



Supplement of

North Atlantic Ocean SST-gradient-driven variations in aerosol and cloud evolution along Lagrangian cold-air outbreak trajectories

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Table S1. The number of measurements used for statistical calculations comparing upwind (C130) and downwind (C130 or *R/V Atlantis*) measurements in Table 3. Measurement counts are included for both upwind and downwind measurements (N_{upwind} , $N_{downwind}$). Ship measurements were averaged over 1 hour, and the ship based AMS and $CN_{>100nm}$ (SMPS) measurements were collected at five minutes intervals, resulting in the different measurement counts between ship and C130 measurements.

		Particle Composition (µg m ⁻³)		Particle Cor (cm	Particle Concentration (cm ⁻³)	
Figure Panel and upwind cloud conditions	Dt (hours)	ΔOrg	ΔSO_4	$\Delta N_{>10nm}^4$	ΔCN _{>100nm}	
Figure 6	Trajectories initialized at the R/V Atlantis					
a _{open-cell}	8	(12,12)	(12,12)	(3600,330)	(12,330)	
b _{open-cell}	11	(12,18)	(12,18)	(3600,494)	(12,494)	
C _{open-cell}	14	(12,15)	(12,15)	(3600,419)	(12,419)	
d _{open-cell}	16	(12,15)	(12,15)	(3600,399)	(12,399)	
emixture ¹	30	(12,27)	(12,27)	(3600,760)	(12,760)	
Figure 7		Trajectories Initialized at the C130				
b _{closed-cell} ²	48	(17, 9)	(17, 9)	(488, 251)	(488, 251)	
$b_{cloud-free}{}^3$	48	(16,14)	(16,14)	(410,397)	(410,397)	

¹Upwind measurements are near the transition between cloud-free and closed-cell regions and therefore, downwind measurements possibly represent a mixture of the two regimes.

²Difference between the measurements in the FLT17 closed-cell region and FLT19 low particle concentration region. Specifically, FLT17 measurements in the closed-cell region at 53.5°W (Figures 3, 7b) are compared to downwind FLT19 C130 surface measurements between 37.8°W – 38.5°W (Figures 4, 7b).

³Difference between the measurements in the FLT17 cloud-free region and FLT19 high particle concentration region. Specifically, FLT17 measurements in the cloud-free region at 57.8°W (Figures 3, 7b) are compared to downwind FLT19 C130 surface measurements east of 37.8°W (Figures 4, 7b).

⁴*R/V Atlantis* N_{>13nm} measurements are used for comparison to C130 N_{>10nm} measurements.



Figure S1. (top) 18 September 1945z (bottom) 19 September 1015z. Cyan and red lines represent the flight track at altitudes > 3km and < 3km, respectively, ± 1 hour from the satellite image time. The yellow line represents the ship position ± 1 hour from the satellite image time. The gray line is the entire ship track for the NAAMES3 campaign.



Figure S2. The (a) C130 altitude, (b) surface DMS, (c) carbon monoxide and (d) black carbon for 17 September. All measurements are presented as a function of longitude, consistent with Figure 3 measurements.



Figure S3. The (a) C130 altitude, (b) surface DMS, (c) carbon monoxide and (c) black carbon for 19 September. All measurements are presented as a function of longitude, consistent with Figure 4 measurements.



Figure S4. 3-Day FLEXPART back trajectories initialized at the three western most C130 near-surface legs (red) in a closed-cell region (black), cloud-free region (blue) and the transition zone (green) on September 17.



Figure S5. Vertical profiles of the $CN_{>10nm}$ concentration from C130 flights that occurred on (a) 17 September 2019 and (b) 19 September 2019 (Figures 3a and 4a). The same vertical profiles are used to show potential temperature in Figure 5. The point color represents the longitude at which the measurement was made. Air mass transitions from opened-cell to closed cell clouds and from closed-cell to cloud-free air on the 17 September 2019 flight occurred at approximately 52°W and 56°W, respectively. None of the

vertical profiles are in the closed-cell region for the 19 September 2019 flight. In-cloud measurements are excluded and a 30-second running averages is applied to improve readability.



Figure S6. OSCAR satellite derived 1° resolution sea-surface current vectors averaged for September 2017.



Figure S7. MERRA-2 hourly horizontal wind direction and speed by month from 2007-2020 at 56°N, 54°W, which coincides with a point on the transition line between the closed-cell and cloud-free regions during NAAMES3.



Figure S8. September 2017 averaged satellite chlorophyll-a. The boxed area represents the area analyzed in Figure 8.



Figure S9. Monthly satellite derived cloud fraction and chlorophyll-a averaged over 2008-2019.