Supplemental material:

Pilot preparation for the comparison flight:

Both aircraft cannot appear at the same location at the same time due to safety concerns. Thus, the approval of a formation (inter-comparison) flight was acquired six months before the campaign through DOE Pacific Northwest Site Office (PNSO) and the Office of Aviation Management (OAM). Essential risk mitigation was also discussed and approved by the Pacific Northwest National Laboratory Aviation Risk Management Committee (PNNL ARMC). During the IOP, both aircraft crew and scientists teams set up a meeting to discuss the potential flight plan. After the flight plan was formed, both pilots briefed the plan to the Brazilian Air Force (BAF) and Airport Traffic Control (ATC). The clear-sky flight would be under Visual Flight Rules (VFR), which means good weather and no cloud and pilots communicate with each other using an air-to-air frequency. For coordinated flights in cloudy condition, the G1 and the HALO were both on Instrument Flight Rules (IFR) flight plan.

The coordinated flight on October 1, 2014, was initially designed to be a coordinated flight under a cloudy condition, which means the G1 and the HALO flew the same flight leg with at least 300 m altitude offset and at least 5 minutes apart. However, the coordinated two flight legs (~900 m and ~1200 m) are all below the cloud. Thus, the comparison focus on the correlation between two aircraft measurements, not vertical profiling.

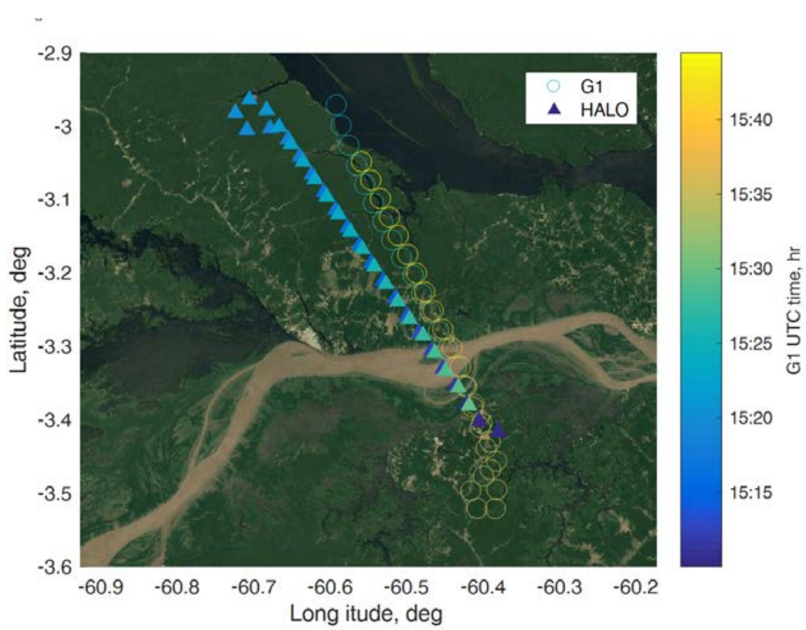


Figure S1. Time colored flight track of the G1 (circle) and the HALO (triangle) on October 1, 2014, during a cloudless coordinated flight.

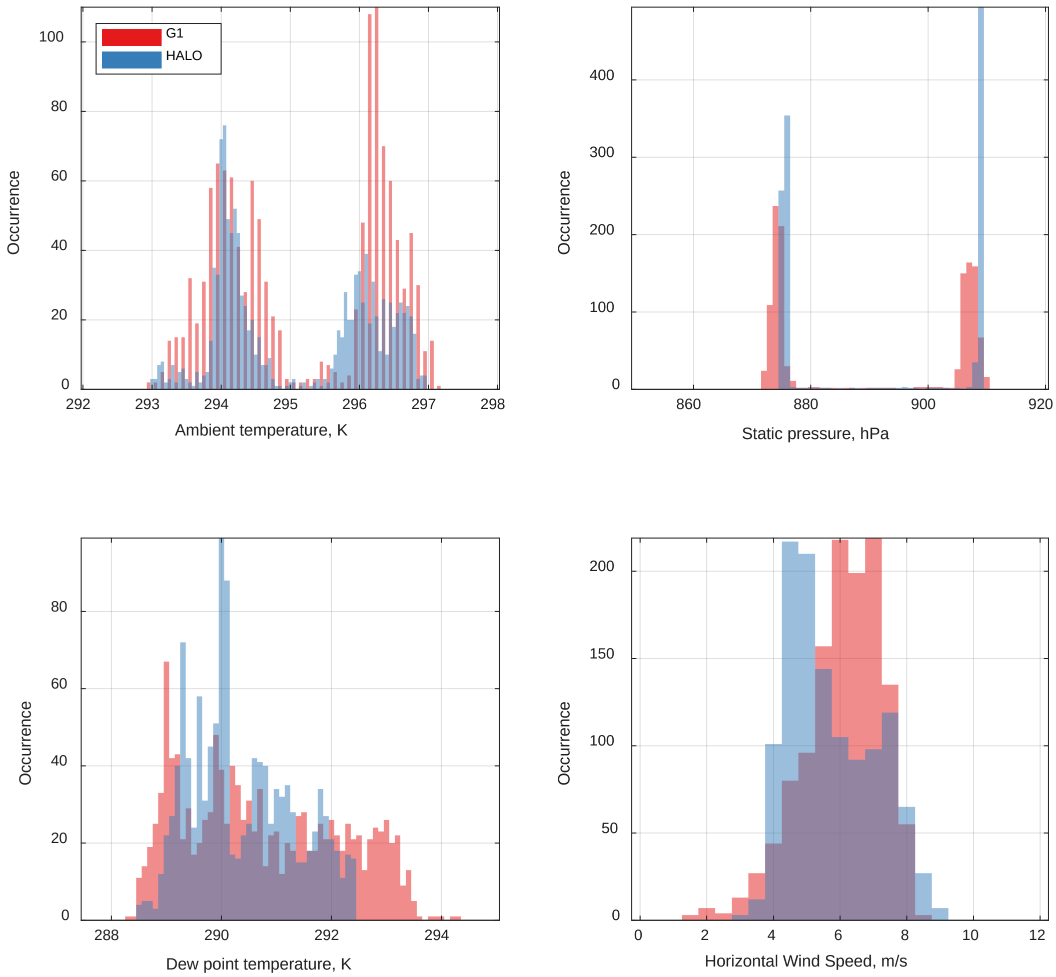
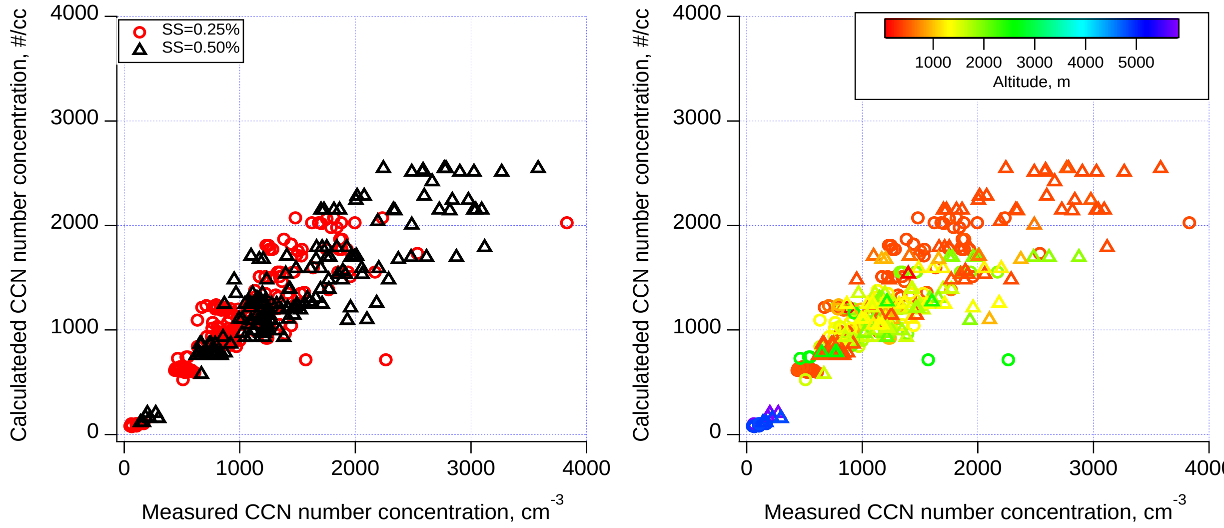


Figure S2, Atmospheric parameters observed by the G1 and the HALO on October 1, 2014.

To further examine the relative importance of mixing state and chemical composition, the CCN concentrations were calculated from aerosol particle size distribution, and chemical composition measured onboard the G1. The calculation was based on *κ*-Köhler parameterization, (Kohler, 1936; Petters and Kreidenweis, 2007, 2008, 2013) and the detail of the approach was described by Mei et al., 2013b. For the flight on September 9, 2017, the CCN number concertation calculated from the G1 UHSAS size distribution and chemical composition exhibits underestimation at a supersaturation of 0.5% (Fig. 3S(a)) and when the altitude is below 1000 m (Fig 3S(b)). This underestimation suggests that the UHSAS size range (90-500 nm) did not fully cover the aerosols with the critical activation diameter ($D_{p,50}$) at high supersaturation. Thus, the FIMS measurements onboard the G1 was the more appropriate size distribution for both the CCN closure study.



(a) (b)

Figure S3 Comparison of calculated CCN with measured CCN using the averaged 1 min measurements from the G1: (a) colored by different supersaturations. (b) colored by different altitude. (Note that both plots used the calculated CCN number concentration from UHSAS size distribution.)

The CCN concentration calculated using the size distribution from FIMS agrees well with the measurement (Fig. S4). The scattering of the comparison data in Figure 15 is likely due to the chemical composition and mixing state effect on aerosol hygroscopicity.

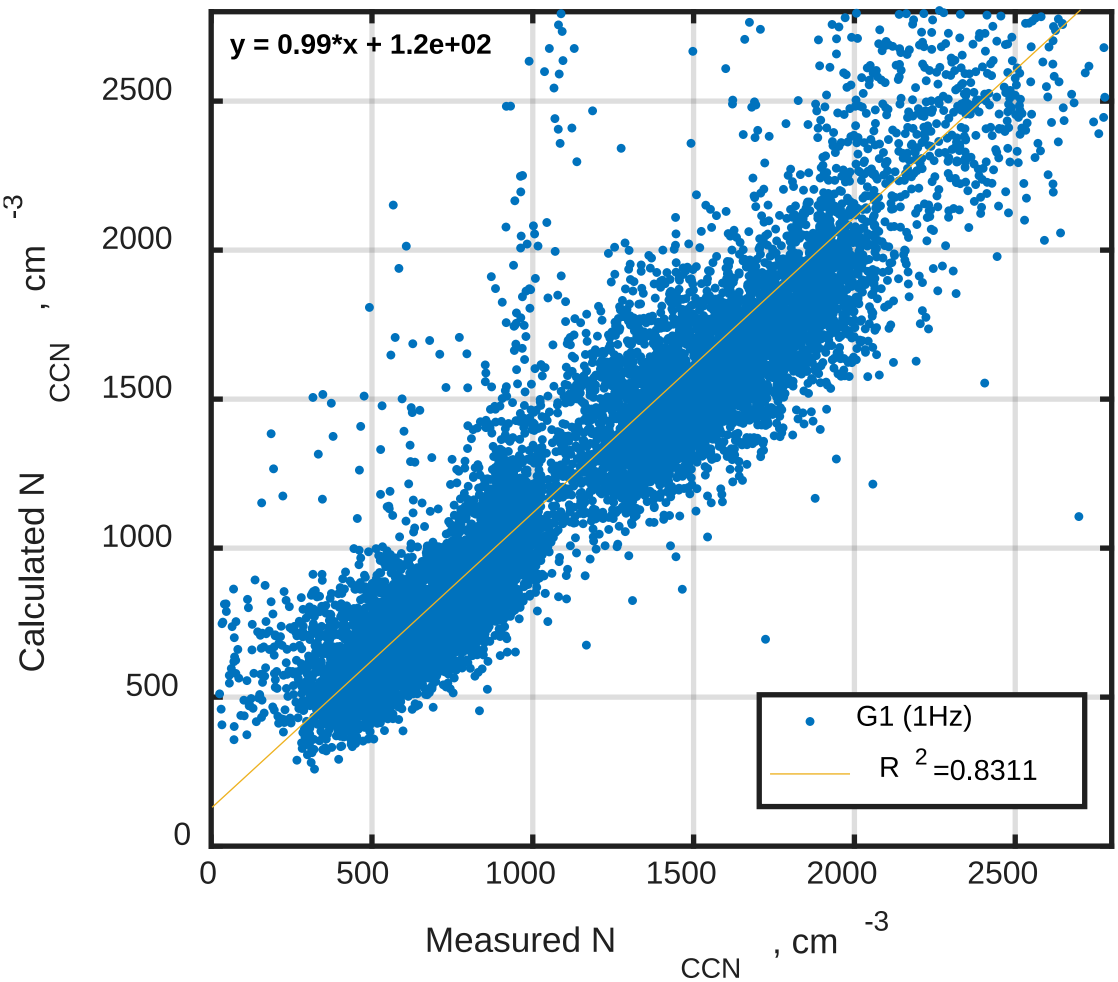


Figure S4. The scatter plot of the calculated CCN number concentration using FIMS size distribution compared with the measured CCN number concentration

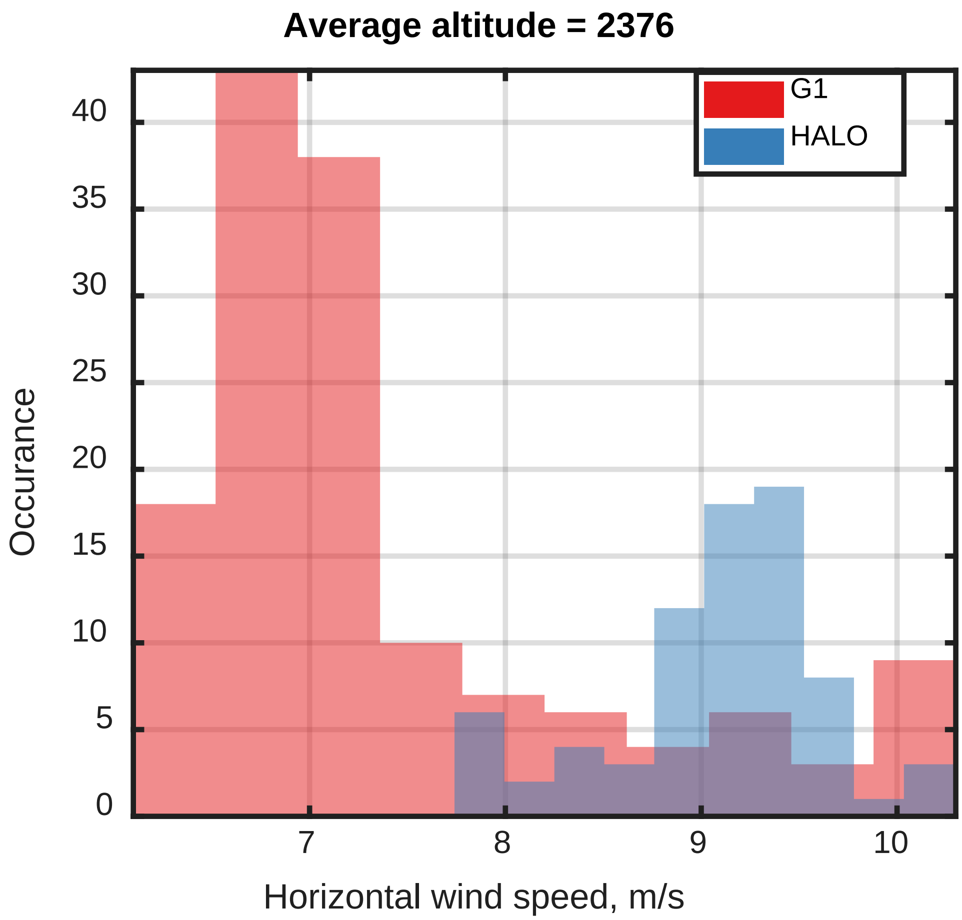
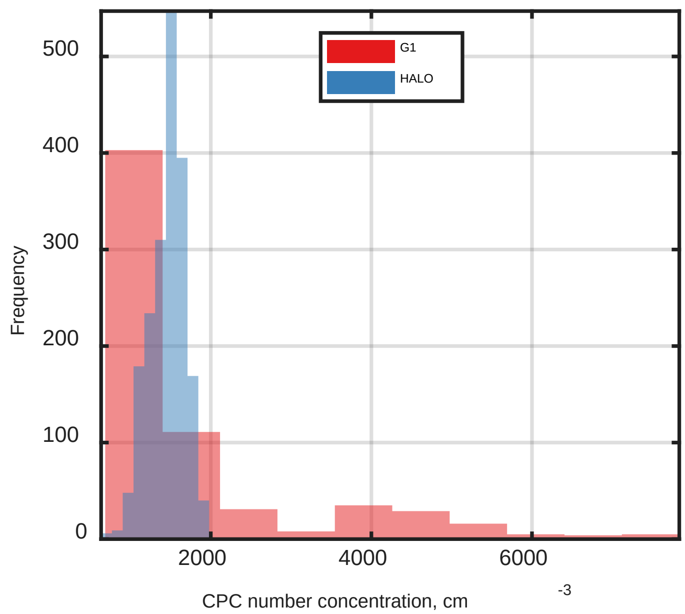
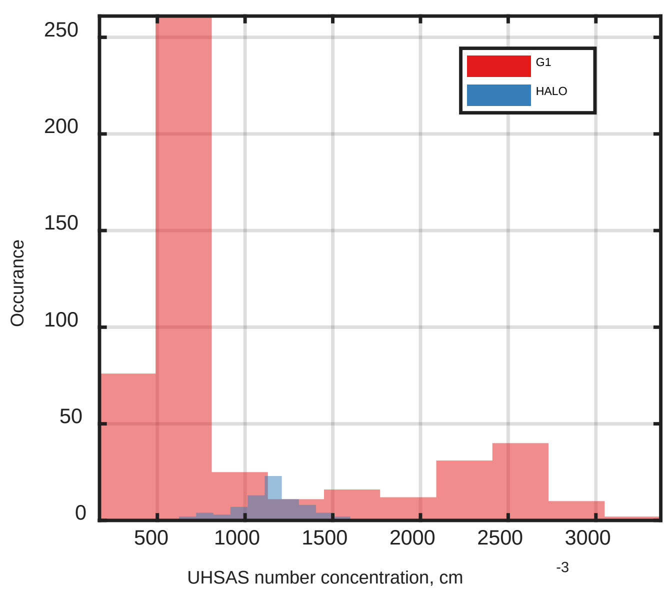


Figure S5. Horizontal wind speed between 2000-3000 m altitude on September 21, 2014.

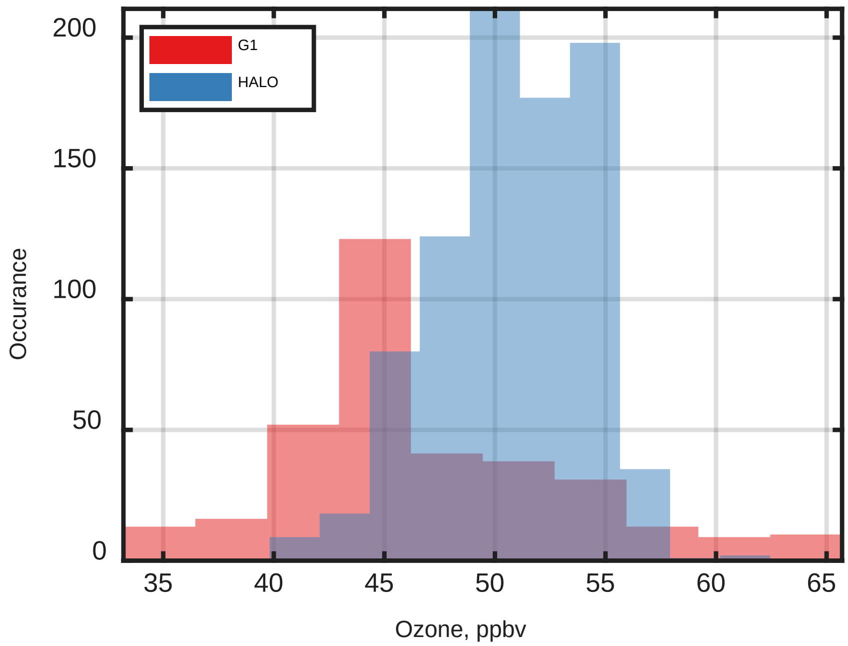


(a)

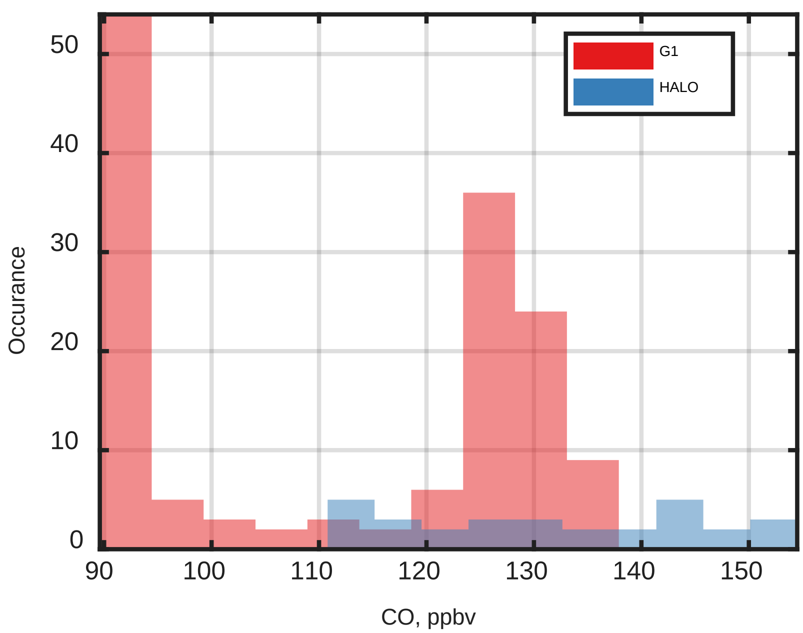


(b)

Figure S6. The total aerosol particles number concentration between 2000-3000 m altitude on September 21, 2014: (a) CPC measurement; (b) UHSAS measurement.



(a)



(b)

Figure S7. The trace gas concentration between 2000-3000 m altitude on September 21, 2014: (a) Ozone measurement; (b) CO measurement.

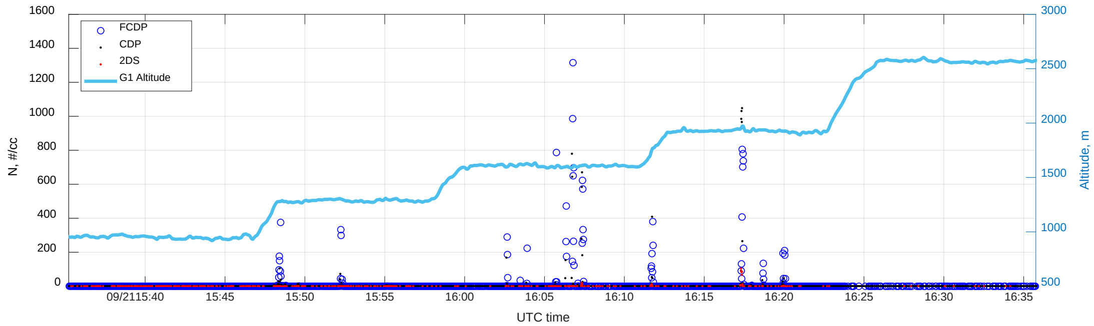


Figure S8. The cloud droplet number concentration from the G1 aircraft on September 21.

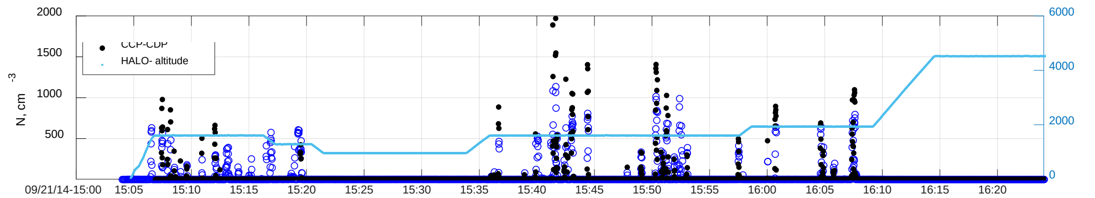
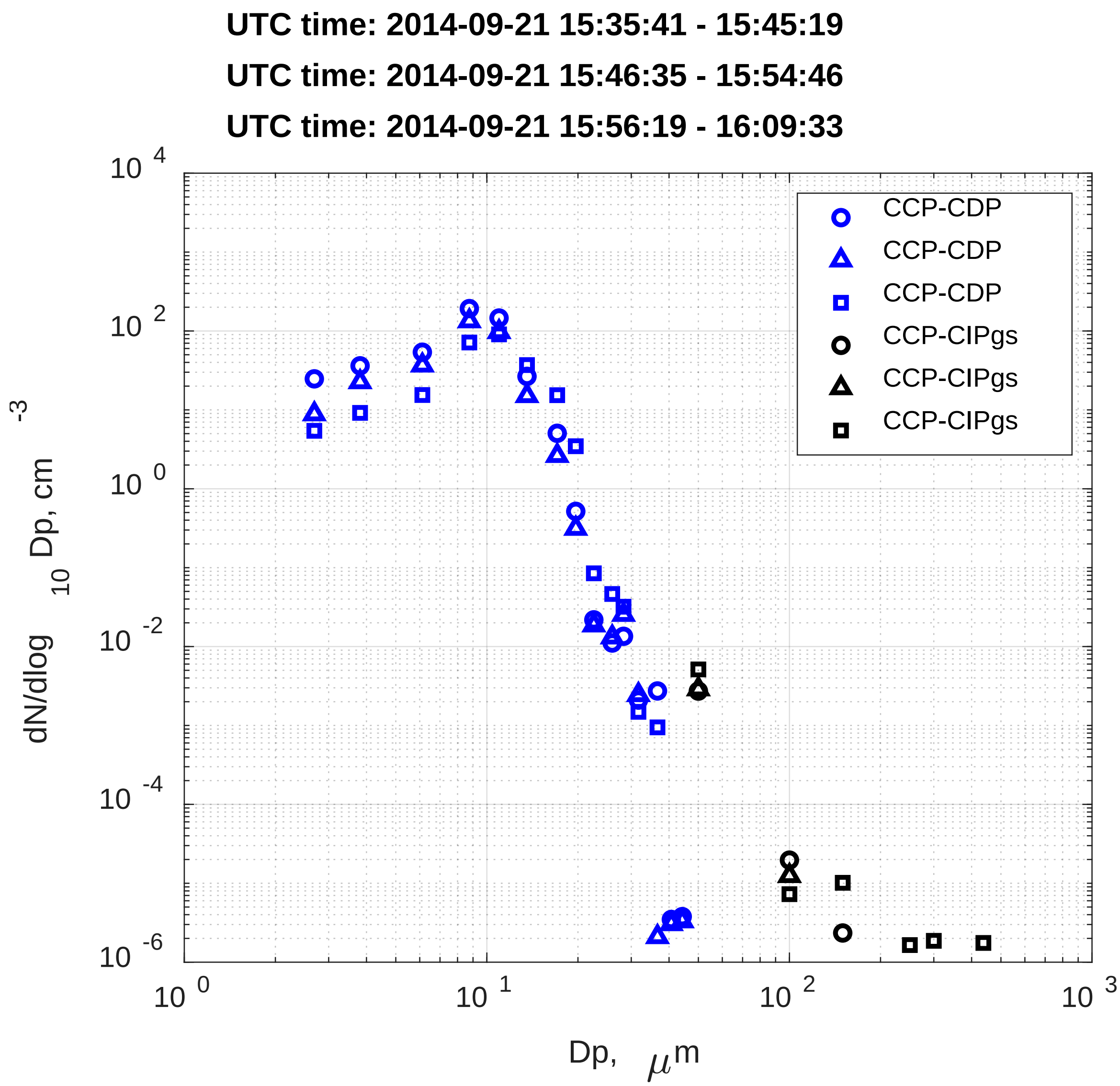


Figure S9. The cloud droplet number concentration from HALO on September 21.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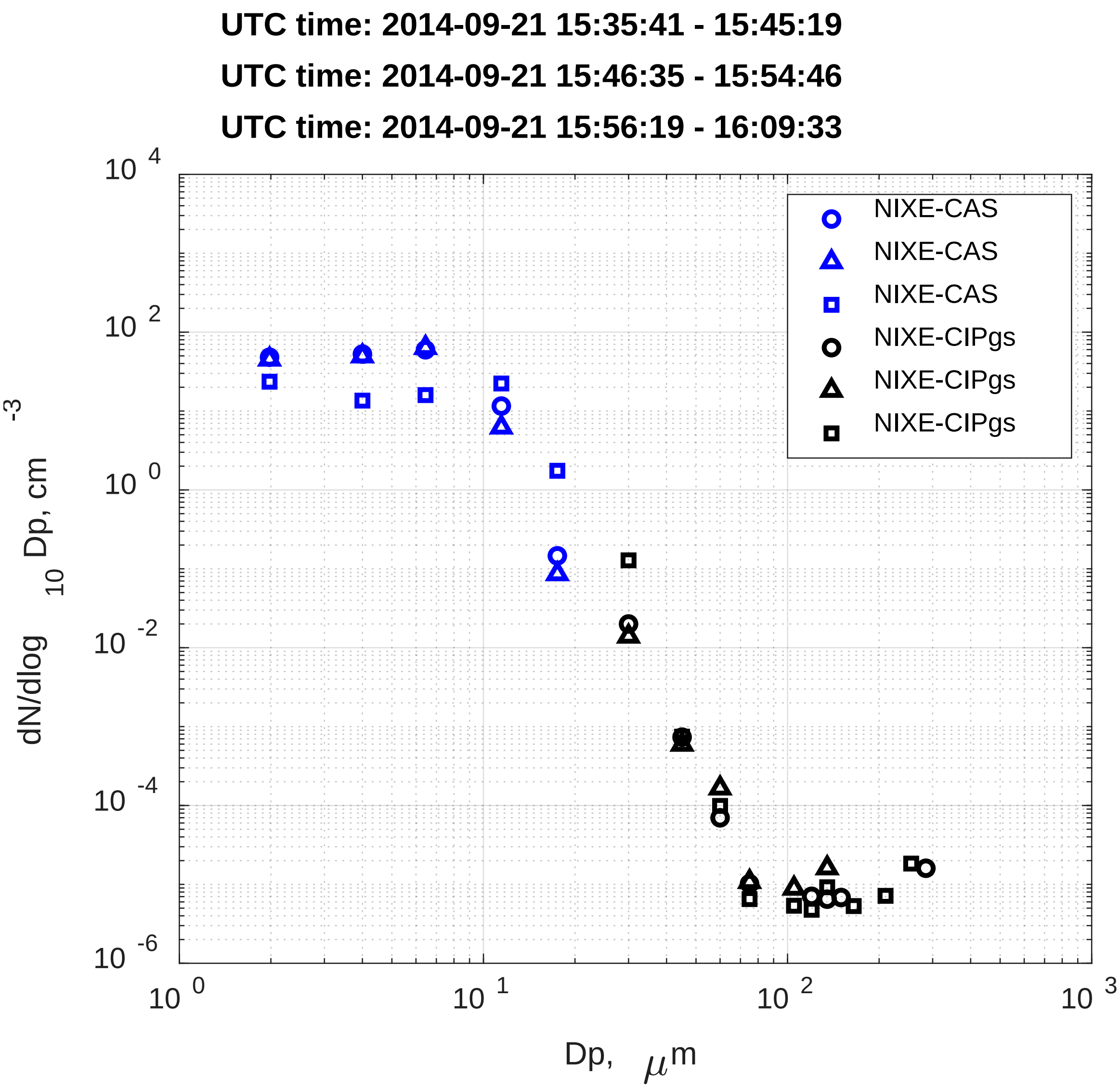
UTC time: 2014-09-21 15:46:35 - 15:54:46
UTC time: 2014-09-21 15:56:19 - 16:09:33
CCP-CDP
CCP-CDP
CCP-CDP
CCP-CIPgs
CCP-CIPgs
CCP-CIPgs
dN/dlog10 Dp, cm-3
Dp, μm

69

70 (a)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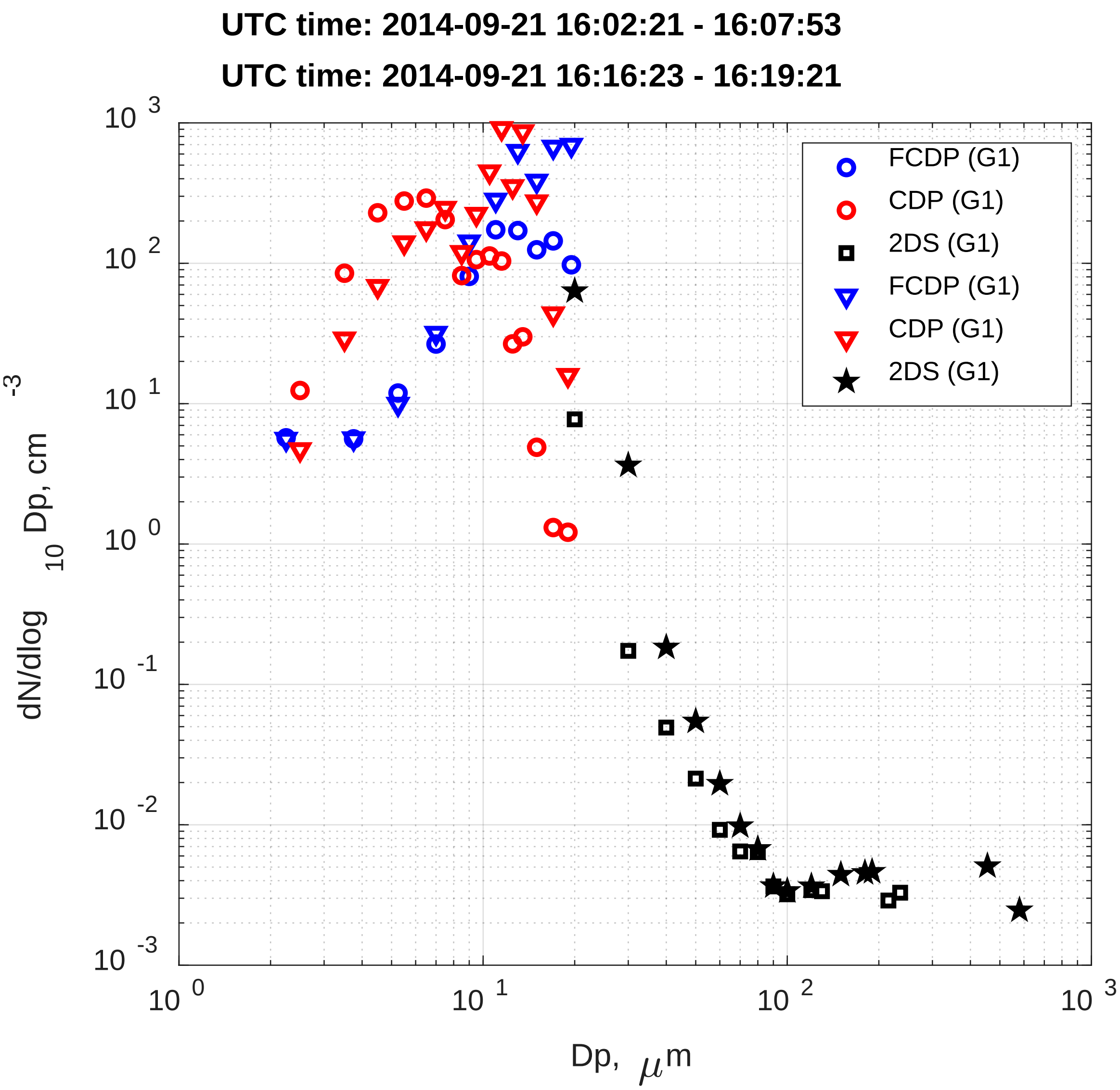
dN/dlog10 Dp, cm-3
Dp, μm

(b)



(c)

Figure S10. The averaged cloud droplet size distributions from HALO on September 21, (a) CCP probes; (b) NIXE-CAPS probes; (c) Cloud probes on board the G1.