonal averages of black carbon (BC) surface mass concentration (middle column) and associated anomalies on low- $N_d$  days (left column) and high- $N_d$  days (right column). The red box represents sub-domain C-N for which the analysis was conducted.

- 135
- 136





138 Figure S22: Average local accumulated effect (ALE) profiles based on GBRT modeling for 139 surface mass concentrations of the following parameters: a) relative humidity at 950 hPa (RH<sub>950</sub>), (b) relative humidity at 800 hPa (RH<sub>800</sub>), (c) rain rate, and (d) CAO index. Blue and 140 141 red profiles represent ALEs of DJF and JJA, respectively. Shaded areas show the ALE 142 ranges stemming from the variability of the obtained models from the cross-validation resampling procedure. Markers on the bottom and top x-axes denote the values of 5<sup>th</sup>, 25<sup>th</sup>, 143 144 50<sup>th</sup>, 75<sup>th</sup>, and 95<sup>th</sup> percentiles for each input variable; note that the first three markers on 145 the x-axes in panel (c) are very close and thus on top of each other.