



Supplementary Materials for

Locally narrow droplet size distributions are ubiquitous in stratocumulus clouds

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Science **384**, 528 (2024)
DOI: 10.1126/science.adi5550

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Supplementary materials

Materials and Methods

Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA): The ACE-ENA campaign is an atmospheric research field campaign to study low-level clouds in the Atlantic Ocean sponsored by the Department of Energy's (DOE) Atmospheric Radiation Measurement (ARM) research facility. The eastern North Atlantic is a region marked by persistent annual coverage of low-level stratocumulus clouds maintained by the Azores high. ACE-ENA had a total of 39 research flights with the objective of better characterizing the marine low-level clouds and the associated boundary layer properties. The region also lies in the path of long-range aerosol transport from three major continents making it ideal to study aerosol-cloud interactions. The flights are evenly split into two intensive operational periods (IOPs) between the summer (June-July) of 2017 and the winter (Jan-Feb) of 2018. For this study, we compiled Holographic Detector for Clouds (HOLODEC) data from a total of 31 cloud transects from four research flights (3 in summer and 1 in winter) each containing well-developed stratocumulus cloud systems under different boundary layer conditions. These flights also have identifiable drizzle embryo populations (as defined by Hocking radius 40 microns) which hold the key to drizzle initiation (42). The segments from each flight are selected to be at a constant altitude so as to aim at horizontal homogeneity. The details of the different flight segments analyzed are reported in Tables S1 and S2.

Holographic Detector for Clouds(HOLODEC): HOLODEC works on the principle of digital inline holography and captures instantaneous 3D snapshots of small cloud volumes($\approx 10\text{cm}^3$) at successive intervals. In addition to the particle diameter, HOLODEC is capable of revealing the position of each particle in its sample volume. HOLODEC measures all parti-

cles (liquid and ice) in the size range from 6 – 2000 microns; for this study, a lower-cutoff of 10 microns was employed. The operational frequency is 3.3 Hz thereby capturing around 3-4 holograms every second. Aboard an aircraft moving at 100 m/s, this corresponds to a spatial separation of around 30 m. It differs from traditional cloud measurement probes which measure one particle at a time and require a larger path length to get size distribution information. HOLODEC on the other hand acquires all targets in a sample volume large enough to generate size distribution information and hence is best suited to look at local cm scale differences. The National Center for Atmospheric Research (NCAR) Earth Observing Laboratory (EOL) web page for HOLODEC (<https://www.eol.ucar.edu/instruments/holographic-detector-clouds>) has more technical information on HOLODEC along with the data sets from the different field campaigns.

Algorithm: The algorithm employs non-parametric hypothesis testing and data clustering techniques to classify all the regions of a cloud containing a similar population of cloud droplets. In other words, it is a measure of similarity for cloud sample size distributions at different points in space or time. It inputs a sequence of spatially/temporally tagged cloud data (size distributions) and outputs clusters of these that are statistically similar. When applied to cloud data from HOLODEC, it classifies holograms that can be defined by the same underlying drop-size distribution into clusters. To do this, the algorithm uses hypothesis testing and a data-clustering algorithm to find the different size distribution shapes found within a cloud segment. These different size distribution shapes constitute the different characteristic distributions. Each hologram then is either assigned to a cluster or, if it doesn't have a distribution that is similar enough to any of the characteristic distributions, remains unassigned. We specify a minimum of at least 10 holograms for each of these characteristic distributions.

The algorithm can be simplified into two parts. The first step in the algorithm is to look at the similarity of any two distributions via hypothesis testing. The non-parametric and widely used

Kolmogorov-Smirnov (KS) test is employed to test the hypothesis that the two samples (from two holograms) have the same distribution shape (43). The KS test uses the maximum distance between these distributions' cumulative distribution functions (CDFs) to define a test statistic. If the probability of observing this test statistic given that the hypothesis was true is less than the prescribed significance level the hypothesis is rejected. This significance level called the alpha value is set to be 5 percent for our calculations. The results of KS tests are expressed in binary fashion with zero indicating that the hypothesis is true (the distributions come from the same population) and 1 indicating that the hypothesis is false (the distributions are not from the same population). The KS test operates on the CDF of the samples, the step size of which depends on the number of drops in each hologram. So to avoid any possible discrepancies in KS test results due to the different concentrations for different holograms, we sample each hologram to a uniform number of particles given by 70 percent of the mean droplet counts from all holograms in the flight segment. This entire hologram-hologram intercomparison is done an ensemble number of times (here performing the KS test 1000 times for each hologram pair) to mitigate sampling uncertainties. The thousand binary KS test results performed for each hologram pair are averaged to determine a final value between 0 and 1. A value closer to 0 means that the null hypothesis is true (that the size distributions are samples that come from a common underlying size distribution) and a value closer to 1 means that the null hypothesis is false and they are dissimilar.

All the cloud samples are compared against each other and so for n holograms we have n^2 of these KS results with values from 0 to 1 with a step size of 0.001. The algorithm then constructs a matrix of size $n \times n$ with each cell having values between 0 and 1 with the row and column of the matrix indicating the compared holograms. This matrix of KS results or "KS matrix" is now fed into a density-based clustering algorithm. Here we employ Ordering Points To Identify the Clustering Structure (OPTICS) (44, 45). Previously in Allwayin et al.

(2022), Density-Based Spatial Clustering of Applications with Noise (DBSCAN) was used for clustering points in the KS matrix (46, 47). DBSCAN requires three parameters; ϵ specifying the maximum distance between two points in a cluster, *MinPoints* that specifies the minimum number of points to form a cluster, and another parameter to specify the metric used to compute the distance between the points. The third parameter is significant as this algorithm does not operate in an Euclidean space but rather in an abstract space where the distribution of points is defined by this parameter. For us, this space for a single hologram is defined by the average KS scores it has with the other holograms. The distances vary from 0 – 1 with holograms with smaller KS scores lying closer and more likely to be similar to each other. OPTICS is an upgrade to DBSCAN addressing its shortcoming of not being very effective in identifying clusters having different mean distances between its points and thus not capable of dealing with points of varying densities in space. OPTICS solves this issue by estimating a “reachability distance” for every point in this abstract space and using it to generate a dendrogram-like plot with valleys in this plot forming the different clusters. In practice, compared to DBSCAN this reduces the number of parameters to one—namely the minimum number of points required to form a cluster. For OPTICS, the appropriate ϵ is determined by the algorithm, and the metric to define the distances is already given by the KS Matrix.

The clustering algorithm, here OPTICS, thus segments the KS matrix into different clusters of holograms with similar size distributions. These size distributions define the characteristic size distributions. As in any clustering algorithm, some points do not get classified into any of the defined clusters. In our case, these are the holograms that are not sufficiently similar to the characteristic distributions. In some cases, these are holograms with droplets similar to one of the characteristic distributions but not similar enough to be categorized in the same cluster. The others are holograms representing cloud regions with disparate distributions.

Though the algorithm is mostly non-parametric, a few parameters can be optimized, and

changing them can affect the results in non-trivial ways (For more information see Allwayin et al. 2022). Typical values are chosen for the parameters-alpha for KS tests (0.05), sampling cutoff for hologram droplets (70 percent of mean droplets in a hologram for the flight segment), and the minimum number of cluster points (greater than 10) are chosen. This set of parameters is the standard set that is general enough to get reasonable clustering results for all cloud segments analyzed. For each of the cloud segments, these parameters can be optimized individually. For both cases, the major clusters are always identified and the qualitative nature of the results is preserved. The algorithm is mostly non-parametric works relatively unsupervised and importantly does not assume any functional form for the distribution types. Additional information about the impacts of particular parameter choices is given in Allwayin et al. 2022.

Supplementary Text

Kolmogorov-Smirnov test for local vs global comparison: KS test is also employed to evaluate the statistical similarity between local hologram cloud volumes to the global average size distribution. For each such comparison, the droplets forming the global distribution are re-sampled to match the droplet count of the local sample. This is repeated 1000 times to constitute a statistical ensemble and the results are averaged to form a statistical similarity score. A value close to one indicates maximum similarity and a value close to zero indicates no similarity. Only the holograms having more than 70 percent of the mean droplet count are evaluated. For the segment from Fig. 1, cloud droplets at a local scale show no resemblance to the global average with practically no holograms having a similarity score greater than 0.5 (Fig. S1). Comparisons to other segments show that the number of holograms resembling the global average is almost always less than 10 percent.

Calculation of mass autoconversion rates: Autoconversion represents the first stage of precipitation formation where cloud droplets collide with each other to form drizzle embryos (I). When these drizzle embryos become sufficiently large ($\approx 40 \mu\text{m}$ diameter), accretion becomes the dominant growth mechanism. For the results reported in this paper, we use the three-moment mass autoconversion rate (P_L) (37, 48), given by,

$$P_L = \left(\frac{3}{4\pi\rho_w} \right)^2 k_l \frac{\Gamma(\alpha_d)\Gamma(\alpha_d + 3, x_{cq})\Gamma(\alpha_d + 6, x_{cq})}{\Gamma^3(\alpha_d + 3)} \frac{L^3}{N} \left[\frac{g}{cm^3 s} \right]. \quad (1)$$

Here, $x_{cq} = r_{cr}/\beta_d$, where r_{cr} is the critical radius for starting autoconversion and is taken to be $10.3 \mu\text{m}$ (49). α_d and β_d are the shape and scale parameters for the gamma distribution fit of the cloud droplet size distribution. They account for the contribution of the shape of the distribution. $k_l = 1.9 \cdot 10^{11} cm^{-3} s^{-1}$ is the Long collision kernel coefficient and L and N is the liquid water content and number concentration of the sample.

For our local autoconversion rate calculations, the droplet size distribution from each hologram above the 70 percent cutoff threshold is fitted to a gamma distribution to obtain the shape and scale parameters. The *MATLAB* gamma fitting function *gamfit* is used to fit the cloud droplet size distribution. This together with the liquid water content and number concentration of the hologram are used in the local autoconversion rate calculation. For the global value, we pool together all the cloud droplets from the segment (above threshold) to fit a gamma to this distribution and use the global average liquid water content and number concentration. It is worth mentioning that a gamma distribution may not be the best-fit option for the local size distribution but is done here for consistency when compared to the global average. The mean of the local process rates calculated for all the different clusters, the unclustered holograms, and the global average value are reported in Tables S1 and S2.

Note that the only time in this work that the measured distributions were fitted to a gamma distribution was for this autoconversion rate calculation; as mentioned earlier, the data clustering algorithm to identify cluster membership was based on hypothesis testing and a machine-learning clustering algorithm only.

Discussion on relationships to updraft velocity: We report our investigations into the relationships of different clusters to updraft velocity for the segment from July 18, 2017, illustrated in the paper. For this particular case, no evident correlations to updraft velocities are observed (Fig. S2). This may be perhaps due to the fact that for stratocumulus cloud systems the fluctuations in updraft velocities are of the order of the mean velocity thereby making it difficult to isolate correlations. Preliminary investigations into such relationships with the other cloud segments were also inconclusive.

Mixing diagram: Here we illustrate the mixing diagram for the segment from July 18, 2017 (Fig. 4 in the paper) in the more traditional coordinates of mean volume diameter and number concentration in Fig. S3. The different colors correspond to the different clusters with the grey

lines showing the unclustered holograms. Consistent with what is described in the main text, the constant mean volume diameters associated with the different clusters point to inhomogeneous mixing signatures. Note that as stated previously in the paper, this result is not evident in the absence of results from the algorithm.

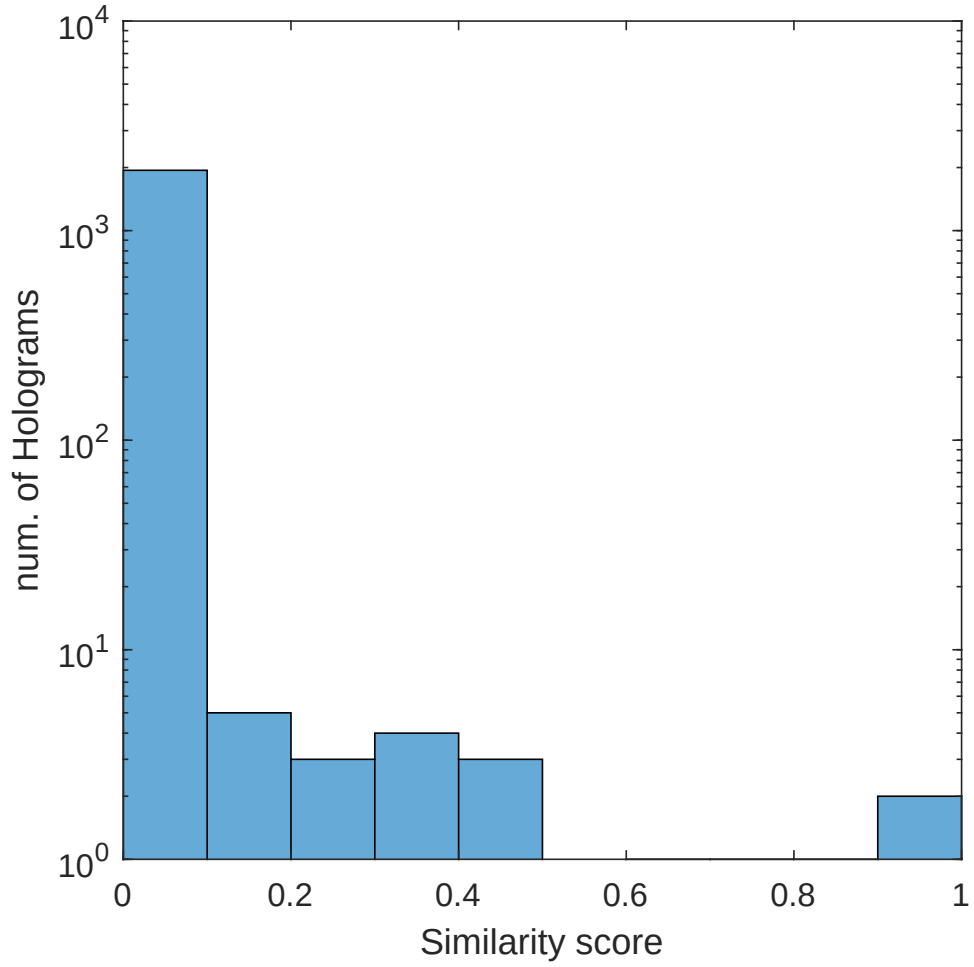


Fig. S1. Histogram of Kolmogorov-Smirnov test results of statistical similarity between local hologram size distributions and the global segment average. The results are from an ensemble of Kolmogorov-Smirnov tests for each hologram with a value of 1 indicating maximum similarity and a value of 0 indicating least similarity. Almost all holograms (except 2 – about 0.1% of the sample) have a similarity score of less than 0.5. The results are for the segment from the flight on July 18, 2017 shown in the paper.

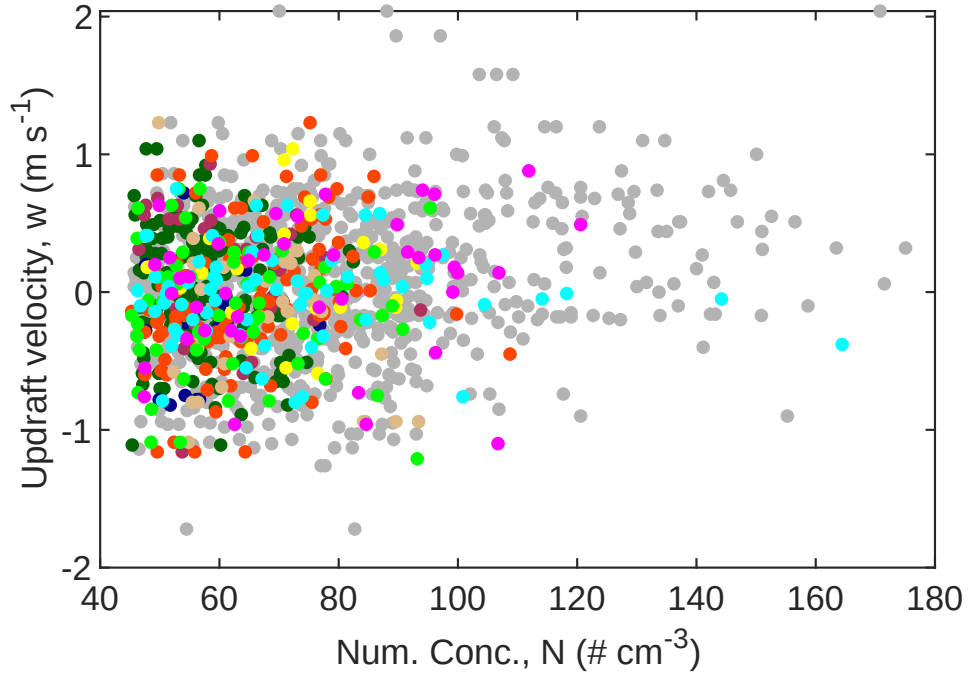


Fig. S2. Relationship between the identified characteristic distributions to the updraft velocity for the cloud segment from July 18, 2017, illustrated in the main paper. The different colors correspond to the different clusters identified by the algorithm with the grey dots representing the unclustered holograms. No relationship between the updraft velocities and cluster identity is immediately evident.

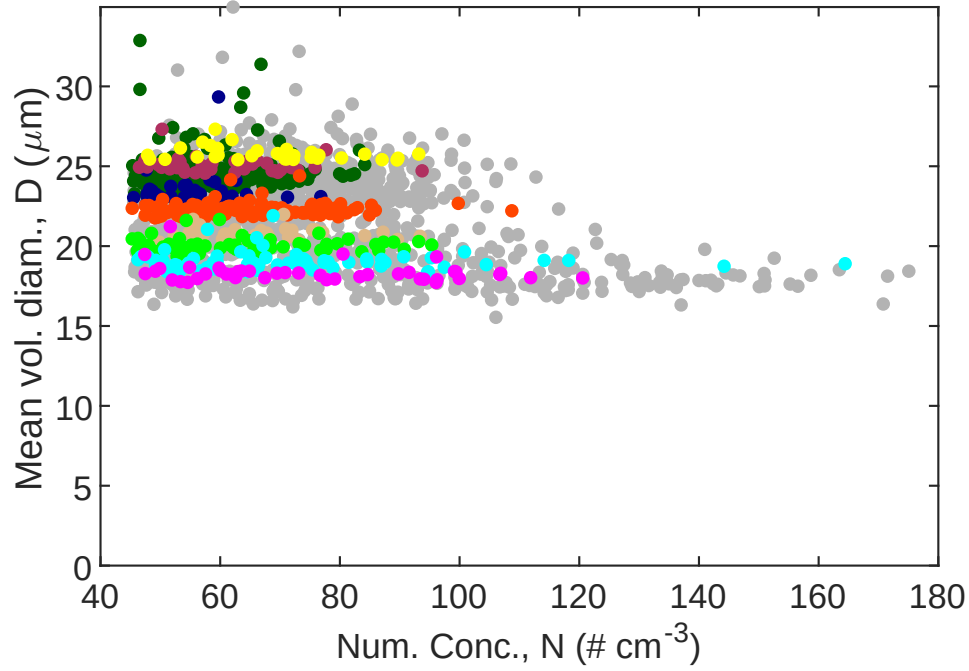


Fig. S3. Mixing diagram in coordinates of mean volume diameter and number concentration for the cloud segment from July 18, 2017, illustrated in the main paper. The different colors correspond to the different clusters identified by the algorithm with the grey dots representing the unclustered holograms. The constant mean volume diameters associated with the different clusters point to inhomogeneous mixing. The clusters may then be interpreted as cloud parcels with different histories that are inhomogeneously mixed. This signature is lost without information on the characteristic distributions.

Time (seconds after 00:00 UTC)	Altitude (meters)	Similarity to ensemble avg (percentage)	Classified holograms (above thresh) (percentage)	Global DSD properties		Clusters count	Local DSD properties: for Clusters		Clusters	Mass autoconversion rates (in log10 scale) ($g\,m^{-3}\,s^{-1}$)			
				Mean	IQR		Mean (micro-meters)	IQR		Unels	Rate of avg. DSD	Avg. of local rates	
AEJ RF16 40400-40800 40900-41200 41200-41400 41400-41900 41800-42500 43950-46100 46100-46300 46400-46800 46900-47300 47480-47980 49900-50410	977	12	98	15.3	3.5	2	15.3,12.6	4.1,1.8	-14.39, -13.23	-13.12	-13.80	-15.38	
	994	8	47	16.8	3.1	6	18.2,17.2,16.6,16.1, 15.0,19.3	4.1,3.6,3.8,3.5, 2.3,4.1	-12.59, -13.02, -13.23, -13.76, -15.71, -12.14	-12.64	-12.72	-13.30	
	1016	6	54	14.4	3.1	4	15.6,14.0,13.4,12.8	3.9,2.7,2.1,1.5	-14.29, -16.50, -16.29, -19.36	-13.93	-14.20	-15.57	
	1040	1	62	16.1	4.5	5	14.1,12.9,16.6,17.4, 18.6	2.7,1.9,3.1,2.9, 3.5	-15.72, -18.36, -13.79, -13.34, -12.43	-12.82	-13.00	-13.79	
	1024	4	28	17.9	4.4	9	16.7,17.8,18.8,18.2, 15.7,14.8,20.2,22.6, 26.1	3.4,3.4,2.3,2, 3.0,2.4,5.0,5.3, 3.6	-13.84, -13.18, -12.57, -12.92, -14.04, -14.91, -12.12, -11.49, -10.92	-12.23	-12.12	-12.68	
	1145	3	30	21	10.8	4	17.6,16.3,19.9,23.4	4.1,4.2,8.4,10.5	-10.95, -12.32, -11.37, -10.94	-10.70	-10.79	-11.14	
	1110	1	66	19.3	8.6	4	22.2,8.8,16.5,14.1	9.6,10.7,5.2,2.1	-10.95, -10.65, -13.20, -12.80	-11.03	-11.08	-11.63	
	1191	2	38	20.3	7.5	6	20.6,18.5,18.0,17.3, 23.3,19.3	8.4,4.6,4.2,3.8, 10.1,4.4	-11.09, -11.84, -11.77, -12.02, -10.66, -12.05	-10.82	-10.83	-11.25	
	1251	16	60	23.6	10.1	2	24.4,20.6	10.1,6.1	-10.64, -11.20	-10.64	-10.65	-10.80	
	1280	11	39	23.3	9	4	23.5,22.6,23.6,21.1	10.5,7.5,6.3,7	-10.89, -11.12, -11.11, -11.59	-10.55	-10.67	-10.84	
	1209	2	39	21	8.6	6	18.4,16.6,21.0,24.2, 27.8,29.3	5.2,4.3,6.3,9.9, 6.5,6.0	-11.98, -11.88, -10.47, -10.77, -10.46, -10.25	-11.00	-10.88	-11.23	
AEJ RF18 4000-35000 35000-35800 35900-36500 36600-37300 39600-40300 40400-41000 41200-41800 42900-43200	561	6	62	15	3.6	1	16.3	4.8	-11.31	-12.36	-11.49	-14.76	
	754	0	43	19.2	5.7	15	150.0,159.16,5.17,7, 140.0,18.3,18.8,20.5, 22.0,21.9,19.2,19.7, 19.7,14.0,13.2	3.1,3.2,3.0,4.8, 2.5,2.3,1.4,4, 4.9,4.0,3.1,3.2, 2.7,2.3,2.0	-13.70, -13.46, -13.35, -12.40, -15.50, -12.52, -11.72, -11.58, -10.89, -11.02, -11.68, -11.76, -11.56, -15.76, -14.97	-11.01	-11.19	-11.72	
	852	0	41	21	6.4	9	23.5,22.6,24.2,21.7, 24.9,20.2,19.2,18.1, 17.4	4.4,3.8,4.3,3.4, 4.2,3.5,4.7,4.7, 4	-10.64, -10.86, -10.58, -11.09, -10.34, -11.49, -11.68, -11.91, -12.26	-10.75	-10.78	-10.97	
	977	3	38	20.7	5.3	15	22.8,21.9,22.1,21.3, 20.8,20.2,19.8,20.1, 19.2,19.0,18.2,17.6, 17.0,18.9,25.6	4.3,4.0,4.4,4.1, 4.2,4.5,4.5,5.0, 4.3,7.3,9.4,0, 3.6,4.8,3.9	-10.62, -10.86, -10.83, -11.22, -11.17, -11.42, -11.66, -11.32, -11.50, -11.48, -12.06, -12.40, -13.02, -11.65, -10.26	-10.73	-10.82	-11.04	
	661	4	26	15.6	3.7	3	15.7,13.3,16.5	3.8,2.0,2.3	-12.10, -11.34, -10.97, -12.19, -11.50	-12.54	-10.95	-13.66	
	812	5	42	19.4	5	5	18.2,20.6,22.4,17.1, 19.7	4.1,5.4,5.1,4.3, 3.7	-10.71, -10.69, -10.96	-11.33	-11.39	-11.64	
	946	11	38	23.5	5.2	3	24.1,23.6,22.7	5.7,5.0,4.8	-10.95, -10.78, -11.00, -11.26, -11.43, -10.23, -11.59	-10.56	-10.63	-10.74	
	897	5	28	22.9	6.9	7	23.6,24.6,22.6,22.1, 21.0,26.9,19.8	6.5,6.4,5.7,6.4, 6.0,8.7,6.0	-10.95, -10.78, -11.00, -11.26, -11.43, -10.23, -11.59	-10.69	-10.73	-10.88	

Table S1. Detailed information of all cloud transects analyzed for flights RF16 and RF18 from IOP1. These segments are from horizontal penetrations of the cloud at constant altitudes and an airspeed of $100\,m\,s^{-1}$ thereby spanning 10s of kilometers. The table lists the all calculated

metrics- the percent of holograms that have a similar shape to the global average (rounded to the nearest integer), the percent of classified holograms (rounded to the nearest integer), the distribution shape parameters (mean and interquartile range(IQR)) of the global average and also their mean values for the different local characteristic distributions, the mass autoconversion rates of the local hologram samples, their average and the rate for the mean size distribution.

Time (seconds after 00:00 UTC)	Altitude (meters)	Similarity to ensemble avg (percentage)	Classified holograms (above thresh) (percentage)	Global DSD properties Mean (micro-meters)	Global DSD properties IQR (micro-meters)	Clusters count	Local DSD properties- Mean (micro-meters)	Local DSD properties- IQR	Clusters	Mass autocorrelation rates (in log10 scale) (gcm ⁻³ s ⁻¹)	Uncs	Rate of avg. of local rates
AE1 RF19 41000-41500 42000-42500 42500-43000 43000-43500 43800-44200 44200-44400	2002	6	42	25.5	15.2	2	27.4,22.9	15.0,12.0	-9.35, -9.73	-9.49	-9.46	-10.16
	1829	6	24	23.4	12.4	5	27.7,32.4,24.1,21.7, 18.3	12.7,17.6,11.1,9.0, 7.1	-9.82, -9.53, -9.51, -10.54, -11.23	-9.97	-9.93	-10.52
	1681	7	38	23.8	12.3	3	26.7,16.8,15.2	13.4,4.2,2.9	-9.86, -11.21, -13.14	-9.95	-9.93	-10.58
	1675	5	50	24.1	12.5	4	19.9,24.1,28.1,30.7	8.7,10.9,15.4,14.9	-10.95, -10.76, -10.27, -10.07	-9.98	-10.19	-10.66
	1353	4	38	22	9.8	6	28.8,23.2,21.3,18.8, 15.7,38.4	9.1,9.6,8.7,6.1, 3.4,12.9	-10.39, -11.20, -10.13, -11.77, -13.27, -9.14	-9.86	-9.89	-10.89
	775	18	35	18.3	6.5	2	18.1,19.0	6.1,7.5	-11.94, -11.64	-10.23	-10.39	-11.77
AE2 RF04 42300-42800 4300-43500 43500-44200 44250-44500 44500-44600 44600-45300	1295	0	77	19.4	6.2	3	19.6,22.9,15.6	3.6,4.8,3.9	-11.54, -11.54, -11.80	-11.72	-11.64	-12.13
	1367	0	53	21.1	8.8	8	25.1,26.6,27.9,23.4, 19.0,18.4,17.2,15.5	5.4,5.9,5.8,7.0, 3.8,3.8,3.7,3.3	-11.19, -11.05, -9.58, -11.29, -12.38, -12.56, -13.09, -14.61	-10.19	-10.39	-11.43
	1366	0	42	18	6.1	16	22.3,21.6,21.0,20.1, 22.9,18.6,17.7,17.3, 16.8,16.3,15.9,15.2, 14.9,14.2,13.7,2.8	3.6,3.5,4.0,4.6, 3.4,3.0,3.5,3.9, 3.7,3.1,3.7,3.4, 3.1,3.1,2.7,3.8	-11.03, -11.20, -11.36, -11.63, -10.95, -12.29, -12.49, -12.54, -12.78, -13.13, -13.31, -14.03, -14.20, -15.06, -16.03, -10.74	-11.58	-11.54	-11.92
	1433	2	54	26.5	7.5	8	23.0,24.1,25.3,28.5, 29.6,30.5,31.1,21.5	4.4,4.6,5.1,6.1, 5.3,5.8,5.1,4.0	-10.71, -10.55, -10.28, -10.10, -10.02, -9.80, -9.45, -11.01	-9.96	-10.01	-10.17
	1539	7	42	23.9	4.5	3	23.6,24.0,24.5	4.2,4.2,4.5	-10.33, -10.25, -10.28	-10.26	-10.27	-10.29
	1549	0	45	28.2	7.8	10	24.3,25.7,27.5,28.2, 29.0,31.4,32.1,32.1, 33.0,34.8	4.4,5.1,3.1,3.2, 3.4,6.0,6.1,4.6, 6.0,5.9	-10.64, -10.44, -10.18, -10.03, -9.95, -9.68, -9.72, -9.71, -9.62, -9.51	-9.84	-9.88	-10.0

Table S2. Same as Table S1. but for flights RF19 and RF04 from IOP1 and IOP2 respectively.

References and Notes

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