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Measurement report: Closure analysis of aerosol–cloud composition in tropical maritime warm convection

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Table S1: Cloud water sample merged properties

	Timestamp [UTC s]		Altitude [m]	LWC [g sm^{-3}]	RWC	N_d [scm^{-3}]	w [m s^{-1}]	σ_w	CO [ppm$_v$]	CH_4
Case I: 2019-09-19	100350	100430	2862	1.61	0.11	148	0.07	0.94	0.135	1.952
	100430	100460	2845	2.57	0.59	200	1.93	1.54	0.135	1.952
	100560	100700	2586	0.57	0.06	69	0.07	0.57	0.139	1.956
	100725	100805	2517	0.49	0.03	67	0.00	0.46	0.137	1.953
	100805	100910	2518	0.76	0.02	261	0.63	0.96	0.135	1.950
	100910	100965	2513	0.64	0.01	224	0.37	0.85	0.136	1.950
	101055	101175	2380	0.83	0.07	255	-0.08	0.89	0.137	1.952
	101175	101235	2108	0.86	0.17	108	-0.18	0.40	0.138	1.953
	101235	101305	1938	0.44	0.12	103	0.02	0.34	0.139	1.953
	101305	101380	1860	0.43	0.15	115	-0.05	0.27	0.136	1.952
	101380	101425	1867	0.92	0.24	162	0.05	0.36	0.135	1.950
	101425	101475	1871	1.10	0.14	195	0.12	0.36	0.138	1.951
	101475	101515	1869	1.07	0.06	204	0.00	0.26	0.135	1.951
	101515	101585	1864	1.00	0.26	200	0.35	0.83	0.136	1.950
	101585	101655	1863	0.74	0.26	175	0.06	0.61	0.136	1.949
	101655	101695	1867	0.76	0.08	208	0.06	0.22	0.135	1.950
	101695	101750	1866	0.63	0.01	208	0.03	0.17	0.135	1.951
	101750	101925	1861	0.48	0.02	176	-0.01	0.29	0.135	1.950
	101925	102020	1682	0.56	0.20	231	-0.02	0.29	0.136	1.949
	102020	102090	1357	0.26	0.24	13	-0.16	0.28	0.136	1.949
	102090	102135	1208	0.36	0.28	213	0.15	0.42	0.135	1.948
	102135	102205	1219	0.86	0.54	481	0.76	1.42	0.138	1.949
	102205	102295	1216	0.18	0.18	0.2	-0.31	0.41	0.135	1.946
	102295	102355	1220	0.38	0.28	156	-0.10	0.45	0.134	1.946
	102630	102745	1215	0.67	0.40	302	-0.23	0.78	0.137	1.950
	102745	102815	1156	0.29	0.29	1.2	0.09	0.42	0.136	1.948
	103740	103770	483	0.25	0.25	1.2	-0.10	0.19	0.139	1.948
Case II: 2019-09-23	97490	97505	3761	5.65	0.08	782	-0.45	2.76	0.139	1.956
	97585	97675	3762	3.45	0.06	391	0.09	2.14	0.131	1.954
	98050	98110	3279	2.55	0.07	381	0.15	2.36	0.143	1.957
	98115	98165	3274	0.94	0.00	198	-0.54	1.44	0.142	1.958
	98185	98265	3278	1.01	0.00	295	-0.54	2.05	0.136	1.955
	98730	98760	2624	1.13	0.00	380	-0.66	1.38	0.147	1.954
	98835	98880	2635	1.02	0.03	462	2.32	3.34	0.150	1.957
	98890	98940	2620	0.70	0.04	214	-0.34	0.94	0.149	1.959
	98950	99010	2626	0.74	0.04	323	0.45	0.69	0.146	1.955
	99255	99295	2052	1.28	0.03	493	3.41	2.30	0.157	1.964
	99295	99335	2115	0.68	0.21	231	0.62	0.69	0.151	1.959
	99335	99425	2020	0.66	0.02	420	0.16	0.75	0.151	1.958
	99425	99470	1983	0.67	0.18	447	-0.11	0.82	0.152	1.958
	99745	99900	1355	0.35	0.12	190	0.04	0.96	0.157	1.964
	99980	100065	1348	0.63	0.06	456	1.37	1.65	0.159	1.966
	100350	100440	695	0.34	0.09	915	0.89	0.95	0.163	1.971
	101495	101550	231	0.27	0.27	0.2	-0.20	0.36	0.159	1.968
Case III: 2019-09-15	95310	95350	4220	0.88	0.02	402	-0.13	2.47	0.223	1.862
	99205	99240	4204	0.90	0.15	292	-2.35	2.10	0.231	1.863
	99445	99475	3292	0.67	0.27	294	-1.76	1.50	0.318	1.871
	99625	99685	2477	0.51	0.02	627	-0.43	3.14	0.386	1.877
	99825	99910	1812	0.42	0.02	957	0.28	1.14	0.391	1.878
	100110	100175	1285	0.65	0.04	1271	2.42	3.10	0.614	1.896
	100785	100835	623	0.21	0.01	1095	0.50	0.47	0.680	1.903

Table S2: LWC sensitivity to LWC threshold method. L* corresponds to the use of a LWC weight in calculating the mean, rather than a binary mean-over-threshold. Δ is the percentage change from the default calculation using a 0.1 g sm^{-3} threshold.

	Timestamp [UTC s]		LWC (g sm^{-3}) [Δ] - using variable threshold listed							
			L*		0.02		0.05		0.15	
Case I: 2019-09-19	100350	100430	2.26	[+41%]	1.43	[-11%]	1.53	[-5%]	1.72	[+7%]
	100430	100460	2.99	[+16%]	2.57	[0%]	2.57	[0%]	2.57	[0%]
	100560	100700	0.80	[+40%]	0.43	[-24%]	0.52	[-9%]	0.64	[+12%]
	100725	100805	0.70	[+43%]	0.41	[-17%]	0.46	[-6%]	0.57	[+17%]
	100805	100910	1.18	[+56%]	0.55	[-28%]	0.65	[-14%]	0.87	[+16%]
	100910	100965	0.75	[+16%]	0.42	[-35%]	0.48	[-26%]	0.67	[+4%]
	101055	101175	1.00	[+21%]	0.73	[-11%]	0.79	[-5%]	0.84	[+2%]
	101175	101235	0.95	[+11%]	0.86	[0%]	0.86	[0%]	0.86	[0%]
	101235	101305	0.49	[+11%]	0.44	[-1%]	0.44	[-1%]	0.46	[+4%]
	101305	101380	0.56	[+31%]	0.39	[-9%]	0.39	[-9%]	0.49	[+15%]
	101380	101425	0.97	[+5%]	0.92	[0%]	0.92	[0%]	0.92	[0%]
	101425	101475	1.18	[+7%]	1.10	[0%]	1.10	[0%]	1.10	[0%]
	101475	101515	1.19	[+11%]	1.07	[0%]	1.07	[0%]	1.07	[0%]
	101515	101585	1.13	[+13%]	1.00	[0%]	1.00	[0%]	1.00	[0%]
	101585	101655	0.88	[+19%]	0.74	[0%]	0.74	[0%]	0.74	[0%]
	101655	101695	0.81	[+7%]	0.76	[0%]	0.76	[0%]	0.76	[0%]
	101695	101750	0.74	[+17%]	0.60	[-5%]	0.61	[-3%]	0.63	[0%]
	101750	101925	0.60	[+26%]	0.39	[-18%]	0.43	[-11%]	0.51	[+6%]
	101925	102020	0.60	[+7%]	0.56	[0%]	0.56	[0%]	0.56	[0%]
	102020	102090	0.27	[+4%]	0.20	[-21%]	0.21	[-18%]	0.31	[+19%]
	102090	102135	0.42	[+15%]	0.36	[-1%]	0.36	[-1%]	0.38	[+4%]
	102135	102205	1.13	[+30%]	0.65	[-25%]	0.68	[-21%]	0.98	[+14%]
	102205	102295	0.16	[-14%]	0.12	[-33%]	0.14	[-23%]	0.24	[+34%]
	102295	102355	0.44	[+14%]	0.36	[-7%]	0.36	[-5%]	0.41	[+7%]
	102630	102745	0.83	[+25%]	0.54	[-18%]	0.60	[-10%]	0.68	[+2%]
	102745	102815	0.32	[+12%]	0.17	[-41%]	0.21	[-27%]	0.37	[+28%]
	103740	103770	0.23	[-7%]	0.15	[-41%]	0.19	[-24%]	0.31	[+23%]
Case II: 2019-09-23	97490	97505	6.67	[+18%]	5.65	[0%]	5.65	[0%]	5.65	[0%]
	97585	97675	5.27	[+53%]	2.99	[-13%]	3.45	[0%]	3.63	[+5%]
	98050	98110	4.23	[+66%]	2.27	[-11%]	2.43	[-5%]	2.89	[+14%]
	98115	98165	1.48	[+56%]	0.92	[-3%]	0.92	[-3%]	1.00	[+5%]
	98185	98265	1.33	[+31%]	0.73	[-28%]	0.87	[-15%]	1.01	[0%]
	98730	98760	1.48	[+31%]	1.04	[-8%]	1.08	[-4%]	1.18	[+4%]
	98835	98880	1.39	[+36%]	0.84	[-18%]	0.97	[-6%]	1.12	[+9%]
	98890	98940	0.87	[+24%]	0.61	[-12%]	0.64	[-8%]	0.72	[+2%]
	98950	99010	1.47	[+101%]	0.51	[-30%]	0.62	[-16%]	0.95	[+30%]
	99255	99295	1.53	[+20%]	1.21	[-5%]	1.24	[-3%]	1.28	[0%]
	99295	99335	0.88	[+29%]	0.59	[-13%]	0.66	[-3%]	0.76	[+12%]
	99335	99425	0.75	[+14%]	0.49	[-26%]	0.57	[-13%]	0.71	[+8%]
	99425	99470	0.83	[+23%]	0.51	[-24%]	0.55	[-18%]	0.77	[+14%]
	99745	99900	0.50	[+43%]	0.20	[-44%]	0.26	[-25%]	0.41	[+16%]
	99980	100065	0.82	[+30%]	0.49	[-23%]	0.55	[-14%]	0.69	[+9%]
	100350	100440	0.34	[+2%]	0.24	[-28%]	0.27	[-21%]	0.37	[+9%]
	101495	101550	0.31	[+14%]	0.22	[-17%]	0.24	[-13%]	0.31	[+14%]
Case III: 2019-09-15	95310	95350	1.52	[+72%]	0.69	[-21%]	0.80	[-10%]	0.93	[+5%]
	99205	99240	1.19	[+32%]	0.67	[-26%]	0.74	[-18%]	0.95	[+6%]
	99445	99475	0.87	[+30%]	0.56	[-16%]	0.60	[-11%]	0.76	[+13%]
	99625	99685	0.67	[+29%]	0.42	[-19%]	0.45	[-12%]	0.59	[+15%]
	99825	99910	0.51	[+21%]	0.31	[-26%]	0.36	[-14%]	0.47	[+10%]
	100110	100175	0.84	[+31%]	0.54	[-17%]	0.60	[-8%]	0.76	[+18%]
	100785	100835	0.21	[-2%]	0.15	[-28%]	0.19	[-13%]	0.26	[+20%]

Table S3: Summary statistics of CW properties for each case

Species		Air Equiv. Mass Conc. (μg sm^{-3})		Aqueous Conc. (mg L^{-1})	
		mean	min / median / max	mean	min / median / max
SS	I	8.5	2.5 / 6.1 / 21.7	16.6	2.8 / 7.6 / 28.4
	II	8.9	2.3 / 6.2 / 15	11.3	1.8 / 5.7 / 32.7
	III	5.2	1.6 / 5.5 / 8.5	12.6	2.5 / 8.4 / 31.3
$_{nss}SO_4^{2-}$	I	2.7	0.9 / 2.5 / 4.9	4.7	1.3 / 3.4 / 8.9
	II	3.5	1.2 / 2.5 / 3.8	3.6	1.1 / 2.3 / 8.9
	III	3.6	1.5 / 3.2 / 6.3	8.5	2.2 / 6.2 / 18
NO_3^-	I	1.6	0.5 / 1.1 / 3.4	3.0	0.7 / 1.4 / 4.6
	II	4.8	1.2 / 3.1 / 7.2	5.8	1.1 / 2.9 / 16
	III	3.6	1.3 / 4.1 / 5.9	8.6	2 / 5.5 / 20.9
NH_4^+	I	0.6	0.2 / 0.6 / 0.9	1.0	0.3 / 0.8 / 1.9
	II	1.6	0.7 / 1.2 / 1.9	1.7	0.5 / 1.3 / 3.9
	III	3.2	1.7 / 2.6 / 5.2	7.0	2.5 / 6.1 / 12.3
Sum of organic ions (total mass)	I	0.4	0.2 / 0.3 / 0.7	0.7	0.2 / 0.6 / 1.2
	II	1.0	0.5 / 0.7 / 1.8	1.0	0.6 / 0.8 / 1.9
	III	4.5	2.6 / 3.5 / 7.3	9.7	3.7 / 8.7 / 17.2
TOC	I	0.5	0.2 / 0.4 / 0.8	0.7	0.4 / 0.6 / 1.1
	II	0.9	0.5 / 0.8 / 1.8	1.1	0.7 / 1 / 1.5
	III	5.4	3.3 / 4 / 8.8	8.8	5 / 7.7 / 13.7
		Air Equiv. Mass Conc. (ng sm^{-3})		Aqueous Conc. (μg L^{-1})	
$_{nss}K^+$	I	-	-	-	-
	II	52	0 / 1 / 136	67	0 / 1 / 285
	III	225	72 / 191 / 426	539	107 / 373 / 1189
$_{nss}Ca^{2+}$	I	49	0 / 1 / 171	125	0 / 2 / 277
	II	648	45 / 142 / 1024	397	51 / 178 / 1265
	III	175	28 / 176 / 323	472	53 / 245 / 1434
Glycolate	I	16	5 / 10 / 22	35	8 / 16 / 34
	II	27	4 / 11 / 33	15	5 / 12 / 29
	III	-	- / - / 10	-	- / - / 14
Acetate	I	134	60 / 124 / 210	201	86 / 205 / 304
	II	421	107 / 275 / 921	338	284 / 330 / 409
	III	1671	780 / 1506 / 2548	3582	1167 / 3120 / 6259
Formate	I	64	10 / 44 / 151	105	14 / 88 / 211
	II	237	74 / 164 / 464	253	69 / 239 / 499
	III	1561	760 / 1360 / 2457	3107	1311 / 3149 / 5322
MSA	I	7	2 / 6 / 10	13	5 / 8 / 15
	II	15	5 / 10 / 15	14	3 / 11 / 33
	III	3	1 / 3 / 5	6	4 / 6 / 8
Pyruvate	I	31	4 / 30 / 70	54	4 / 48 / 109
	II	27	14 / 21 / 48	34	9 / 24 / 75
	III	112	44 / 80 / 225	253	51 / 214 / 529
Glutarate	I	3	1 / 2 / 8	8	2 / 4 / 20
	II	13	2 / 5 / 22	11	2 / 6 / 35
	III	105	44 / 91 / 176	236	65 / 191 / 459
Adipate + Succinate	I	39	6 / 24 / 83	73	11 / 62 / 134
	II	74	17 / 60 / 160	106	16 / 56 / 372
	III	557	241 / 400 / 1139	1265	345 / 922 / 2715
Maleate	I	-	-	-	-
	II	-	-	-	-
	III	7	5 / 7 / 8	11	9 / 11 / 12
Oxalate	I	116	49 / 93 / 215	213	78 / 160 / 329
	II	204	58 / 134 / 277	226	56 / 134 / 562
	III	511	208 / 499 / 871	1232	313 / 806 / 2894
pH (unitless)	I			4.36	3.96 / 4.39 / 4.63
	II			4.56	4.27 / 4.54 / 5.03
	III			4.16	3.82 / 4.26 / 4.43

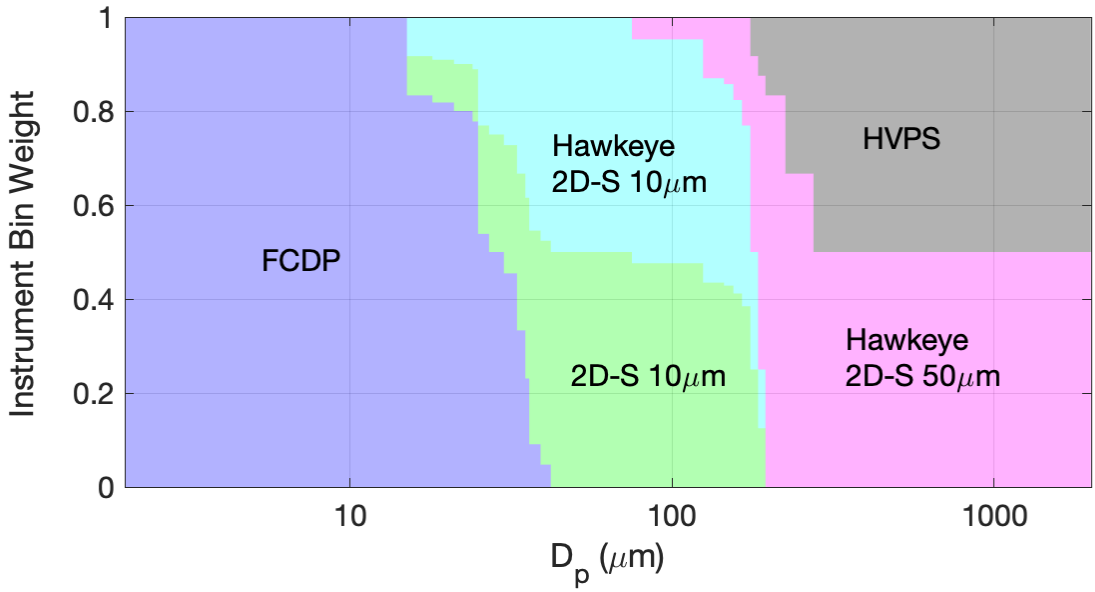

Figure S1: Stitching weight diagram for the estimate of the combined drop size distribution and integrated quantities

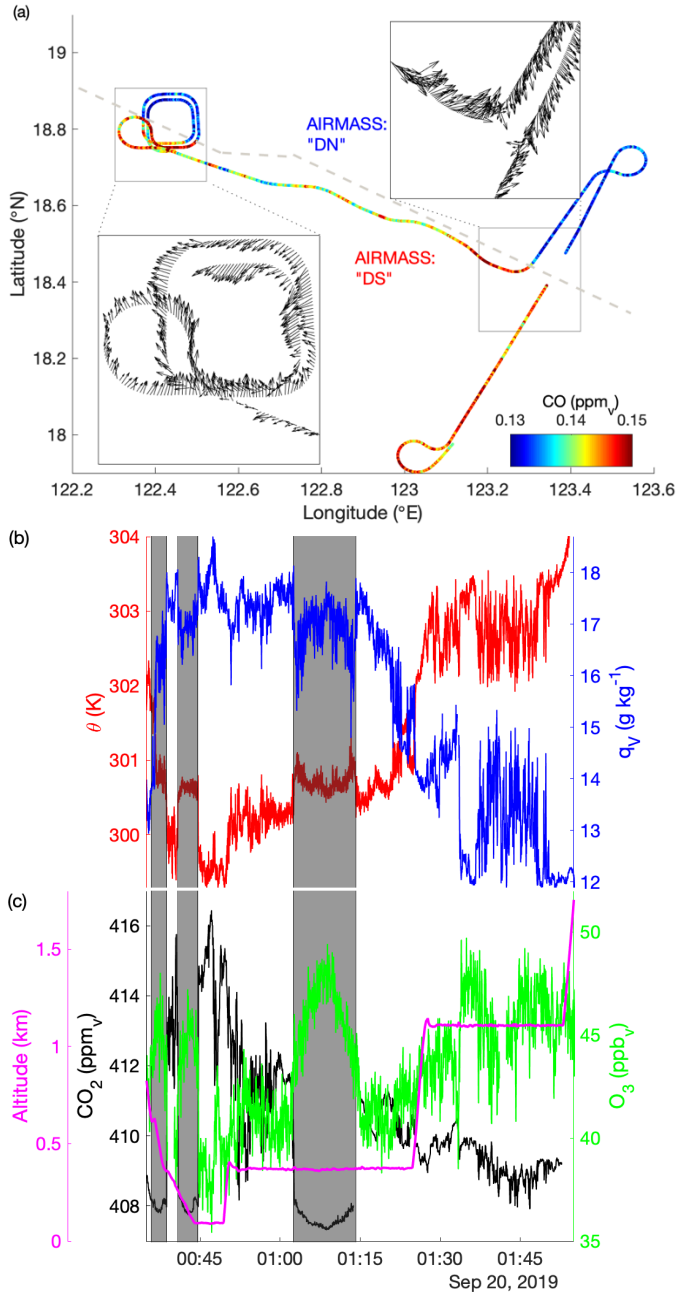

Figure S2: (a) Relationship between horizontal wind anomaly and CO concentration along an airmass boundary sampled downwind of the cloud module in Case I. (b-c) Time series of supporting properties highlighting the cool, moist, O_3-poor and CO_2-enhanced downwind-south (DS) airmass.

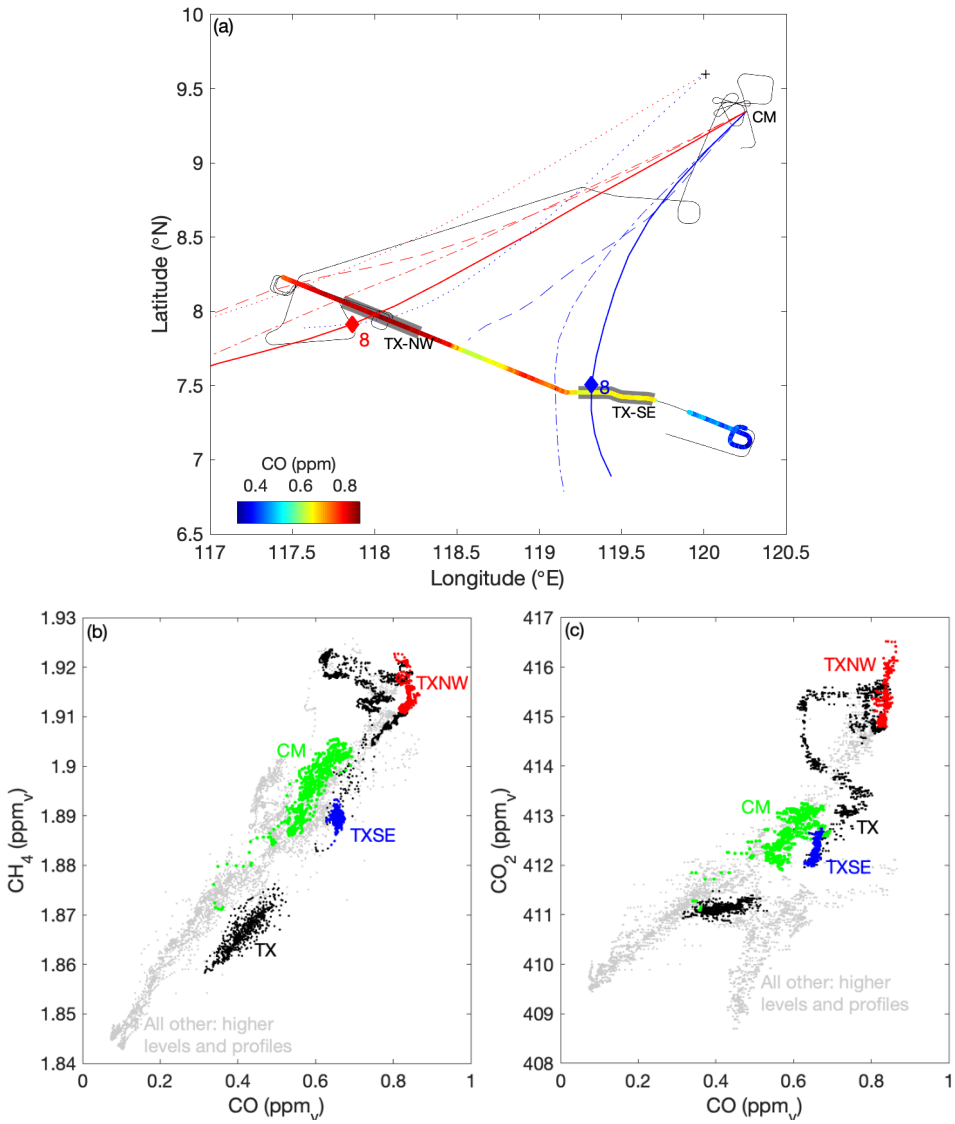

Figure S3: (a) Back trajectory analysis at the cloud module location for Case III with relevant flight segment included [thin black]. Back trajectories were initiated from the center of the sub-cloud [lowest] level leg at the leg altitude [600 m, solid blue] and just above cloud base [1000 m, solid red] at 0400Z September 16. The temporal sensitivity is investigated by repeating trajectories at 00Z [dash] and 08Z [dot-dash]. Spatial sensitivity is investigated by displacing the trajectory end point 0.25° north and 0.25° west [dot]. The diamond markers indicate the projected -8 hour airmass location demonstrating the estimated transit time between the upwind transect and the cloud module. The upwind transect is colored by CO mixing ratio and grey zones represent the regions used to calculate the southeast (TX-SE) and northwest (TX-NW) airmass properties, selected to coincide with the back trajectory uncertainty. (b) and (c) show gas ratios for the entire transect (TX), the TX-NW and TX-SE subsections and the cloud module (CM).

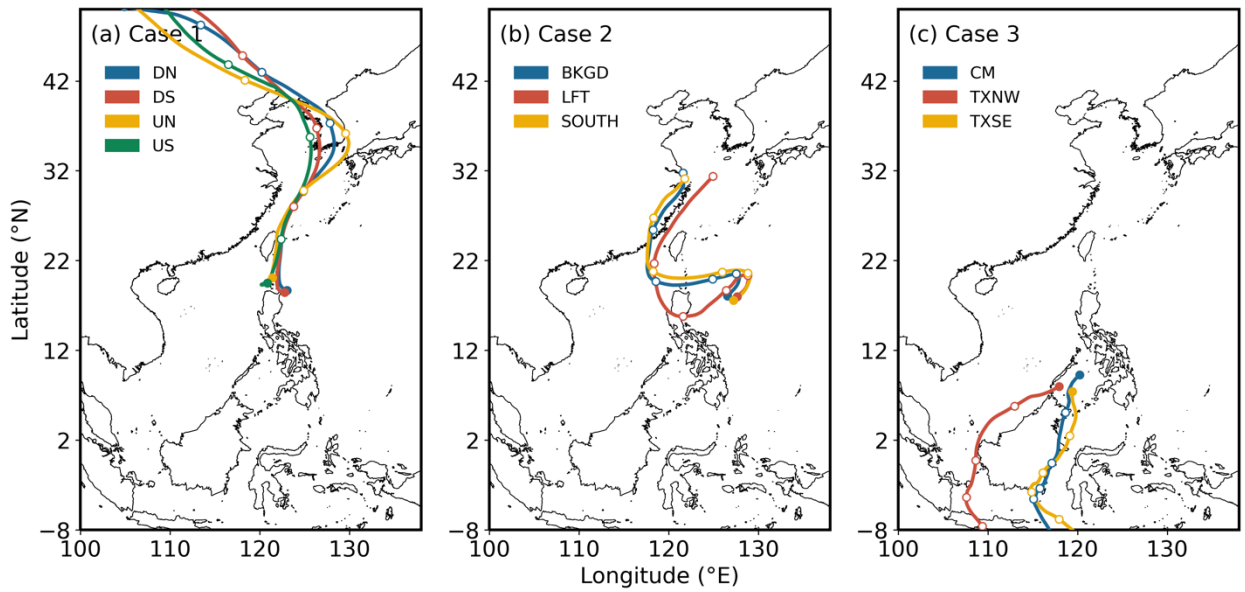

Figure S4: 120 hour back trajectories showing the mean location of the airmasses identified in each case (see text for airmass descriptions). Circles represent intervals of 24 hours. Note that in Case III, the sub-airmasses (TXNW and TXSE) are shown in place of the entire upwind transect (TX) because of the bifurcation of the airmass origin.