Table S1: Two-mode, lognormal model results for aerosol number concentration (N) for different bins of distance relative to LaRC. N_1 represents the mode corresponding to the smaller $D_{p,g}$ while N_2 corresponds to the larger $D_{p,g}$. The bolded values represent the measurements made within the smoke layer, which correspond to the FT measurements made on 22 March 2022 in the $550 - 800$ km range. Blank cells indicate unavailable data or when the data can be better represented by a single mode.

Boundary Layer

Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
		N_1 [cm^{-3}]	N_2 [cm^{-3}]	N_1 [cm^{-3}]	N_2 [cm^{-3}]	N_1 [cm^{-3}]	N_2 [cm^{-3}]	N_1 [cm^{-3}]	N_2 [cm^{-3}]
22 March 2022	142	6980.9	6005.5	5367.5	6661.1	4083.6	3414.3	2312.1	1155.2
	143	1469.7	12155.5	2846.9	8121.2	3186.2	4589.1	2282.6	1234.3
18 May 2022	156	5941.9	1790.4	2350.2	3659.6	1548.5	2668.6	1476.5	491.9
	157	5613.6	1466.1	4483.8	2027.0	1389.1	2995.5	1931.4	795.0
21 May 2022	159	4246.9	355.8	2157.1	1119.7	1765.8	968.1	1332.2	837.1
	160	864.7	4256.3	1285.2	562.9	1178.6	517.6	1027.0	562.6
31 May 2022	161	341.1	4005.5	261.3	3170.6	623.5	3172.0	853.5	2506.4
18 June 2022	179	7380.9	3645.1	2943.7	2746.8	882.8	538.3	832.3	324.4

Free Troposphere

Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
22 March 2022	142	2272.1	10005.2	2065.6	9932.0	1866.2	4681.1	1040.3	3488.9
	143	11457.9		1386.9	7239.2	**4118.5**	**2711.0**	2489.1	4157.7
18 May 2022	156	3819.2	4425.2	5308.5	1240.9	3054.6	2454.0	2131.1	503.5
	157	6567.1	4754.9	5989.1	2475.2	4772.4	1161.7	1273.1	952.1
21 May 2022	159	5149.1	1281.6	2923.4	772.5	3379.5	874.6	224.8	4154.9
	160	4326.1	1287.0	1592.8	1149.2	2240.2	228.0	1925.2	529.5
31 May 2022	161	1264.8	3571.9	243.7	3567.2	1205.3	162.6	260.9	2728.3
18 June 2022	179					1908.0	2027.6	1211.8	311.1

Table S2: Two-mode, lognormal model results for geometric mean diameter ($D_{p,g}$) for different bins of distance relative to LaRC. $D_{p,g,1}$ is smaller than $D_{p,g,2}$. The bolded values represent the measurements made within the smoke layer, which correspond to the FT measurements made on 22 March 2022 in the $550 - 800$ km range. Blank cells indicate unavailable data or when the data can be better represented by a single mode.

Boundary Layer

Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
		$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]
22 March 2022	142	39.34	89.88	33.33	76.71	25.56	82.95	25.08	102.94
	143	60.53	81.17	41.79	74.42	29.75	68.04	25.07	103.88
18 May 2022	156	39.70	119.18	38.63	109.72	42.10	110.29	61.69	188.53
	157	46.56	132.17	41.39	114.43	38.72	102.60	57.81	176.39
21 May 2022	159	82.95	228.82	78.12	208.64	80.43	208.48	71.24	198.25
	160	75.13	87.35	55.80	184.25	54.70	183.67	59.82	186.15
31 May 2022	161	37.09	106.94	40.90	97.94	42.08	99.96	45.11	107.20
18 June 2022	179	53.88	118.30	48.97	126.17	43.44	154.91	40.44	168.78

Free Troposphere

Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
		$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]	$D_{p,g,1}$ [nm]	$D_{p,g,2}$ [nm]
22 March 2022	142	31.09	64.22	29.08	64.16	33.54	113.50	29.31	98.99
	143	66.99		28.34	69.30	**95.71**	**207.68**	36.02	118.08
18 May 2022	156	35.22	131.11	69.84	143.16	42.68	119.01	79.89	200.47
	157	53.87	56.97	46.16	93.40	75.81	158.95	61.59	175.73
21 May 2022	159	77.76	217.71	87.56	209.76	97.44	230.23	74.23	143.82
	160	70.39	198.25	81.63	169.62	102.12	191.31	107.08	211.02
31 May 2022	161	76.56	165.07	38.50	108.69	51.37	109.49	34.62	111.32
18 June 2022	179					48.90	144.12	33.51	107.44

Table S3: Two-mode, lognormal model results for the geometric standard deviation parameter (σ_g) for different bins of distance relative to LaRC. $\sigma_{g,1}$ represents the mode corresponding to the smaller $D_{p,g}$ while $\sigma_{g,2}$ corresponds to the larger $D_{p,g}$. The bolded values represent the measurements made within the smoke layer, which correspond to the FT measurements made on 22 March 2022 in the $550 - 800$ km range. Blank cells indicate unavailable data or when the data can be better represented by a single mode.

Boundary Layer

Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
		$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$
22 March 2022	142	1.47	1.64	1.42	1.78	1.55	1.71	1.86	1.28
	143	1.24	1.78	1.34	1.75	1.44	1.90	1.68	1.31
18 May 2022	156	1.49	1.37	1.47	1.52	1.55	1.49	1.76	1.31
	157	1.47	1.33	1.38	1.44	1.46	1.53	1.63	1.38
21 May 2022	159	2.01	1.23	1.68	1.34	1.73	1.35	1.54	1.31
	160	1.36	2.14	1.60	1.34	1.49	1.28	1.49	1.30
31 May 2022	161	1.31	1.75	1.25	1.76	1.26	1.72	1.39	1.67
18 June 2022	179	1.52	1.48	1.46	1.33	1.42	1.40	1.34	1.40

Free Troposphere

| Date	RF	< 300 km		300 − 550 km		550 − 800 km		800 − 1100 km	
		$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$	$\sigma_{g,1}$	$\sigma_{g,2}$
22 March 2022	142	1.29	1.82	1.23	1.96	1.43	1.69	1.52	1.60
	143	1.91		1.22	1.86	**1.57**	**1.38**	1.53	1.63
18 May 2022	156	1.48	1.53	1.91	1.31	1.58	1.50	1.82	1.29
	157	2.22	1.29	1.34	1.43	1.92	1.27	1.60	1.43
21 May 2022	159	1.84	1.43	1.91	1.30	1.84	1.34	1.06	1.66
	160	1.47	1.43	1.87	1.52	1.77	1.25	1.80	1.31
31 May 2022	161	1.28	1.49	1.21	1.77	2.27	1.23	1.30	1.71
18 June 2022	179					1.42	1.55	1.77	1.33
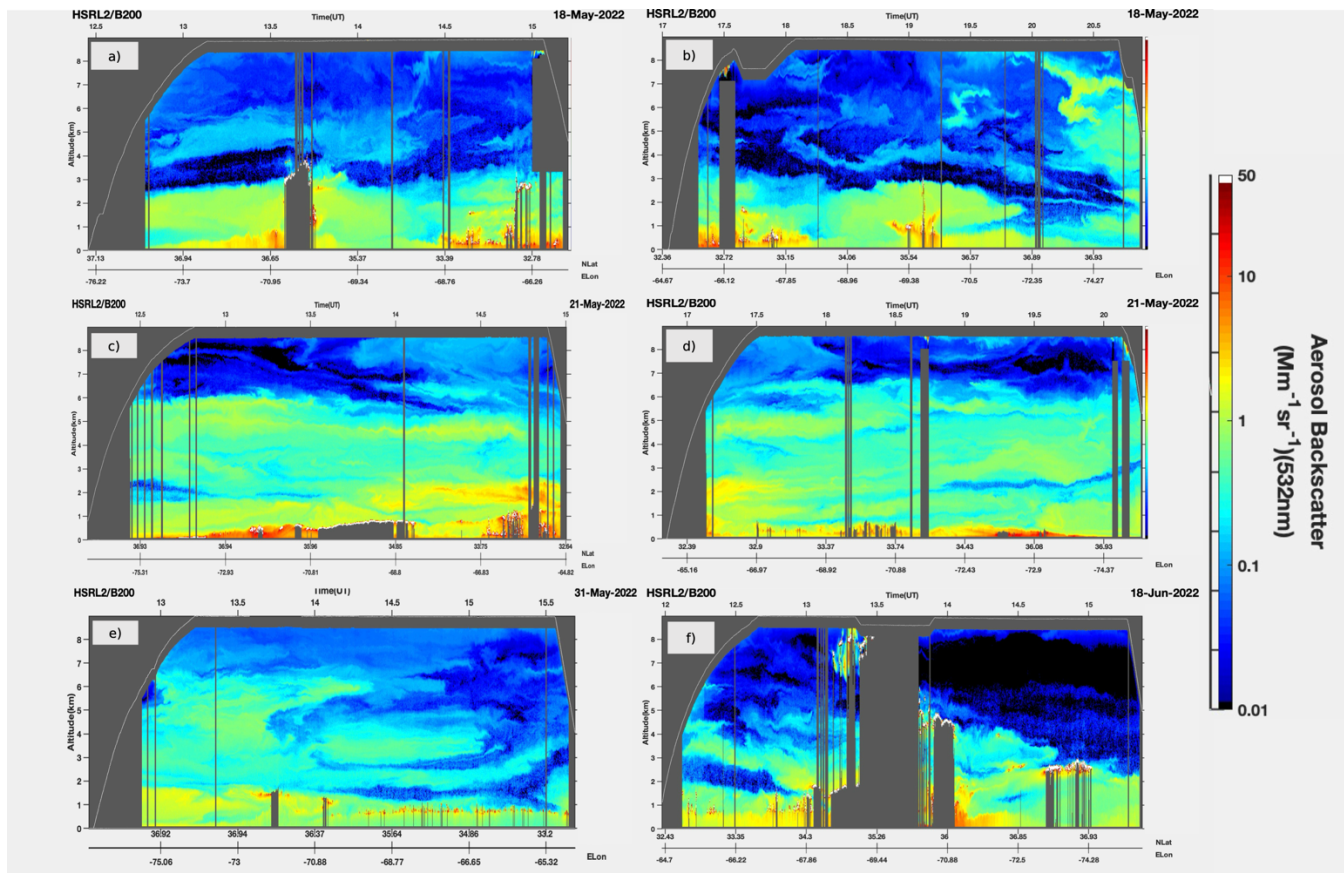
Figure S1: Aerosol backscatter (532 nm) for RFs 156 (a), 157 (b), 159 (c), 160 (d), 161 (e), and 179 (f).
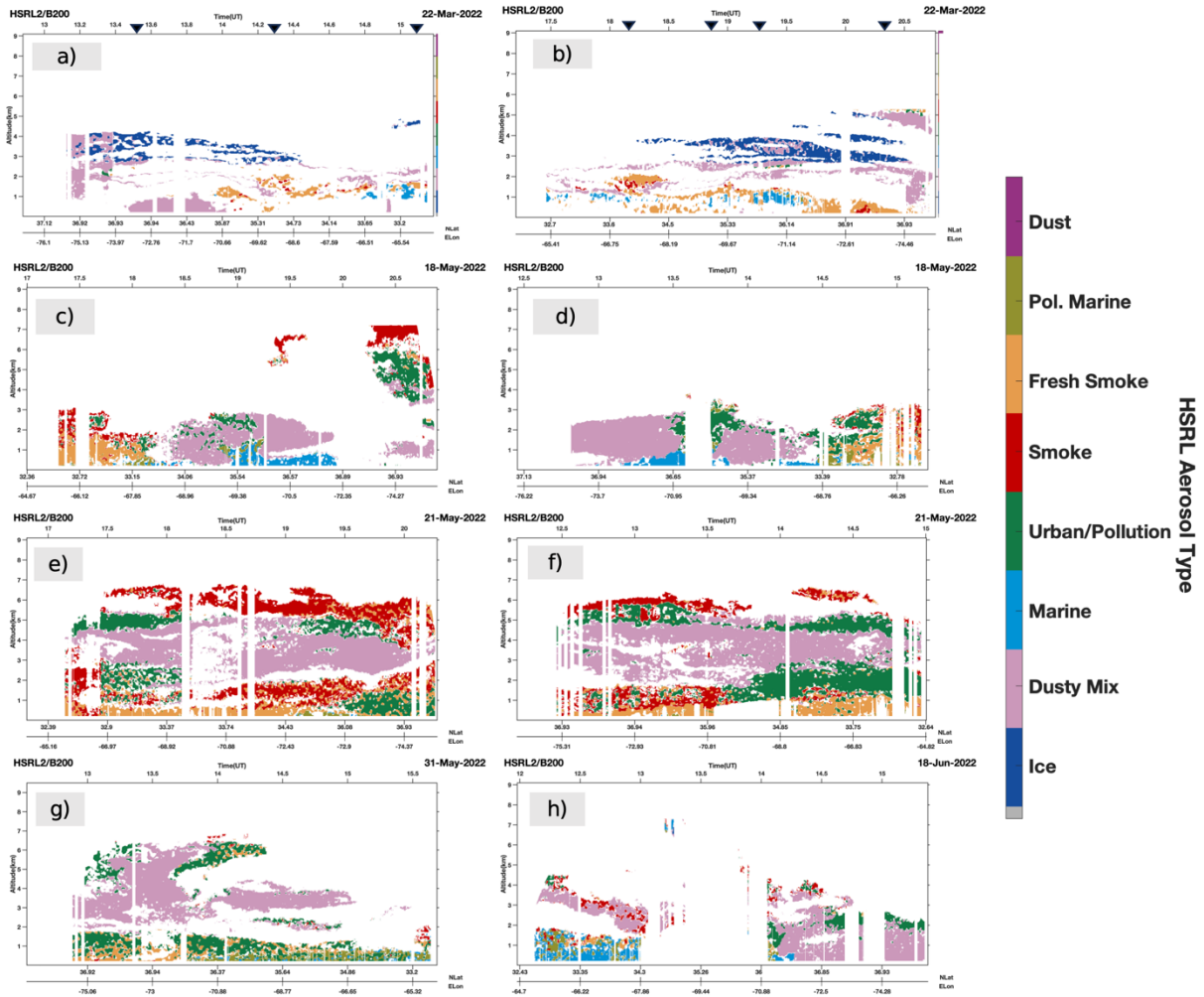
Figure S2: HSLR-2 aerosol type information for all eight transit flights: RFs 142 (a), 143 (b), 156 (c), 157 (d), 159 (e), 160 (f), 161 (g), and 179 (h).
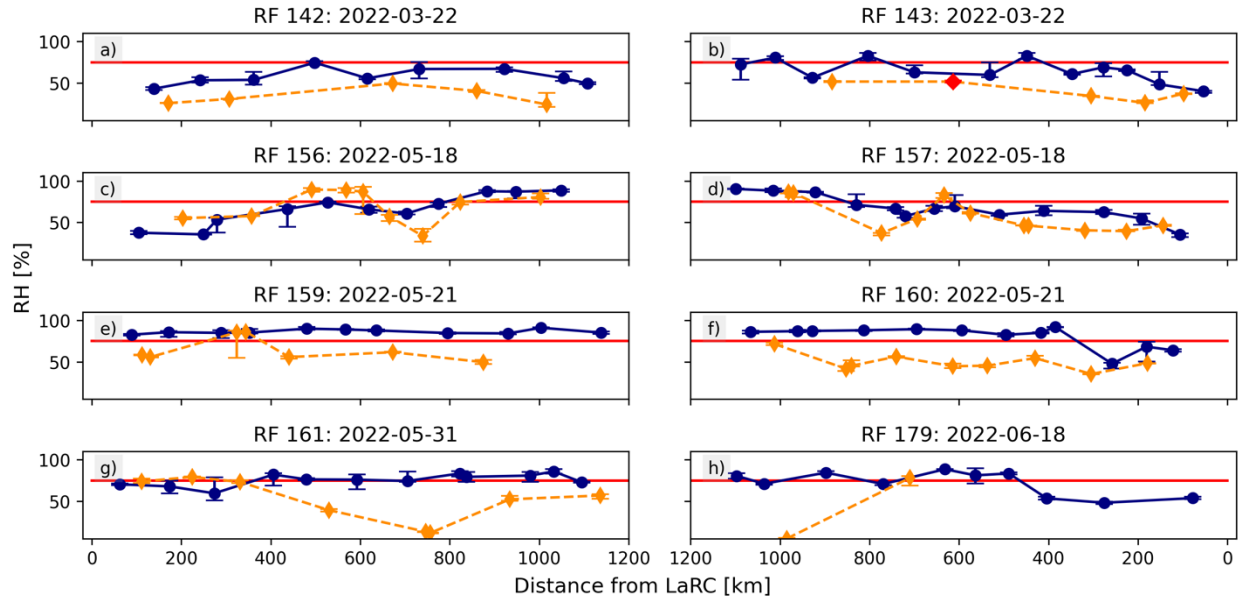
Figure S3: Relative humidity for all eight transit flights as measured by the HU-25 Falcon. Blue and orange represent the BL and FT, respectively. Markers represent median values for 15-minute intervals and whiskers are 25th/75th percentiles. The red line represents 75% RH, which is the deliquescence point of sea salt (pure NaCl) particles. The smoke layer median value is shown in red for RF 143.
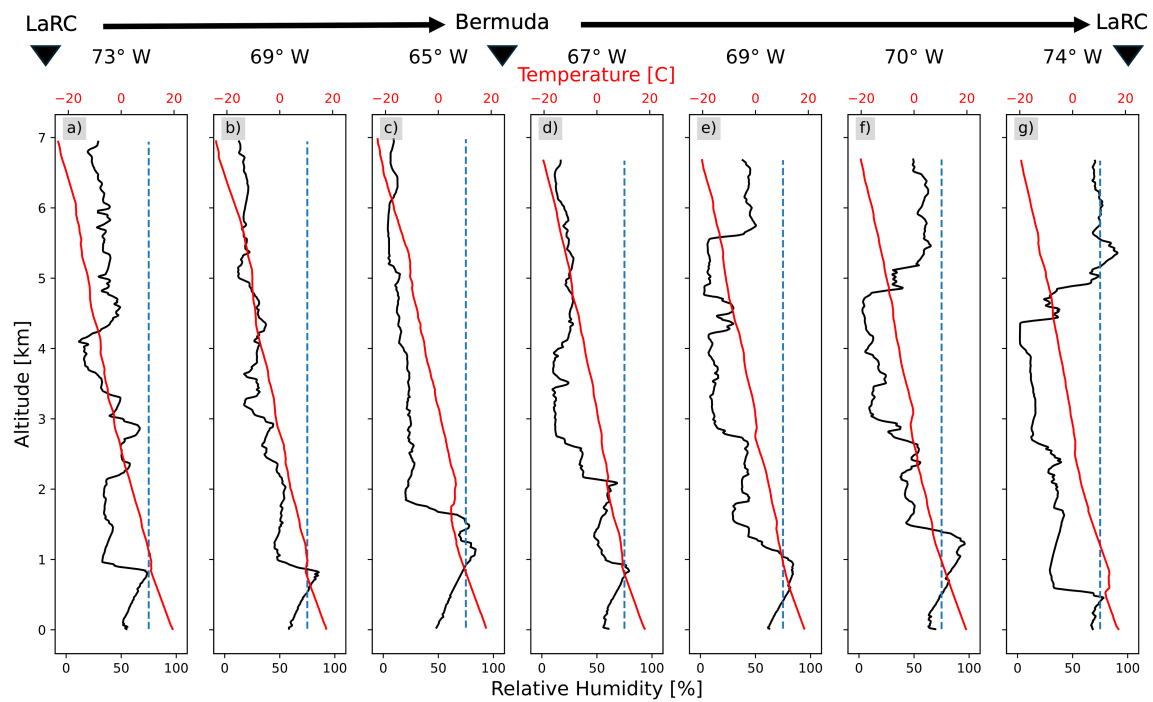
Figure S4: Dropsonde relative humidity (black) and temperature (red) data for the RFs 142-143. The dashed blue vertical line represents 75% RH, which is the deliquescence point of sea salt (pure NaCl) particles. Locations of the dropsonde launches are shown in Figure 3.
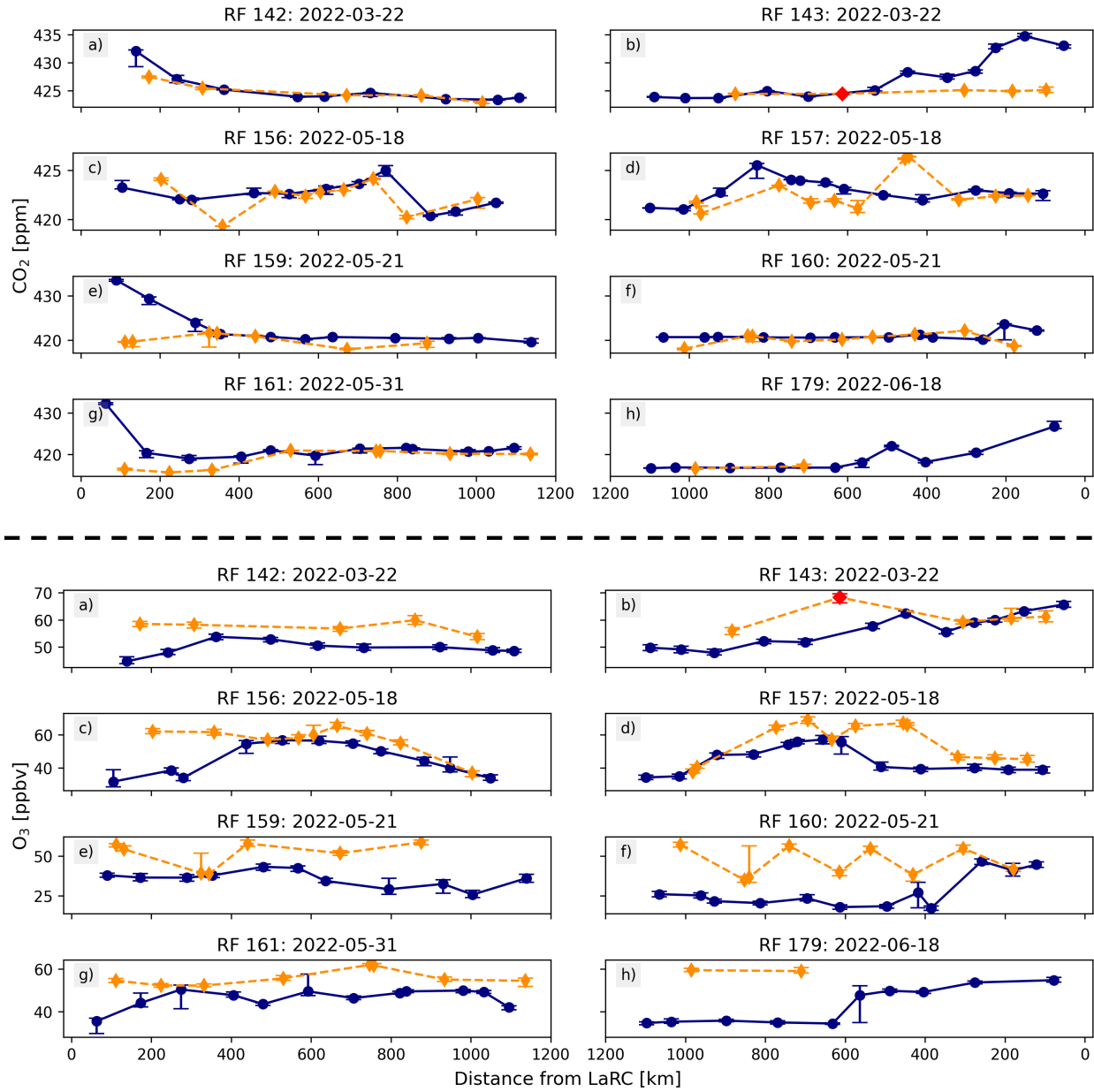
Figure S5: CO_2 and O_3 concentrations as measured by the HU-25 Falcon as a function of offshore distance from LaRC. Blue and orange represent the BL and FT, respectively. Markers represent median values for 15-minute intervals and whiskers are 25th/75th percentiles. The smoke layer median value is shown in red for RF 143.
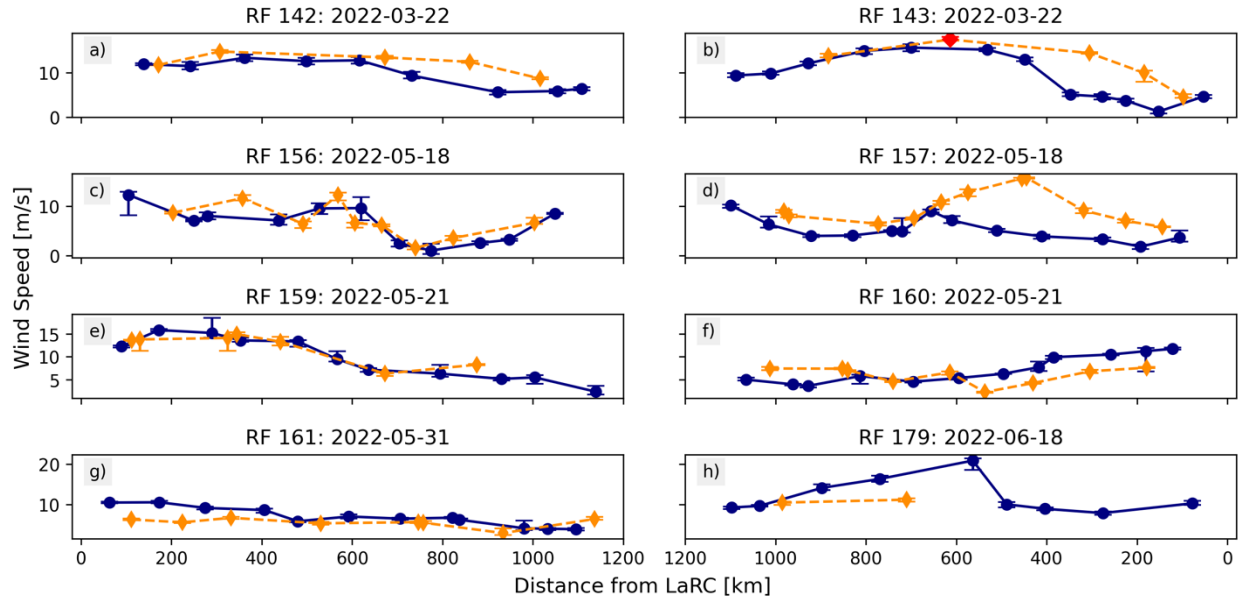
Figure S6: Same as Fig. S5 but for wind speed.
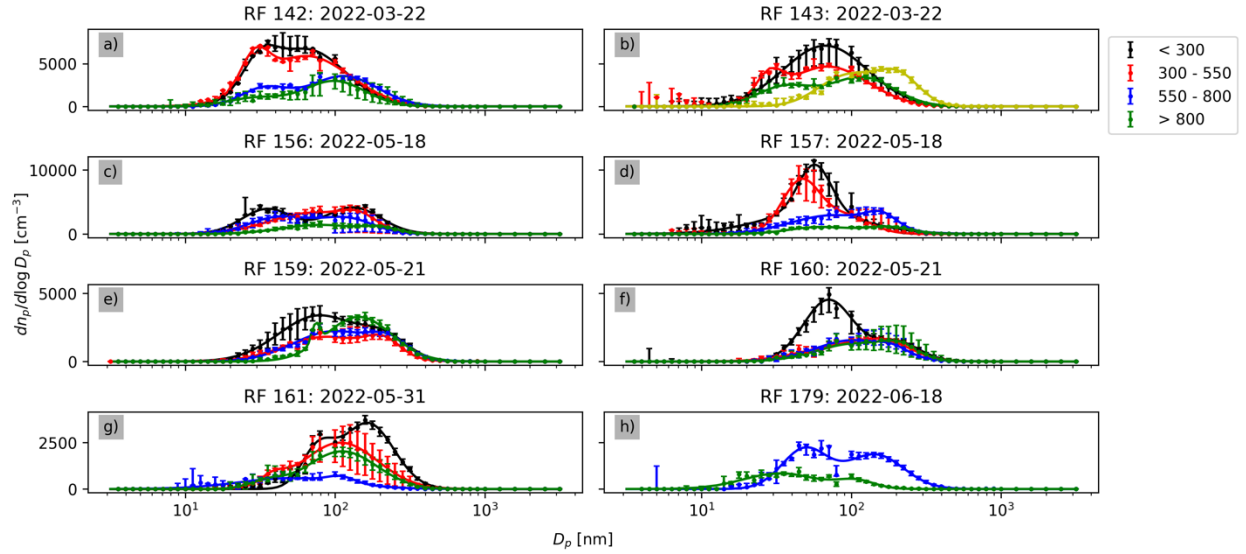
Figure S7: Median particle size number concentration distributions in the free troposphere with best fit models segmented by four offshore distance ranges (in km) relative to LaRC. 25th/75th percentiles are signified with error bars. The smoke layer size distribution from RF143 is shown in yellow.
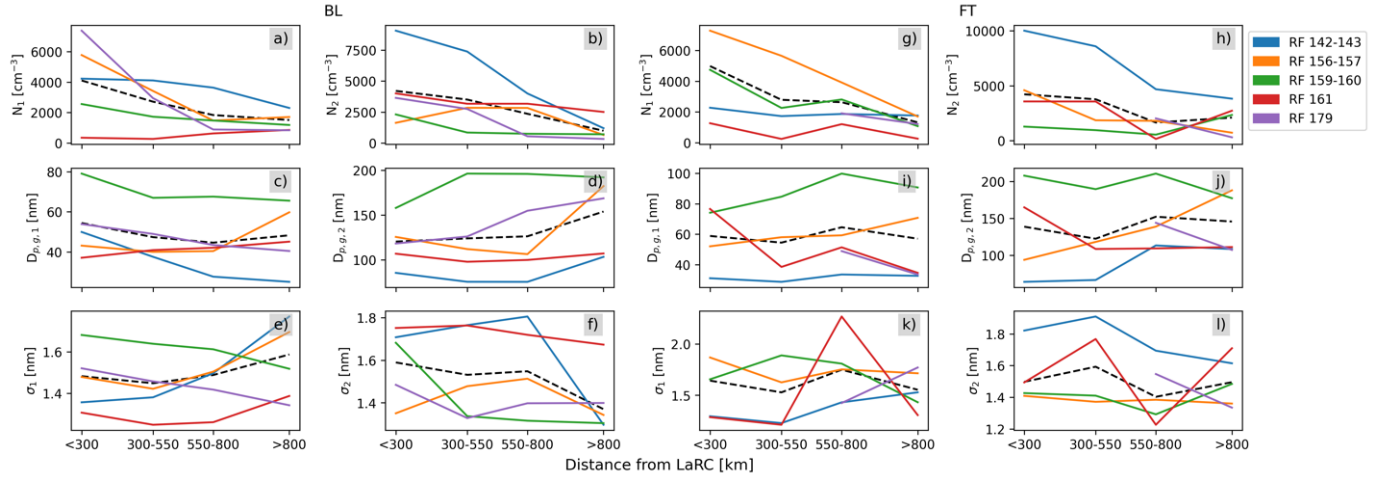
Figure S8: BL (a-f) and FT (g-l) particle number, geometric mean diameter, and geometric standard deviation of the two-mode lognormal fit versus distance from LaRC. Lines are color coded by flight day. Dotted black line displays mean across days.
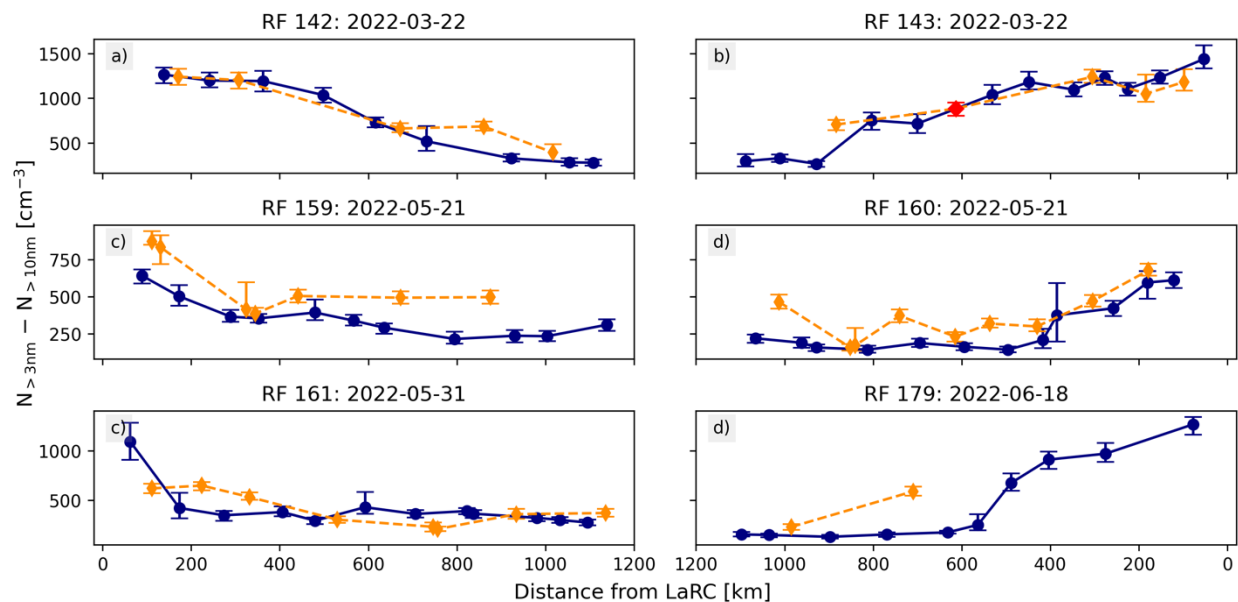
Figure S9: Number concentration of particles with D_p between 3 and 10 nm for all transit flights except RF 156-157 on 18 May 2022 since data were unavailable. Blue and orange represent the BL and FT, respectively. Markers represent median values for 15-minute intervals and whiskers are $25^{th}/75^{th}$ percentiles. The smoke layer median value is shown in red for RF 143.
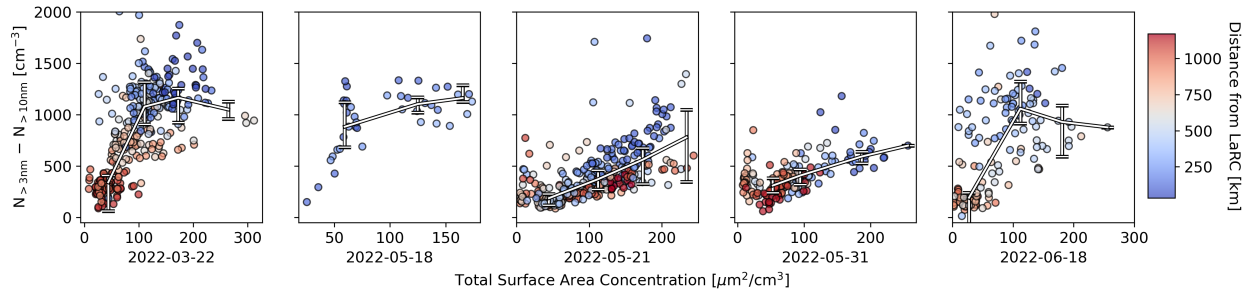
Figure S10: Combined values in the BL and FT for $N_{>3\,nm}$ - $N_{>10\,nm}$ versus total surface area concentration (summed SMPS and LAS data) colored by distance from LaRC. Error bars representing 25th and 75th percentiles from binning data every 100 μm² cm⁻³ are displayed with the white line.