

# Supporting Information for "Dilution of boundary layer cloud condensation nucleus concentrations by free tropospheric entrainment during marine cold air outbreaks"

F. Tornow<sup>1,2</sup>, A. S. Ackerman<sup>2</sup>, A. M. Fridlind<sup>2</sup>, B. Cairns<sup>2</sup>, E. C. Crosbie<sup>3,4</sup>,

S. Kirschler<sup>5,6</sup>, R. H. Moore<sup>3</sup>, C. E. Robinson<sup>3,4</sup>, C. Seethala<sup>7</sup>, M. A. Shook<sup>3</sup>,

C. Voigt<sup>5,6</sup>, E. L. Winstead<sup>3,4</sup>, L. D. Ziembra<sup>3</sup>, P. Zuidema<sup>7</sup>, A. Sorooshian<sup>8,9</sup>

<sup>1</sup>Earth Institute, Columbia University, NY, 10025, NY

<sup>2</sup>NASA Goddard Institute for Space Studies, NY, 10025, NY

<sup>3</sup>NASA Langley Research Center, Hampton, VA 23681, USA

<sup>4</sup>Science, Systems, and Applications, Inc., Hampton, VA 23681, USA

<sup>5</sup>Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, Germany

<sup>6</sup>Johannes Gutenberg-Universität, Mainz, Germany

<sup>7</sup>Rosenstiel School of Marine and Atmosphere Science, University of Miami, FL, 33149, USA

<sup>8</sup>Department of Chemical and Environmental Engineering, University of Arizona, Tucson, Arizona, 85721, USA

<sup>9</sup>Department of Hydrology and Atmospheric Sciences, University of Arizona, Tucson, Arizona, 85721, USA

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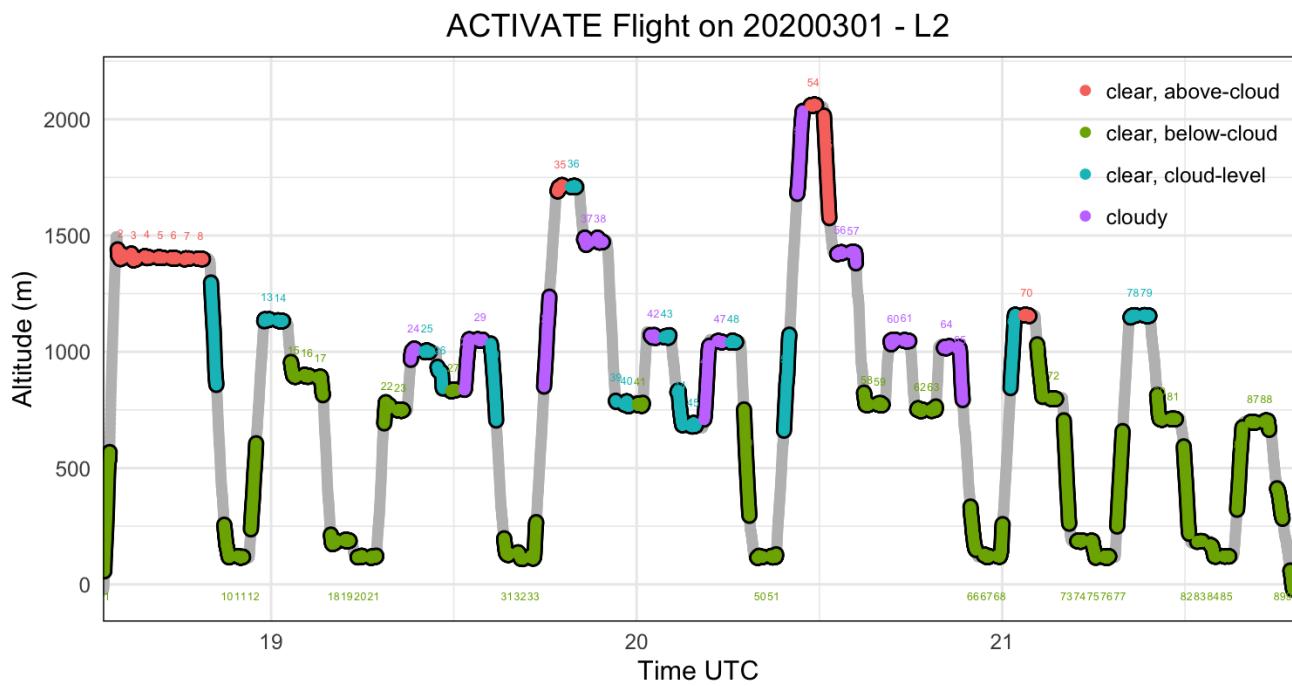
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**Table S1.** 2020 ACTIVATE CAO research flights, the prevalent MBL wind direction, coordinates defining the initial cloud edge, and instrument limitations relevant to this study (see text).

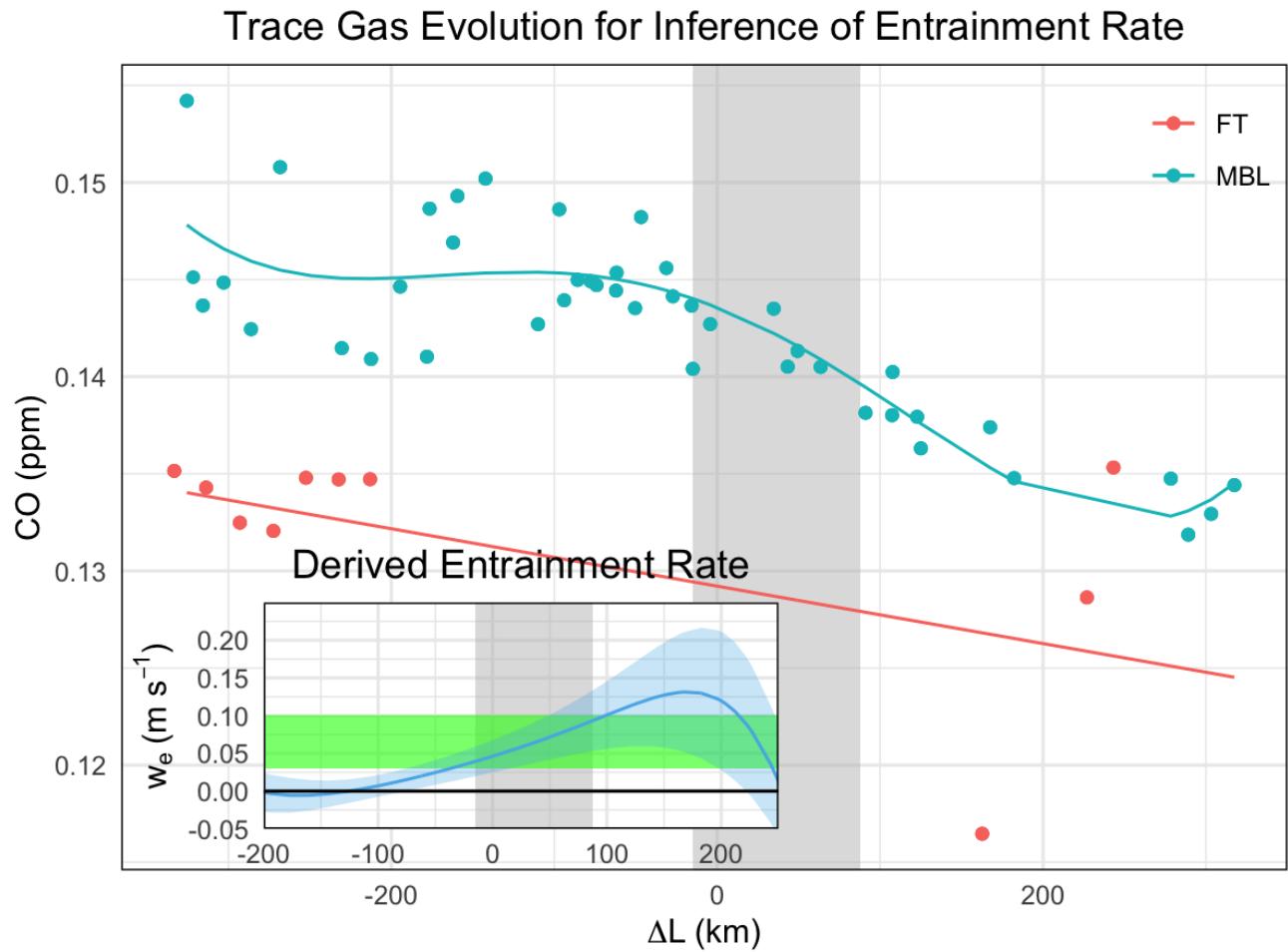
Date / Leg	#	Wind Dir.	Cloud edge coordinates	Instrument Limitations
2020-02-21 / 1	RF04	20°	38.0°N 76.4°W – 39.5°N 72.0°W	Falcon only
2020-02-22 / 1	RF05	25°	34.0°N 77.4°W – 38.0°N 71.5°W	Falcon only
2020-02-22 / 2	RF06	25°	34.0°N 77.4°W – 38.0°N 71.5°W	Falcon only
2020-02-27 / 1	RF09	300°	34.0°N 76.0°W – 38.0°N 73.0°W	/
2020-03-01 / 1	RF13	315°	35.0°N 75.0°W – 40.0°N 72.0°W	/
2020-03-01 / 2	RF14	315°	35.0°N 74.0°W – 40.0°N 72.0°W	/
2020-03-08 / 1	RF17	10°	33.0°N 77.0°W – 36.5°N 72.0°W	No RSP
2020-03-08 / 2	RF18	20°	34.5°N 78.0°W – 34.5°N 70.0°W	No RSP

**Table S2.** Instruments, products, and estimated uncertainty used.

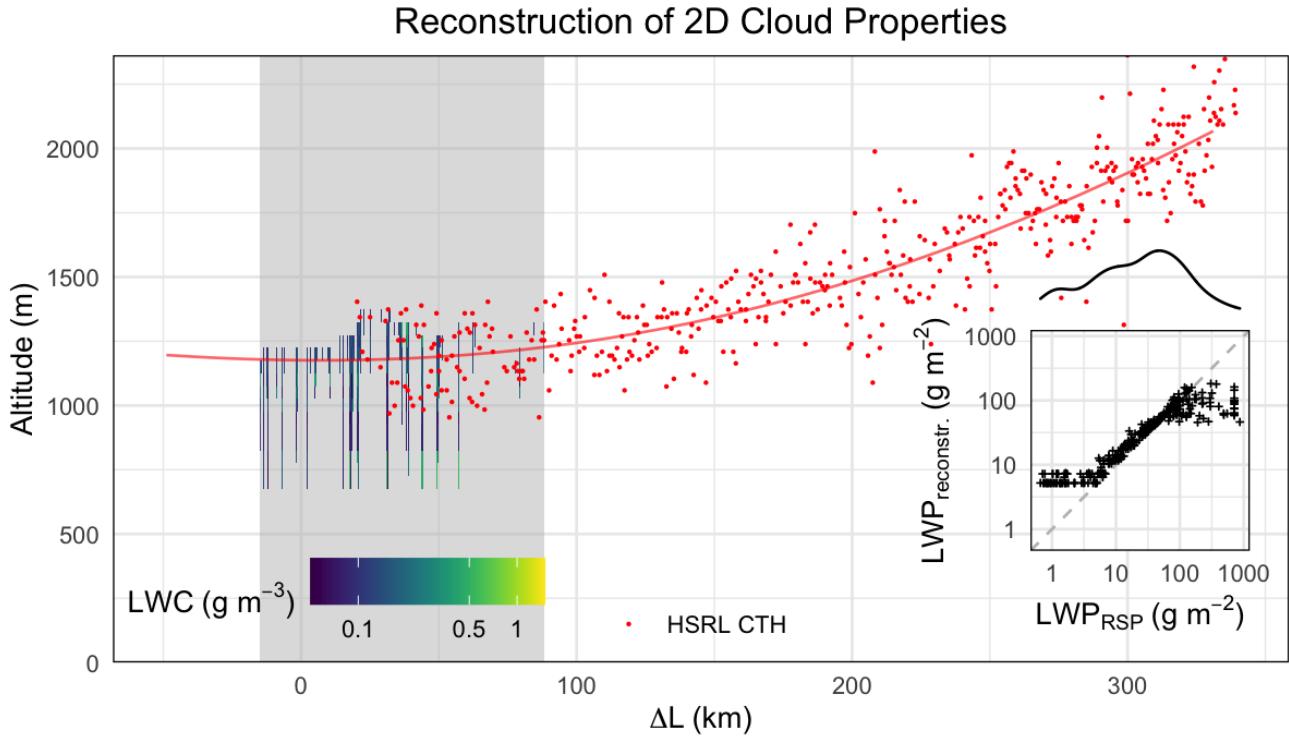
<b>Instrument (in-situ)</b>	<b>Used Products</b>	<b>Uncertainty</b>	<b>Reference</b>
DMT CCN Counter	CCN(s), for either SS=0.43% or SS ∈ [0.2,0.7%]	ΔSS=0.04, δCCN=10%	Lance, Nenes, Medina, and Smith (2006)
TSI CPC-3772	CN-10nm dNa/dlogD for D ∈ [0.1,3.1 um]	10% 20%	/ Froyd et al. (2019)
TSI LAS	dNa/dlogD for D ∈ [0.003,0.089 um]	20%	Moore et al. (2017)
SMPs	Mass conc. for D ∈ [0.05,4.00 um]	/	Sorooshian et al. (2006)
PILS	Mass conc. for D ∈ [0.06,0.60 um]	<50%	DeCarlo et al. (2008)
AMS	dNd/dlogD for D ∈ [3.0,50 um], LWP	/	Knop, Bansmer, Hahn, and Voigt (2021)
SPEC FCDDP	dNd/dlogD for D ∈ [30,1460 um], LWP, IWC	/	Lawson et al. (2006) Kleine et al. (2018) Taylor et al. (2019)
SPEC 2DS	CO gas concentration	2%	/
<b>PICARRO G2401-m</b>			
<b>Instrument (remote)</b>			
HSRL-2	Cloud-top height	15%	Burton et al. (2018)
RSP	Cloud optical thickness, Droplet effective radius		Cairns, Russell, and Travis (1999)
Dropsonde	Temperature, Pressure	0.2 K and 0.05 hPa	Hock and Franklin (1999)



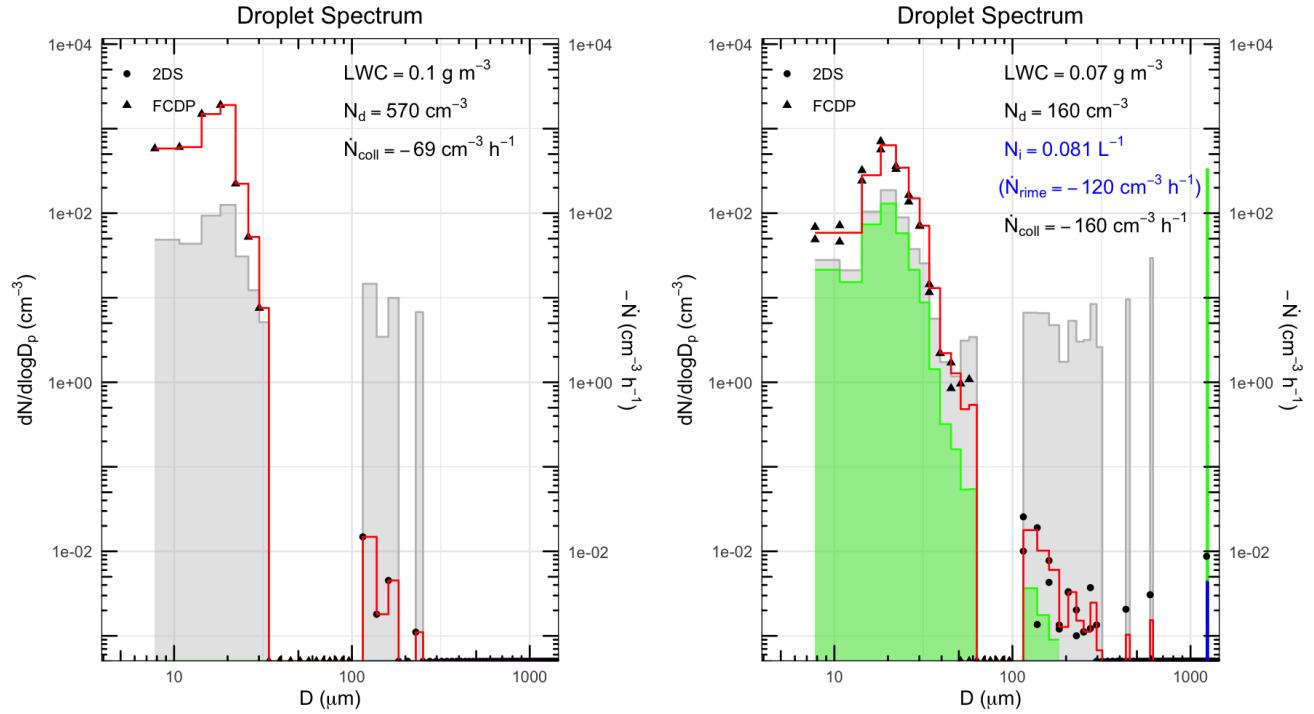
**Figure S1.** Categorization of CCN measurements during RF14 on 1 March 2020 as defined in Section 2.1.



**Figure S2.** CO trace gas measurements during RF14 on 1 March 2020 as a function of distance from cloud edge ( $\Delta L$ ) sorted into altitudes relative to the cloud deck (see legend). Inset: entrainment rates derived from mixed-layer framework (blue) with shaded uncertainties (plus/minus one sigma), and the range found in large-eddy simulations of a similar case (green shading; Tornow et al., 2021). Gray shading indicates distance range of budget analysis.



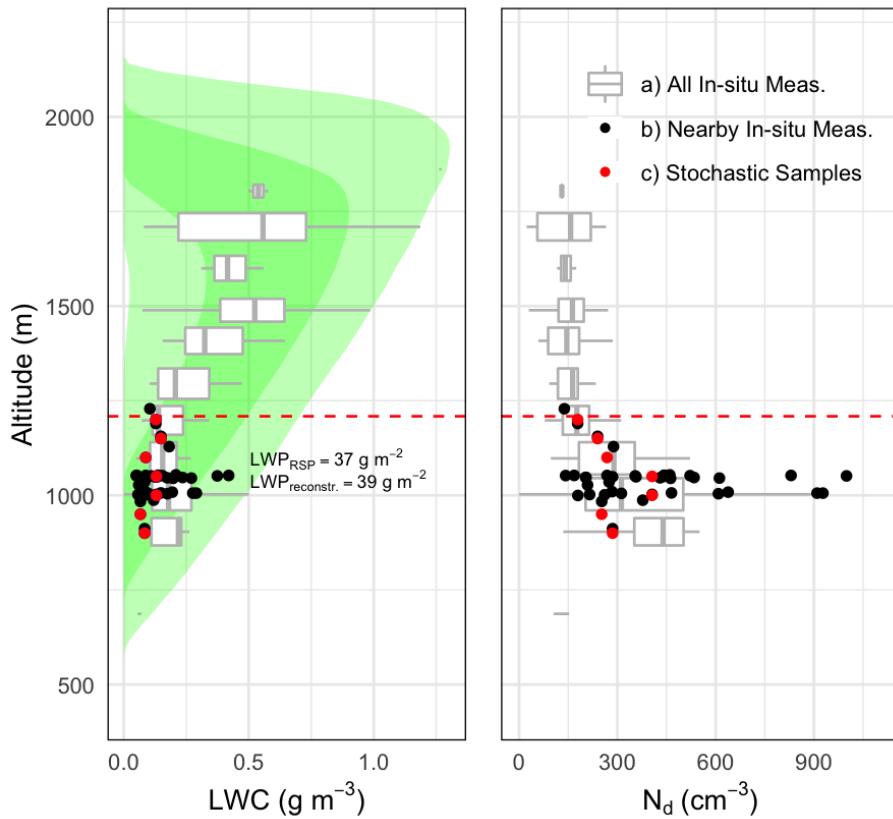
**Figure S3.** Overview of RF14 (1 March 2020) mock-cloud-profiles (LWC shown as colored shading) together with HSRL-2 cloud-top heights (red). The inset compares LWP from reconstructed profiles with the RSP-based LWP values. The curve above the inset indicates the probability density function for RSP-based values.



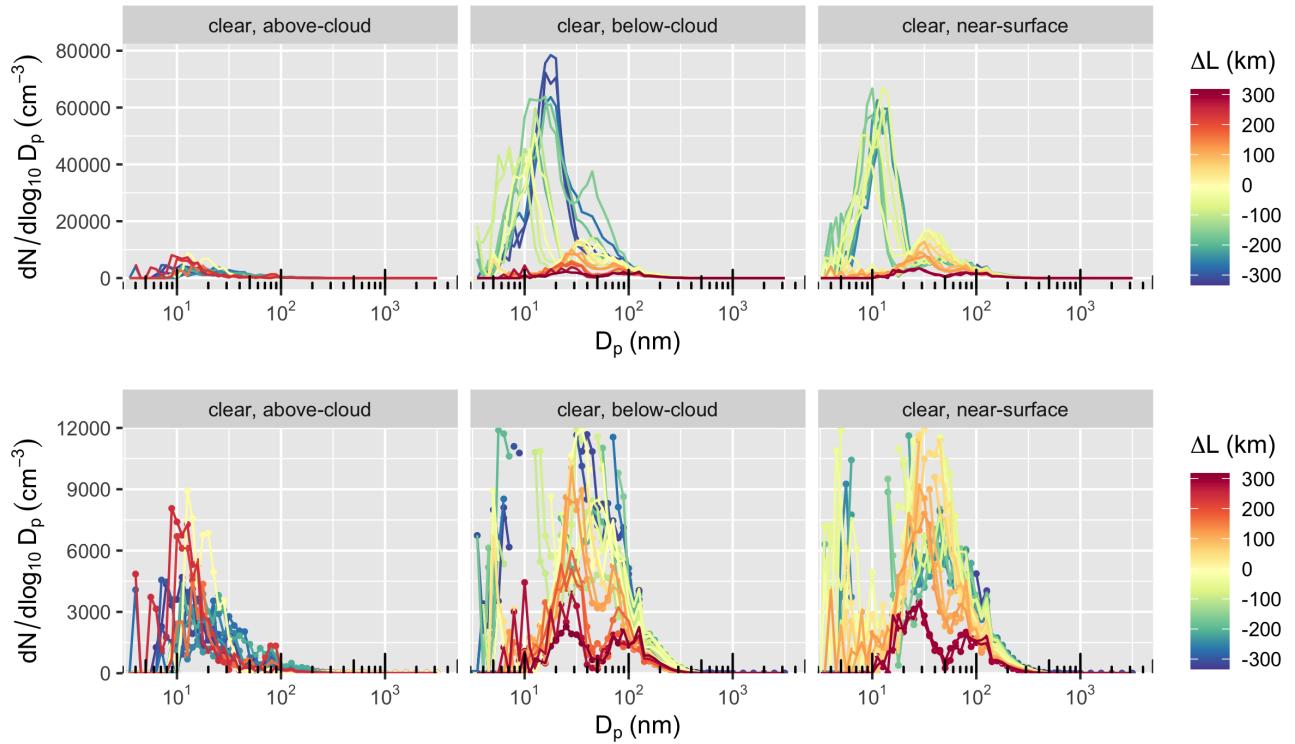
**Figure S4.** Example hydrometeor size distributions (red, scale on left axes) during RF14 at flight time 70495 s (left) and at 73525 s (right) and corresponding computed collision loss rates (listed at top-right corner) with bin-wise contributions (gray shade, scale on right axes). Rates that involved hydrometeors classified as frozen (only in one bin, shown with blue bar) are labelled as “riming” (shown as integral in blue text and as bin-wise contribution through green shading).

### Use of Nearby\* In-Situ Samples for Reconstruction

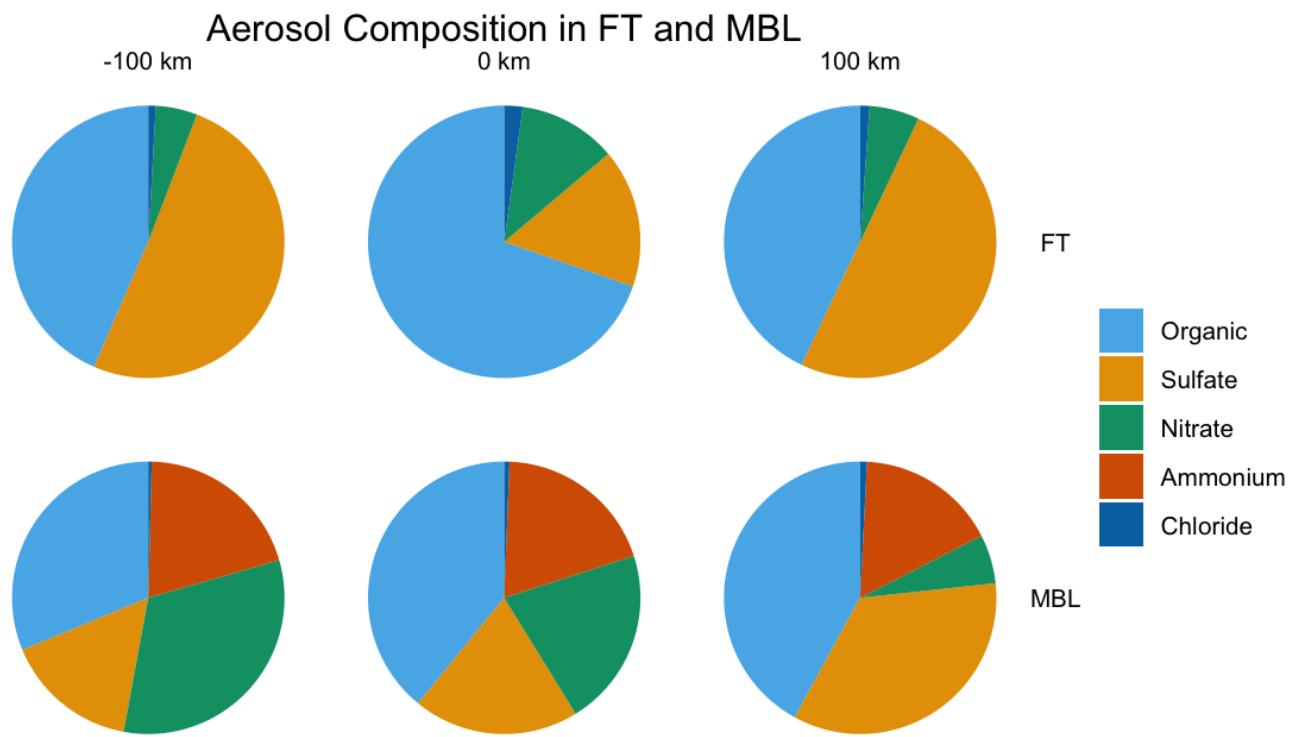
(\* within 100 km horiz. distance; within 900 s of RSP meas.; within 50 m vertically)



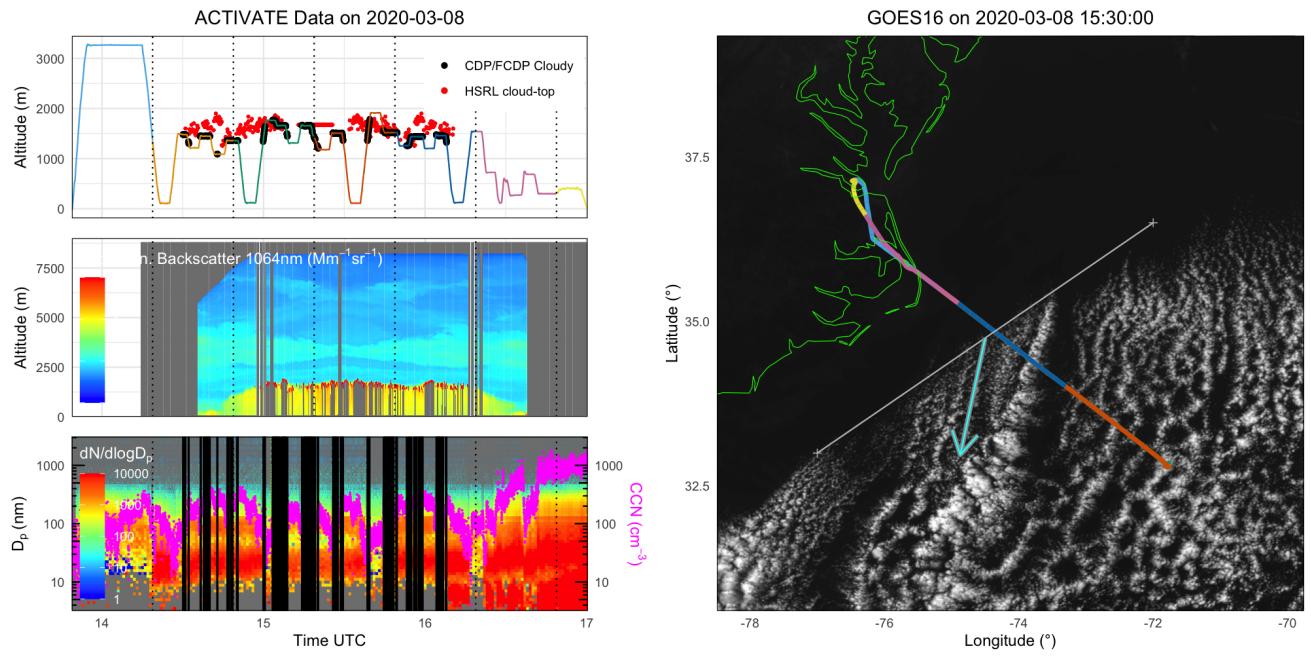
**Figure S5.** Example of RF14 (1 March 2020) in-situ samples (black) to stochastically build a mock-cloud-profile (red), shown for LWC (left) and  $N_d$  (right), until the LWP roughly matches the nearby RSP-sampled value. Gray bars mark the range of all in-situ observations (box ranging between 25<sup>th</sup> and 75<sup>th</sup> percentiles and whiskers extending to 5<sup>th</sup> and 95<sup>th</sup> percentiles). The green shading (lighter shade marks 5<sup>th</sup> to 95<sup>th</sup> and darker shade 25<sup>th</sup> to 75<sup>th</sup> percentiles) shows LWC profiles from large-eddy simulations of a similar case (altitudes shifted 500 m downward). The decrease of  $N_d$  with height is an artifact of MBL deepening downwind where  $N_d$  progressively decreases.



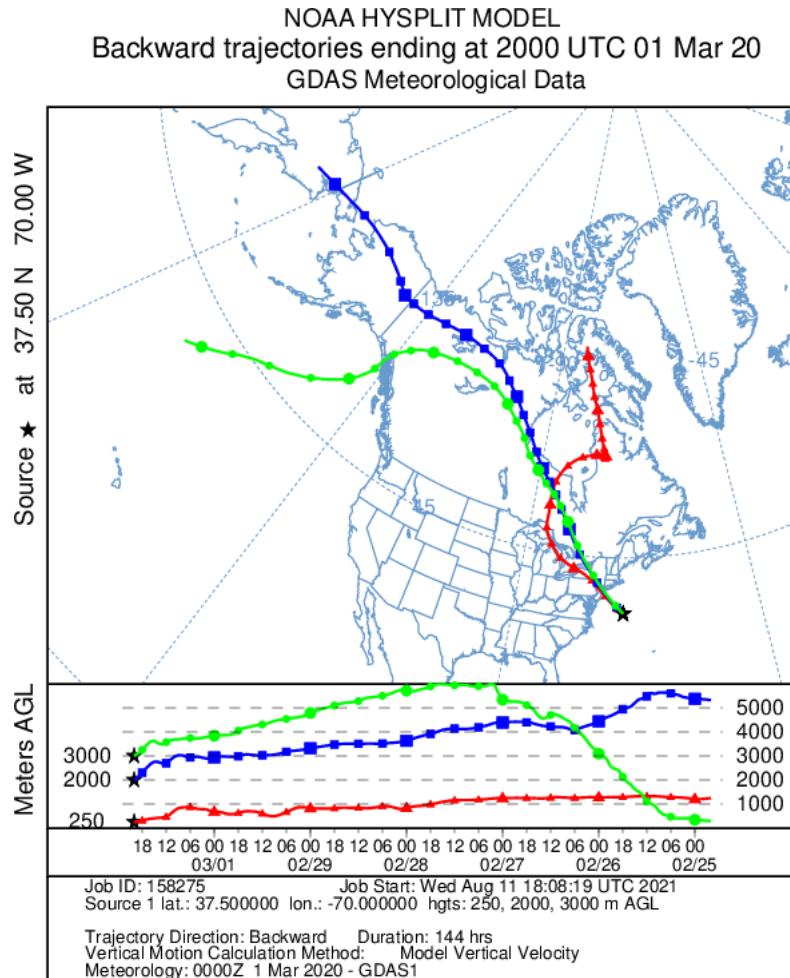
**Figure S6.** Aerosol particle size distributions measured during RF14 (1 March 2020) in the FT and MBL (top; and with reduced y-axis range, bottom). Colors mark the downwind distance from cloud edge,  $\Delta L$ .



**Figure S7.** Aerodyne High-Resolution Mass Spectrometer (AMS) measurements during RF14 (1 March 2020) for the approximate size range 60-600 nm, showing mass proportions in FT (top) and MBL (bottom) air masses and were interpolated to three selected  $\Delta L$  values (horizontal position).



**Figure S8.** As in Figure 1, but for the first research flight on 8 March 2020 (RF17).



**Figure S9.** Back-trajectories based on HYSPLIT (Stein et al., 2015; Rolph et al., 2017) for RF14.