Supplementary material

Results here primarily focus on sensitivity tests of the “large drop isolation trend”, which are shown in Table S1. Results from one sensitivity test are shown in Figure S3 (dependence on drop size threshold). Additional sensitivity tests are available upon request.

<table>
<tr><th colspan="8">Sensitivity Tests</th></tr>
<tr><th>Sensitivity test</th><th colspan="6">Conditions</th><th>Rationale/discussion</th></tr>
<tr><td rowspan="3">Test alternative version of Table 3 with slightly different thresholds</td><td>N</td><td><100</td><td>100–170</td><td>170–240</td><td>240–300</td><td>>300</td><td rowspan="3">Results are consistent with original Table 3. However, note conditions at low drop concentrations are approximately similar, where the isolated large drop trend is observed.</td></tr>
<tr><td>Min ψ for high DCF</td><td>2</td><td>3</td><td>4</td><td>5</td><td>6</td></tr>
<tr><td>Min shell size for low DCF</td><td>$r_{min}+\delta r=$ 0.12 cm</td><td>$r_{min}+\delta r=$ 0.1 cm</td><td>$r_{min}+\delta r=$ 0.09 cm</td><td>$r_{min}+\delta r=$ 0.09 cm</td><td>$r_{min}+\delta r=$ 0.09 cm</td></tr>
<tr><td>Test larger shell sizes</td><td colspan="6">$r_{min}+\delta r=$16cm, 17cm</td><td>-</td></tr>
<tr><td>Test for consideration of different small drop size threshold</td><td colspan="6">D=6 µm, 10 µm, 14 µm</td><td>- The HOLODEC detects drops as small as D=6 µm, but has improved detection for D>10 µm (references therein)</td></tr>
<tr><td rowspan="2">Test DCF>67th percentile for Monte Carlo – Hologram Comparisons</td><td colspan="2">Categories</td><td colspan="2">HILD</td><td colspan="2">OTHER</td><td rowspan="2">-</td></tr>
<tr><td colspan="2">Percentiles</td><td colspan="2">D: 25-30 µm > 95th
D: 30-37.5 µm > 95th
D: 37.5-50 µm > 90th</td><td colspan="2">D: 25-30 µm < 85th
D: 30-37.5 µm < 85th
D: 37.5-50 µm < 85th</td></tr>
<tr><td>Adjust $N_{DCF\_category}$ lower bound</td><td colspan="6">Tested possibilities of $N_{DCF\_category}$ threshold of 1,2,…,10</td><td>Results are consistent for $N_{DCF\_category}$ = 2–10</td></tr>
<tr><td rowspan="2">Extend HOLODEC sample volume</td><td colspan="3">Inner sample volume dimensions</td><td colspan="3">Guard rail dimensions</td><td rowspan="2">-</td></tr>
<tr><td colspan="3">0.7 cm × 0.7 cm × 10.1 cm</td><td colspan="3">0.4 cm × 0.4 cm × 6.1 cm</td></tr>
</table>

Table S1: List of four sensitivity tests. Results from the consideration of the smallest drop size threshold are shown in Figure S1 below.

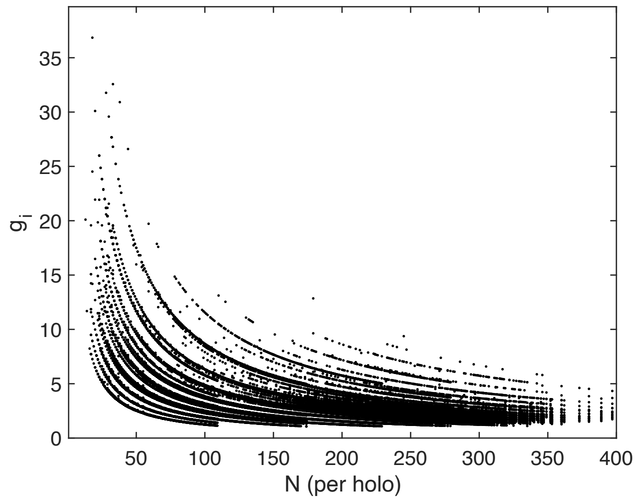

Figure S1: Plot of hologram drop concentration (per sample volume) related to g_i (Eq. 2). All drops used in the analysis as defined in Sections 2 and 3 are included here. Note that only the maximum g_i are shown amongst all shell sizes for a given drop, and a minimum number of neighboring drops is required for a given drop concentration. This is what produces characteristic cut-offs in the datapoints at N=110, 175, etc., which correspond to drop concentration thresholds in Step 3 of Table 2.

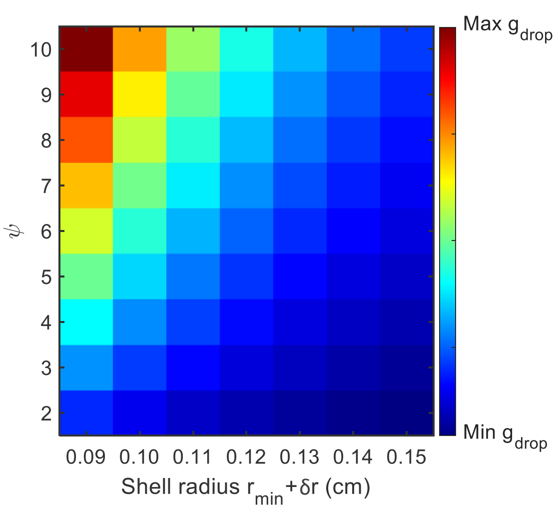

Figure S2: Ranked g_{drop} for varying shell sizes and number of drops within the respective shells.

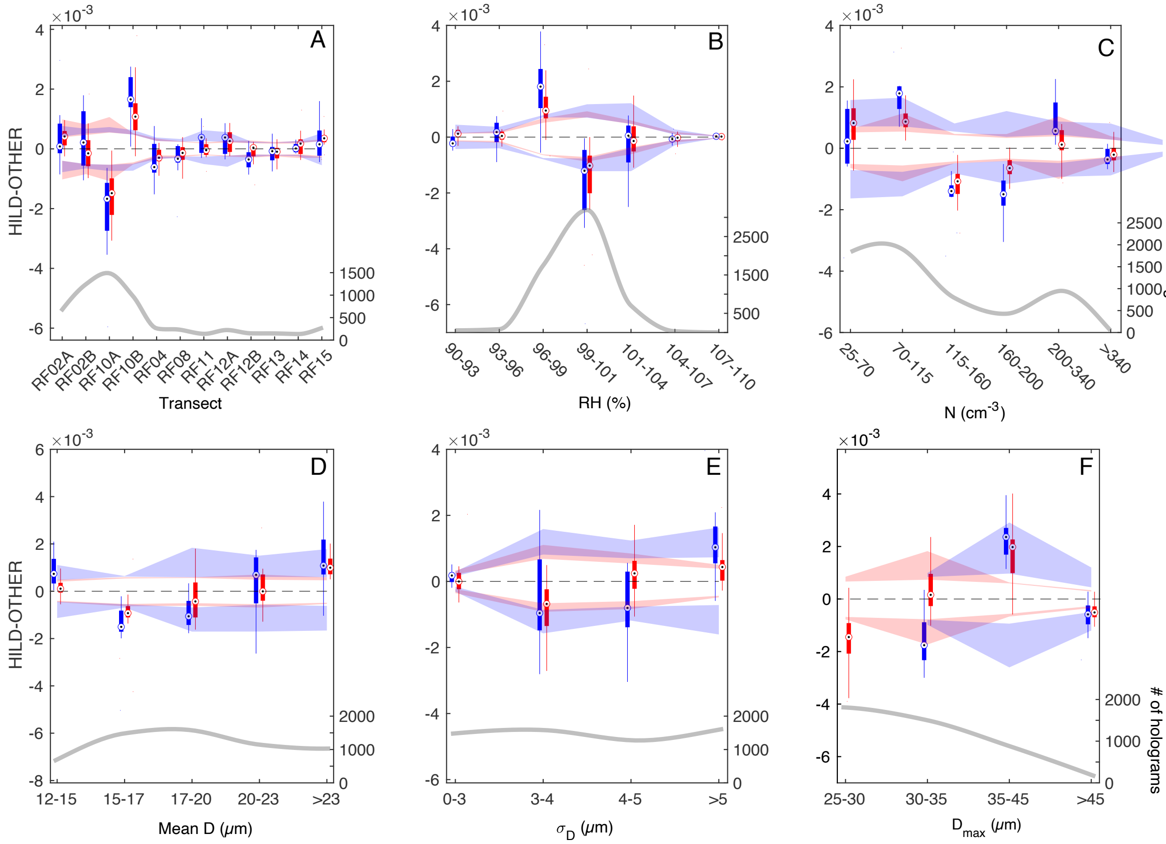

Figure S3: Identical to Figure 7, but using data from all flight legs in Table 1 and not just RF02A,B and RF10A,B.

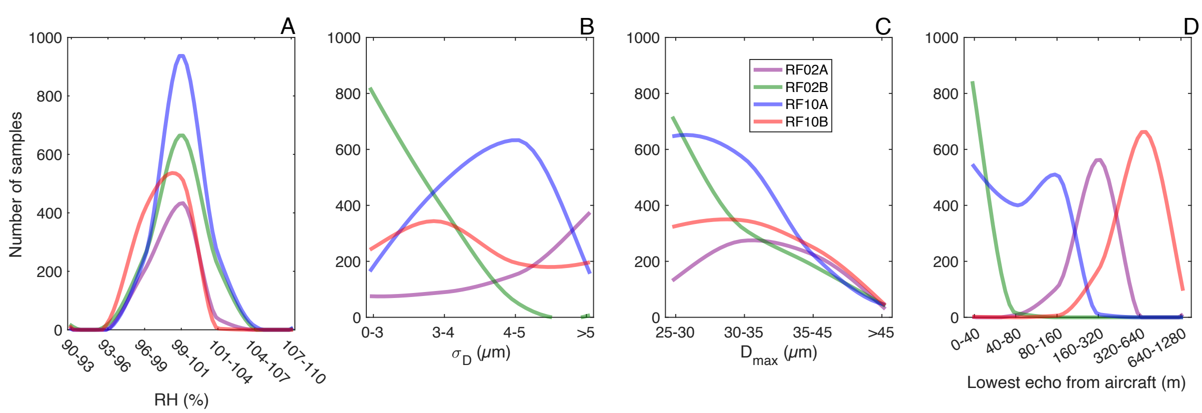

Figure S4: Histograms of *RH* (A), σ_D (B), D_{max} (C), and lowest detected cloud condensate (D) from the four flight legs evaluated in Section 4.2 (colored lines). Results in D) are reported as absolute distance from the aircraft, whereas the corresponding plot in Figure 8D ranks these values over each respective flight leg. A cubic interpolation is applied to the histograms.

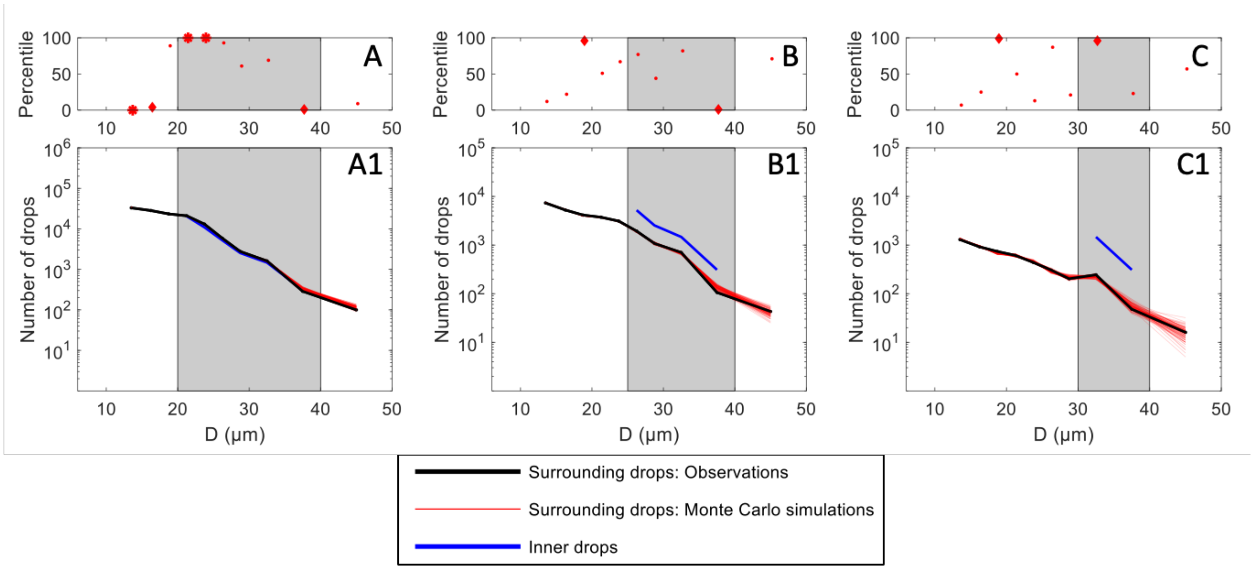

Figure S5: Plots showing percentiles (i.e., likelihoods) that drops of varying sizes will neighbor drops having diameters between 20–40 µm (A), 25–40 µm (B) and 30–40 µm (C). Frequency distributions of the neighboring drops (black lines) and the respective drop ranges (blue lines) are shown in A1, B1 and C1. Drops within these size ranges are termed inner drops. Results from neighboring drops are only shown for high DCF, and from the shell sizes which correspond to those of the maximum g_{drop}. Percentiles are computed similarly to the Monte Carlo-DCF methodology, whose simulated frequency distributions are shown by the thin red lines in A1, B1 and C1. Shaded regions correspond to the inner drops' size range.

Greater likelihoods of drops neighboring similarly sized drops are highlighted here, most notably for drops with diameters from 20–40 µm (S2A), where the highest likelihoods peak at drop sizes closest to D=20 µm (having the greatest occurrence frequencies amongst the size range).