



1	Comparison of Aircraft Measurements during GoAmazon2014/5 and
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26	Abstract. The indirect effect of atmospheric aerosol particles on the Earth's radiation balance
27	remains one of the most uncertain components affecting climate change throughout the industrial
28	period. This issue is partially a result of the incomplete understanding of aerosol-cloud
29	interactions. One objective of the GoAmazon2014/5 and ACRIDICON-CHUVA projects was to
30	improve the understanding of the influence of the emissions of the tropical megacity of Manaus





31 (Brazil) on the surrounding atmospheric environment of the rainforest and to investigate its role in 32 the life cycle of convective clouds. During one of the intensive observation periods (IOPs) in the dry season from September 1 to October 10, 2014, comprehensive instrument suites collected data 33 from several ground sites. In a coordinated way, the advanced suites of sophisticated instruments 34 were deployed in situ both from the U.S. Department of Energy Gulfstream-1 (G1) aircraft and 35 the German High Altitude and Long-Range Research Aircraft (HALO) during three coordinated 36 flights on September 9, 21, and October 1. Here we report on the comparison of measurements 37 collected by the two aircraft during these three flights. Such comparisons are difficult to obtain, 38 but they are essential for assessing the data quality from the individual platforms and quantifying 39 their uncertainty sources. Similar instruments mounted on the G1 and HALO collected vertical 40 profile measurements of aerosol particles number concentration and size distribution, cloud 41 condensation nuclei concentration, ozone, and carbon monoxide concentration, cloud droplet size 42 distribution, and downward solar irradiance. We find that the above measurements from the two 43 44 aircraft agreed within the range given by the measurement uncertainties. Aerosol chemical composition measured by instruments on HALO agreed with the corresponding G1 data collected 45 at high altitudes only. Furthermore, possible causes of discrepancies between the data sets 46 47 collected by the G1 and HALO instrumentation are addressed in this paper. Based on these results, 48 criteria for meaningful aircraft measurement comparisons are discussed. 49

50 1. Introduction

51 Dominated by biogenic sources, the Amazon basin is one of the few remaining continental regions where atmospheric conditions realistically represent those of the pristine or pre-industrial 52 era (Andreae et al., 2015). As a natural atmospheric "chamber", the area around the urban region 53 54 of Manaus in central Amazonia is an ideal location for studying the atmosphere under natural 55 conditions as well as under conditions influenced by human activities and biomass burning events (Andreae et al., 2015; Artaxo et al., 2013; Davidson et al., 2012; Keller et al., 2009; Kuhn et al., 56 2010; Martin et al., 2016b; Pöhlker et al., 2018; Poschl et al., 2010; Salati and Vose, 1984). The 57 Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5) campaign was 58 conducted in 2014 and 2015 (Martin et al., 2017; Martin et al., 2016b). The primary objective of 59 60 GoAmazon2014/5 was to improve the quantitative understanding of the effects of anthropogenic





influences on atmospheric chemistry and aerosol-cloud interactions in the tropical rainforest area.
During the dry season in 2014, the ACRIDICON (Aerosol, Cloud, Precipitation, and Radiation
Interactions and Dynamics of Convective Cloud Systems)-CHUVA (Cloud Processes of the Main
Precipitation Systems in Brazil) campaign also took place to study tropical convective clouds and
precipitation over Amazonia (Wendisch et al., 2016).

A feature of the GoAmazon 2014/5 field campaign was the design of the ground sites' 66 location, which uses principles of Lagrangian sampling to align the sites with the Manaus pollution 67 plume (Figure 1: Source location - Manaus (T1 site), and downwind location - Manacapuru (T3 68 site)). The ground sites were overflown with the low-altitude U.S. Department of Energy (DOE) 69 Gulfstream-1 (G1) aircraft and the German High Altitude and Long Range Research Aircraft 70 (HALO). These two aircraft are among the most advanced in atmospheric research, deploying 71 72 suites of sophisticated and well-calibrated instruments (Schmid et al., 2014; Wendisch et al., 73 2016). The pollution plume from Manaus was intensively sampled during the G1 and HALO 74 flights and also by the DOE Atmospheric Radiation Measurement (ARM) program Mobile Aerosol Observing System and ARM Mobile Facility located at one of the downwind surface sites 75 (T3 site-70 km west of Manaus). The routine ground measurements with coordinated and intensive 76 77 observations from both aircraft provided an extensive data set of multi-dimensional observations in the region, which serves i) to improve the scientific understanding of the influence of the 78 emissions of the tropical megacity of Manaus (Brazil) on the surrounding atmospheric 79 environment of the rainforest and ii) to understand the life cycle of deep convective clouds and 80 81 study open questions related to their influence on the atmospheric energy budget and hydrological 82 cycle.

As more and more data sets are merged to link the ground-based measurements with 83 aircraft observations, and as more studies focus on the spatial variation and temporal evolution of 84 85 the atmospheric properties, it is critical to quantify the uncertainties ranges when combining the data collected from the different platforms. Due to the challenges of airborne operations, especially 86 when two aircraft are involved in data collection in the same area, direct comparison studies are 87 rare. However, this type of study is critical for further combining the datasets between the ground 88 89 sites and aircraft. Thus, the main objectives of the study herein are to demonstrate how to achieve 90 meaningful comparisons between two moving platforms, to conduct detailed comparisons between data collected by two aircraft, to identify the potential measurement issues, to quantify 91





92 reasonable uncertainty ranges of the extensive collection of measurements, and to evaluate the 93 measurement sensitivities to the temporal and spatial variance. The comparisons and the related 94 uncertainty estimations quantify the current measurement limits, which provide realistic 95 measurement ranges to climate models as initial conditions to evaluate their output.

The combined GoAmazon2014/5 and ACRIDICON-CHUVA field campaigns not only 96 97 provide critical measurements of aerosol and cloud properties in an under-sampled geographic region but also provide a unique opportunity to understand and quantify the quality of these 98 measurements using closely orchestrated comparison flights. The comparisons between the 99 100 measurements from similar instruments on the two research aircraft can be used to identify potential measurement issures and quantify the uncertainty range of the field measurements, which 101 include primary meteorological variables (Section 3.1), trace gases concentrations (Section 3.2), 102 103 aerosol particle properties (number concentration, size distribution, chemical composition, and 104 microphysical properties) (Section 3.3), cloud properties (Section 3.4), and downward solar 105 irradiance (Section 3.5). We evaluate the consistency between the measurements aboard the two aircraft for a nearly full set of gas, aerosol particle, and cloud variables. Results from this 106 comparison study provide the foundation not only for assessing and interpreting the observations 107 108 from multiple platforms (from the ground to low altitude, and then to high altitude) but also for providing high-quality data to improve the understanding of the accuracy of the measurements 109 related to the effects of human activities in Manaus on local air quality, terrestrial ecosystems in 110 rainforest, and tropical weather. 111

112 **2. Measurements**

113 2.1 Instruments

The ARM Aerial Facility deployed several in situ instruments on the G1 to measure atmospheric state parameters, trace gases concentrations, aerosol particle properties, and cloud characteristics (Martin et al., 2016b; Schmid et al., 2014). The instruments installed on HALO covered measurements of meteorological, chemical, microphysical, and radiation parameters. Details of measurements aboard HALO are discussed in the ACRIDICON-CHUVA campaign overview paper (Wendisch et al., 2016). The measurements compared between the G1 and HALO are listed in Table 1.

121 2.1.1 Atmospheric parameters





All G1 and HALO meteorological sensors were routinely calibrated to maintain measurement accuracy. The G1 primary meteorological data were provided at one-second time resolution based on the standard developed by the Inter-Agency Working Group for Airborne Data and Telemetry Systems (Webster and Freudinger, 2018). For static temperature measurement, the uncertainty given by the manufacturer (Emerson) is ± 0.1 K, and the uncertainty of the field data is ± 0.5 K. The static pressure had a measurement uncertainty of 0.5 hPa. The standard measurement uncertainties were ± 2 K for the chilled mirror hygrometer and 0.5 ms⁻¹ for wind speed.

129 On HALO, primary meteorological data were obtained from the Basic HALO Measurement and Sensor System (BAHAMAS) at one-second time resolution. The system acquired data from 130 airflow and thermodynamic sensors and from the aircraft avionics and a high-precision inertial 131 reference system to derive the basic meteorological parameters like pressure, temperature, the 3D 132 133 wind vector, aircraft position, and attitude. Water vapor mixing ratio and further derived humidity 134 quantities were measured by the Sophisticated Hygrometer for Atmospheric Research (SHARC) based on direct absorption measurement by a tunable diode laser (TDL) system. The absolute 135 accuracy of the primary meteorological data was 0.5 K for air temperature, 0.3 hPa for air pressure, 136 0.4-0.6 ms⁻¹ for wind, and 5% (±1 ppm) for water vapor mixing ratio. All sensors were routinely 137 calibrated and traceable to national standards (Giez et al., 2017; Krautstrunk and Giez, 2012). 138

139 2.1.2 Gas phase

140 Constrained by data availability, this comparison of trace gas measurements is focused on carbon monoxide (CO) and ozone (O_3) concentrations. Those measurements were made aboard 141 the G1 by a CO/N₂O/H₂O instrument (Los Gatos Integrated Cavity Output Spectroscopy 142 instrument model 907-0015-0001), and an Ozone Analyzer (Thermo Scientific, Model 49i), 143 respectively. The G1 CO analyzer was calibrated for response daily by NIST-traceable commercial 144 standards before the flight. Due to the difference between laboratory and field conditions, the 145 146 uncertainty of CO measurement is about ±5% for one-second sampling periods, An ultra-fast carbon monoxide monitor (Aero Laser GmbH, AL5002) was deployed on HALO. The detection 147 of CO is based on a vacuum-ultraviolet-fluorimetry, employing the excitation of CO at 150 nm 148 and the precision is 2 ppb, and the accuracy is about 5%. The ozone analyzer measures ozone 149 150 concentration based on the absorbance of ultraviolet light at a wavelength of 254 nm. The ozone 151 analyzer (Thermo Scientific, Model 49c) in the HALO payload is very similar to the one on the





G1 (Model 49i), with an accuracy greater than 2 ppb or about $\pm 5\%$ for four-second sampling periods. The G1 ozone monitor was calibrated at the New York State Department of Environmental Conservation testing laboratory at Albany.

155 2.1.3 Aerosol

156 Aerosol number concentration was measured by different condensation particle counters 157 (CPCs) on the G1 (TSI, CPC 3010) and HALO (Grimm, CPC model 5.410). Although two CPCs were from different manufacturers, they were designed using the same principle, which is to detect 158 particles by condensing butanol vapor on the particles to grow them to a large enough size that 159 160 they can be counted optically. Both CPCs were routinely calibrated in the lab and reported the data at one-second time resolution. The HALO CPC operated at 0.6-1 L min⁻¹, with a nominal cutoff 161 of 4 nm. Due to inlet losses, the effective cutoff diameter increases to 9.2 nm at 1000 hPa, and 162 11.2 nm at 500 hPa (Andreae et al., 2018; Petzold et al., 2011). The G1 CPC operated at 1 L min-163 164 ¹ volumetric flow rate and the nominal cut-off diameter D_{50} measured in the lab was ~10 nm. During a flight, the cut-off diameter may vary due to tubing losses, which contribute less than 10 165 166 % uncertainty to the comparison between two CPC concentrations.

167 Two instruments deployed on the G1 measured aerosol particle size distribution. a Fast 168 Integrated Mobility Spectrometer (FIMS) inside of the G1 cabin measured the aerosol mobility size from 15 to 400 nm (Kulkarni and Wang, 2006a, b; Olfert et al., 2008; Wang, 2009). The 169 ambient aerosol particles were charged after entering the FIMS inlet and then separated into 170 different trajectories in an electric field based on their electrical mobility. The spatially separated 171 particles grow into super-micrometer droplets in a condenser where supersaturation of the working 172 fluid is generated by cooling. At the exit of the condenser, a high-speed charge-coupled device 173 camera captures the image of an illuminated grown droplet at high resolution. In this study, we 174 used the FIMS 1 Hz data for comparison. The size distribution data from FIMS were smoothed. 175 176 Aside from the FIMS, The airborne version of the Ultra High Sensitivity Aerosol Spectrometer (UHSAS) was deployed on G1 and HALO. The G1 and HALO UHSAS were manufactured by 177 the same company, and both were mounted under the wing on a pylon. UHSAS is an optical-178 scattering, laser-based particle spectrometer system. The size resolution is around 5% of the 179 180 particle size. The G1 UHSAS typically covered a size range of 60 nm to 1000 nm. HALO UHSAS 181 covered 90 nm to 500 nm size range for the September 9 flight.





182 Based on operating principles, FIMS measures aerosol electrical mobility size and UHSAS 183 measures aerosol optical equivalent size. Thus, the difference in the averaged size distributions from those two types of instruments might be linked to differences in their underlying operating 184 185 principles, such as the assumption in the optical properties of aerosol particles. The data processing in the G1 UHSAS assumed that the particle refractive index is similar to ammonium sulfate (1.55), 186 which is larger than the average refractive index (1.41-0.013i) from a previous Amazon study 187 (Guyon et al., 2003). The HALO UHSAS was calibrated with polystyrene latex spheres, which 188 189 has a refractive index about 1.572 for the UHSAS wavelength of 1054 nm. The uncertainty due to 190 the refraction index can lead to up to 10% variation in UHSAS measured size (Kupc et al., 2018). 191 Also, the assumption of spherical particles affects the accuracy of UHSAS sizing of ambient 192 aerosols.

193 The chemical composition of submicron non-refractory (NR-PM₁) organic and inorganic 194 (sulfate, nitrate, ammonium) aerosol particles was measured using a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) aboard the G1 (DeCarlo et al., 2006; Jayne et al., 2000; 195 Shilling et al., 2018; Shilling et al., 2013). Based on the standard deviation of observed aerosol 196 mass loadings during filter measurements, the HR-ToF-AMS detection limits for the average time 197 of thirteen seconds are approximately 0.13, 0.01, 0.02, 0.01 (3σ values) µg m⁻³ for organic, sulfate, 198 199 nitrate, and ammonium, respectively (DeCarlo et al., 2006). A Compact Time-of-flight Aerosol Mass Spectrometer (C-ToF-AMS) was operated aboard HALO to investigate the aerosol 200 composition. Aerosol particles enter both the C-ToF-AMS and HR-ToF-AMS via constant 201 pressure inlets controlling the volumetric flow into the instrument, although the designs of the 202 inlets are somewhat different (Bahreini et al., 2008). The details about the C-ToF-AMS operation 203 and data analysis are reported in Schulz's paper (Schulz et al., 2018). The overall accuracy has 204 been reported as ~30 % for both AMS instruments (Alfarra et al., 2004; Middlebrook et al., 2012). 205 Data presented in this section were converted to the same condition as the HALO AMS data, which 206 207 is 995 hPa and 300 K.

The number concentration of cloud condensation nuclei (CCN) was measured aboard both aircraft using the same type of CCN counter from Droplet Measurement Technologies (DMT, model 200). This CCN counter contains two continuous-flow, thermal-gradient diffusion chambers for measuring aerosols that can be activated at constant supersaturation. The supersaturation is created by taking advantage of the different diffusion rates between water vapor





and heat. After the supersaturated water vapor condenses on the CCN in the sample air, droplets are formed, counted and sized by an Optical Particle Counter (OPC). The sampling frequency is one second for both deployed CCN counters. Both CCN counters were calibrated using ammonium sulfate aerosol particles in the diameter range of 20-200 nm. The uncertainty of the effective water vapor supersaturation was $\pm 5\%$. (Rose et al., 2008)

218 2.1.4 Clouds

219 Aircraft-based measurements are an essential method for in situ sampling of cloud properties (Brenguier et al., 2013; Wendisch and Brenguier, 2013). Over the last 50-60 years, hot-wire probes 220 221 have been the most commonly used devices to estimate liquid water content (LWC) in the cloud 222 from research aircraft. Since the 1970s, the most widely used technique for cloud droplet spectra measurements has been developed based on the light-scattering effect. This type of instrument 223 224 provides the cloud droplet size distribution as the primary measurement. By integrating the cloud 225 droplet size distribution, additional information, such as LWC can be derived from the high-order 226 data product.

227 Three cloud probes from the G1 were discussed in this manuscript. The Cloud Droplet Probe 228 (CDP) is a compact, lightweight forward-scattering cloud particle spectrometer that measures cloud droplets in the 2 to 50 µm size range (Faber et al., 2018). Using a state-of-the-art electro-229 optics and electronics, Stratton Park Engineering (SPEC Inc.) developed a Fast Cloud Droplet 230 Probe (FCDP), which also use forward-scattering to determine cloud droplet distributions and 231 concentrations in the same range as CDP with up to 100 Hz sampling rate. The G1 also carried a 232 two-dimensional stereo probe (2DS, SPEC Inc.), which has two 128-photodiode linear arrays 233 working independently and electronics produce shadowgraph images with 10 µm pixel resolution. 234 Two orthogonal laser beams cross in the middle of the sample volume, with the sample cross 235 section for each optical path of 0.8 cm^2 . The manufacturer claims the maximum detection size is 236 237 up to 3000 µm for the 2DS. However, due to the counting statistic issue, the data used in this study is from 10–1000 µm only (Lawson et al., 2006). 2DS was upgraded with modified probe tips, and 238 239 an arrival time algorithm was applied to the 2DS data processing. Both efforts effectively reduced the number of small (shattered) particles (Lawson, 2011). For G1 cloud probes, the laboratory 240 calibrations of the sample area and droplet sizing were performed before the field deployment. 241





During the deployment, biweekly calibrations with glass beads were performed with the size variation of less than 5%, which were consistent with the pre-campaign and after-campaign calibrations. Comparison between the LWC derived from cloud droplet spectra with hot-wire LWC measurement was made to estimate/eliminate the coincidence errors in cloud droplet concentration measurements (Lance et al., 2010; Wendisch et al., 1996)

On board of HALO, two cloud probes were operated and discussed in this manuscript, each 247 consisting of a combination of two instruments: the Cloud Combination Probe (CCP) and a Cloud 248 249 Aerosol Precipitation Spectrometer (CAPS, denoted as NIXE-CAPS; NIXE: Novel Ice 250 Experiment). The CCP is a combination of a CDP (denoted as CCP-CDP) with a CIPgs (Cloud 251 Imaging Probe with grey scale, DMT, denoted as CCP-CIPgs). NIXE-CAPS consists of a CAS-Dpol (Cloud and Aerosol Spectrometer, DMT, denoted as NIXE-CAS) and a CIPgs (denoted as 252 253 NIXE-CIPgs). CIPgs is an optical array probe comparable to the 2DS operated on the G1. CIPgs 254 obtains images of cloud elements using a 64-element photodiode array (15µm resolution) to generate two-dimensional images with nominal detection diameter size range from 15 to 960 µm 255 (Klingebiel et al., 2015; Molleker et al., 2014). The CCP-CDP detects the forward-scattered laser 256 light by cloud particles in size range of 2.5 to 46 µm. The sample area of the CCP- CDP was 257 determined to be 0.27 ± 0.025 mm² with an uncertainty of less than 10% (Klingebiel et al., 2015). 258 CAS-Dpol (or NIXE-CAS) is a light scattering probe comparable to the CDP but covers the size 259 range of 0.6 to 50 μ m in diameter, thus including the upper size range of the aerosol particle size 260 spectrum (Luebke et al., 2016). Furthermore, CAS-Dpol measures the polarization state of the 261 particles (Costa et al., 2017). Correspondingly to the G1 CDP, the performance of the CCP-CDP 262 and NIXE-CAS were frequently proven by glass beads calibrations. Prior to or after each HALO 263 flight, CCP-CIPgs and NIXE-CIPgs calibrations were performed by using a mainly transparent 264 spinning disc that carries opaque spots of different but known size. The data of the CCP measured 265 particle concentration on board of HALO are corrected to gain ambient conditions using a 266 267 thermodynamic approach developed by (Weigel et al., 2016). For NIXE-CAPS, the size distributions were provided where NIXE-CAS was merged with the NIXE-CIPgs at 20 µm. 268

269 2.1.5 Solar radiation

The G1 radiation suite included shortwave (SW, 400 - 2,700 nm) broadband total upward and
downward irradiance measurements using Delta-T Devices model SPN-1 radiometers. The





272 radiation data were corrected for aircraft tilt from the horizontal reference plane. A methodology 273 has been developed (Long; et al., 2010) for using measurements of total and diffuse shortwave 274 irradiance and corresponding aircraft navigation data (latitude, longitude, pitch, roll, heading) to 275 calculate and apply a correction for platform tilt to the broadband hemispheric downward SW 276 measurements. Additionally, whatever angular offset there may be between the actual orientation 277 of each radiometer's detector and what the navigation data say is level has also been determined 278 for the most accurate tilt correction.

279 The Spectral Modular Airborne Radiation measurement sysTem (SMART-Albedometer) was 280 installed aboard HALO. Depending on the scientific objective and the configuration, the optical 281 inlets determining the measured radiative quantities can be chosen. The SMART-Albedometer has been utilized to measure the spectral upward and downward irradiances; thereby it is called as an 282 283 albedometer, as well as to measure the spectral upward radiance. The SMART-Albedometer is 284 designed initially to cover measurements in the solar spectral range between 300 and 2,200 nm (Krisna et al., 2018; Wendisch et al., 2001; Wendisch et al., 2016). However, due to decreasing 285 sensitivity of the spectrometers at large wavelengths, the use of the wavelengths was restricted to 286 300 - 1,800 nm. The spectral resolution is defined by the full width at half maximum (FWHM), 287 288 which is between 2 and 10 nm. In this case, the instruments were mounted on an active horizontal stabilization system for keeping the horizontal position of the optical inlets during aircraft 289 movements (up to +/-6 degrees from the horizontal plane). 290

291 2.2 Flight patterns

During the dry season IOP (September 1 – October 10, 2014), two types of coordinated flights were carried out: one flight in cloud-free condition (September 9) and two flights with clouds present (September 21 and October 1). In this study, we compare the measurements for both coordinated flight patterns. The discussion is mainly focused on the flights under cloud-free conditions on September 9 and the flight with clouds present on September 21, as shown in figure 1. The other coordinated flight on October 1 is included in the supplemental document.

For the cloud-free coordinated flight, the G1 took off first and orbited around an area from the planned rendezvous point until HALO arrived in sight. It then coordinated with HALO and performed a wing-to-wing maneuver along straight legs around 500 m above sea level, as shown in Figure 2. The normal G1 average sampling speed is 100 m s⁻¹, and the normal HALO average





sampling speed is 200 m s⁻¹. During the coordinated flight on September 9, both aircraft also adjusted their normal sampling speed by about 50 m s⁻¹ so that they could fly side by side.

For the second type of coordinated flights, the G1 and HALO flew the stacked paten at their 304 own normal airspeed. On September 21, the G1 also took off from the airport first, followed by 305 HALO 15 minutes later. Then, both aircraft flew above the T3 ground site and subsequently flew 306 several flight legs stacked at different altitudes. The two aircraft were vertically separated by about 307 330 m and sampled below, inside, and above clouds. Due to the different aircraft speeds, the flight 308 309 distance between two aircraft flight paths continued increasing from 15 min to 1 hour, as shown in Figure 3. On October 1, the G1 focused on the cloud microphysical properties and contrasting 310 polluted versus clean clouds. HALO devoted the flight to the cloud vertical evolution and life cycle 311 and also probed the cloud processing of aerosol particles and trace gases. The G1 and HALO 312 coordinated two flight legs between 950–1250 m above the T3 site under cloud-free conditions. 313 Following that, HALO flew to the south of Amazonia, and the G1 continued sampling plume-314 influenced clouds above the T3 site, and then flew above the Rio Negro area. 315

In this study, to perform a meaningful comparison of in situ measurements, all the data from 316 instruments were time synchronized with the aircraft (G1 or HALO) navigation system. For AMS 317 318 and CPC data, the time shifting due to tubing length and instrument flow had been corrected. For the coordinated flight on Sep. 9, the data compared was from the same type of measurements with 319 the same sampling rate. For the measurements with the different sampling rate, the data were 320 binned to the same time interval for comparison. For the flight with the cloud presents (Sep.21 and 321 322 Oct. 1), the following criteria are used: 1) the data collected by the two aircraft must be less than 30 mins apart from each other; 2) the comparison data was binned to 200 m altitude interval; and 323 3) the cloud flag was applied to the aerosol measurements, and the data affected by the cloud 324 shattering are eliminated from the comparisons of aerosol measurements. Moreover, additional 325 326 comparison criteria are specified for individual measurements in the following section. Tables 2 shows the total number of points used for the comparison. 327

328 3. Results

329 3.1 Comparison of the G1 and HALO measurements of atmospheric state parameters

The atmospheric state parameters comprise primary variables observed by the research aircraft.The measurements provide essential meteorological information not only for understanding the



(1)



atmospheric conditions but also for providing the sampling conditions for other measurements,such as those of aerosol particles, trace gases, and cloud microphysical properties.

For cloud-free coordinated flights, the comparison focused on the near side-by-side flight leg at around 500 m, as shown in Figure 2. Table 3 shows the basic statistics of the data for primary atmospheric state parameters. In general, the atmospheric state parameters observed from both aircraft were in excellent agreement. The linear regression achieved a slope was near 1 for four individual measurements. The regression is evaluated using the below equation 1.

$$R^2 = 1 - \frac{SS_{regression}}{SS_{Total}}$$

Where the sum squared regression error is calculated by $SS_{regression} = \sum (y_i - y_{regression})^2$, and the sum squared total error is calculated by $SS_{Total} = \sum (y_i - \bar{y})^2$, y_i is the individual data point, \bar{y} is the mean value, and $y_{regression}$ is the regression value. When the majority of the data points are in a narrow value range, using the mean is better than the regression line and the R^2 will be negative (Neg in Table 3).

345 The difference between the average ambient temperatures on the two aircraft was 0.5 K, and the difference between the average dew point temperatures was about 1 K. For temperature and 346 humidity, the G1 data were slightly higher than the HALO data. The main contributions to the 347 observed differences include the error propagation in the derivation of the ambient temperature 348 349 from the measured temperature, instrumental-measurement uncertainty, and the temporal and spatial variability. The average horizontal wind speed measured by HALO is 0.4 m s⁻¹ higher than 350 the average horizontal wind speed measured by the G1. The uncertainty source of wind estimation 351 is mainly due to the error propagation from the indicated aircraft speed measurement and the 352 353 aircraft ground speed estimation from GPS. The static pressure distribution measured aboard HALO showed a smaller standard deviation (0.9 hPa) compared to the value of the G1 (1.5 hPa). 354 355 The standard deviation (std) was also 0.6 hPa narrower. Part of the reason for this difference is a 356 more substantial variation of the G1 altitude during level flight legs when G1 flew at around 50 357 m/s higher than its normal airspeed. Thus, any biases caused by their near side-by-side airspeeds being different from their normal airspeeds would be undetected during these coordinated flights. 358 For the coordinated flights under cloudy conditions, we used the criteria from Section 2 to 359 360 compare ambient conditions measured by the G1 and HALO aircraft. As shown in Figure 4, the 361 linear regression slopes for ambient temperature, dew point temperature, and pressure were also





close to 1 between the G1 and HALO measurements during the September 21 coordinated flight. The R^2 value is also close to 1. These results suggest that the G1 and HALO measurements achieved excellent agreements. The dew point temperature from the G1 measurement was erroneous between 2200–2700 m and above 3700 m (Figure 4(c)) because the G1 sensor was skewed by wetting in the cloud. The HALO dew point temperature was calculated from the total water mixing ratio measured by TDL, and that measurement in the cloud was more accurate than the measurement made by the chilled mirror hydrometer aboard the G1.

The lower value of the R^2 value in horizontal wind speed means the ratio of the regression error 369 370 and total error in wind measurement is much higher than the temperature and pressure 371 measurements. The main contributions to this difference are the error propagation during the horizontal wind speed estimation and also the temporal and spatial variance between two aircraft 372 sampling location. We observed differences between the two aircraft data of up to 2 m s⁻¹, caused 373 374 by the increasing sampling distance as the two aircraft were climbing up. For example, the G1 flew a level leg above T3 around 2500 m between 16:20-16:30, while HALO stayed around 2500 375 m for a short period and kept climbing to a higher altitude. Due to strong vertical motion, 376 turbulence, and different saturations (evaporation-condensation processes), the variances in the 377 378 horizontal wind speed (Figure 4(d)) were also more significant compared to the variances of 379 temperature and pressure measurements.

380 3.2 Comparison of trace gas measurements

For the cloud-free coordinated flight, ozone is the only trace gas measurement available on both aircraft. The linear regression slope shows that the HALO ozone concentration was about 8% higher than the G1 concentration. The difference between the averaged ozone concentrations was 4.1 ppb. The primary source of bias is probably the different ozone loss in the sampling and transfer lines.

386

The comparison made on September 21 flight in Figure 5 shows good agreement for the vertically averaged ozone measurements. Comparing the statistics data from September 9, the ozone measurement is not sensitive to the temporal and spatial changes. The G1 CO measurement shows a low R^2 value which is mainly from the systematic bias between the two instruments with different operation principles. The larger variance between 2000-3000 m in CO concentration





392 indicates the spatial variation contribution, while the G1 and HALO were sampling different air

393 masses, as indicated in Figure S7.

394 3.3 Comparison of aerosol measurements

Aerosol particles exhibited strong spatial variations, both vertically and horizontally, due to 395 complex atmospheric processing in the Amazon basin, especially with the local anthropogenic 396 397 sources at Manaus. Thus, any spatially resolved measurement is critical to characterizing the properties of the Amazonian aerosols. The cloud-free coordinated flights provide us with suitable 398 data to compare the G1 and HALO aerosol measurements and thus will enable further studies 399 400 combined with the ground measurements. The vertical profiles obtained using the G1 and HALO 401 platforms in different aerosol regimes in the Amazon basin have contributed to many studies (Fan et al., 2018; Martin et al., 2017; Wang et al., 2016). 402

When comparing the measurements from the two aircraft, the inlet system is a critical item, 403 especially for sampling aerosol particles (Wendisch et al., 2004). Inlet design and characterization 404 can actively modify the measured aerosol particle number concentration, size distribution, and 405 406 chemical composition. The G1 aerosol inlet is a fully automated isokinetic inlet. Based on the 407 manufacturer wind tunnel test and peer-reviewed publications, this inlet operates for aerosol up to 5 µm, with transmission efficiency around 50 % at 1.5 µm (Dolgos and Martins, 2014; Kleinman 408 et al., 2007; Zaveri et al., 2010). The HALO sub-micrometer Aerosol Inlet (HASI) was explicitly 409 designed for HALO. Based on the numerical flow modeling, optical particle counter 410 measurements, and field study evaluation, HASI has a cut-off size of 3 µm, with transmission 411 efficiency larger than 90 % at 1 µm (Andreae et al., 2018; Minikin et al., 2017). 412

413 3.3.1 Aerosol particle number concentration

For the cloud-free coordinated flight, the linear regression of CPC and UHSAS between the G1 and HALO measurements are also included in Table 3. Also, the total number concentration of HALO CPC data was about 20 % lower than the total number concentration from the G1 CPC, as shown in Figure 6. The typical uncertainty between two CPCs is 5-10%.(Gunthe et al., 2009; Liu and Pui, 1974) Many factors can contribute to the rest 10-15% difference. Those include systematic uncertainties, such as systematic instrument drifts, different aerosol particle losses inside of the two CPCs, and different inlet transmission efficiencies in the two aircraft.





421 The CPC data in Figure 6 are color-coded with UTC time. The general trend is that the aerosol 422 number concentration increased with aircraft sampling through the Manaus plume between 15:30-15:40. A similar trend was observed in aerosol particle number concentration (Figure 7) measured 423 424 by the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)-Airborne version (referred to as UHSAS). The total number concentration data given by UHSAS (Figure 7) is integrated over the 425 overlapping size range (90 - 500 nm for the September 9 flight) for both the G1 and HALO 426 UHSAS. The linear regression shows that the total aerosol particle concentration from HALO 427 428 UHSAS is about 16.5% higher than the total aerosol concentration from the G1 UHSAS. The 429 discrepancy between the two UHSAS measurements is mainly due to the error propagation in the 430 sampling flow, the differential pressure transducer reading, the instrument stability, and calibration repeatability, which is consistent with the other UHSAS study (Kupc et al., 2018). In the airborne 431 432 version of UHSAS, mechanical vibrations have a more significant impact on the pressure transducer reading than the case for the bench version of UHSAS. 433

For the coordinated flight on September 21, the G1 and HALO data are averaged to a 200 m 434 vertical altitude interval, as shown in Figure 8. There was a good agreement in the CPC 435 comparison, especially at altitudes of 200 m and above (<10 % variance). However, the linear 436 437 regression slope significantly decreased, which was primarily related to the temporal and spatial differences in aerosol number concentration. Especially between 2000 - 3000 m altitude, the 438 difference between the G1 and HALO measurement is largely due to the different aerosol srouces, 439 as shown in Figure S6(a). The size range was integrated from 100-700 nm for UHSAS on 440 441 September 21. The change of the size range was because the overlap of size distribution from both UHSAS instruments was changed. The linear regression slope and the R^2 value slightly decreased 442 in the UHSAS comparison as shown in Figure 8(b). And we can expect that the main contribution 443 to the UHSAS measurement difference is from instrument systematic drift and the spatial/temporal 444 445 variance of particle concentration in the ambient environment as shown in Figure S6(b). However, combining with the CPC measurement comparison, it indicates that the smaller size aerosol 446 447 particles (< 100 nm) have a more profound variance due to the temporal and spatial change.

448 3.3.2 Aerosol particle size distribution

For the cloud-free coordinated flight, the averaged aerosol size distribution measured during one flight leg is compared in Figure 9. Based on the comparison plot, at the size range less than 90





451 nm, the G1 UHSAS overestimated the particle counts, which is due to the error in the counting efficiency correction. The UHSAS detection efficiency is close to 100% for particles larger than 452 100 nm and concentrations below 3000 cm⁻³ but decreases both for smaller particles and for higher 453 concentrations considerably (Cai et al., 2008). The aerosol counting efficiency correction 454 determined from the lab condition does not represent the real correction during the flight operation. 455 Between 90 nm and 250 nm, FIMS agreed well with the G1 UHSAS, whereas HALO UHSAS is 456 about 30 % higher than the other two instruments. For the size range of 250–500 nm, FIMS had 457 458 good agreement with HALO UHSAS, whereas FIMS is about 30-50 % higher than the G1 UHSAS 459 depending on the particle size. Because the UHSAS has a simplified "passive" inlet, the large size 460 aerosol particle loss in the UHSAS inlet was expected to increase with the increase of the aircraft speed. Thus, the lower G1 UHSAS counts at a larger aerosol particle size are likely related to the 461 particle loss correction. 462

For the September 21 flight, the aerosol size distribution vertical profiles were averaged into 463 464 100 m altitude intervals (Figure 10). Overall, all size distribution measurements captured the mode near 100 nm between 800–1000 m, which is at the top of the convective boundary layer, as 465 indicated by the potential temperature (Figure 10(d)), which starts from a maximum near the 466 467 ground and then becomes remarkably uniform across the convective boundary layer. With the increase in altitude, we observe the peak of the aerosol size distribution shifted from 100 nm to 468 150 nm. Note that due to data availability, the aerosol size distribution data from the HALO 469 UHSAS has a less spatial resolution. 470

471 3.3.3 A significant contribution of small aerosol particles

Comprehensive characterization of aerosol particles, especially small ones (<50 nm) 472 during GoAmazon2014/5 has demonstrated that high concentrations of those small particles in the 473 lower free troposphere are transported from the free troposphere into the boundary layer by a 474 475 strong convective downdraft and sustain the population of particles in the pristine Amazon boundary layer. This important observation improved the current understanding of the aerosol 476 influence on cloud properties and climate under natural conditions (Fan et al., 2018; Wang et al., 477 2016). However, the aerosol particle size distribution measurement, especially for sizes less than 478 479 50 nm, is very rare due to the lack of high-frequency airborne measurements. The most common 480 aerosol size spectrometer, UHSAS, covers aerosol particle sizes larger than 60 nm. The scanning





mobility particle spectrometer cannot obtain size distribution in 1 Hz time resolution. The other approach is to estimate particle size distribution by extrapolating the UHSAS or Passive Cavity Aerosol Spectrometer Probe (size range 100 – 3000 nm) measured aerosol size distribution to smaller size ranges (down to 10 nm). The accuracy of the third approach is limited by the nature of the aerosol size distribution, and the aerosol particle concentration can be significantly underestimated if there is a dominant nucleation mode in the aerosol particle size distribution, such as during a new particle formation event.

As shown in Figure 11, we compared the integrated aerosol number concentrations 488 between one wet season flight (on March 7), which was influenced by a long-range transport plume 489 490 from Africa (Moran-Zuloaga et al., 2018) and one typical dry season flight (September 21). The agreement of the small aerosol number concentration between the FIMS-measured size 491 distribution and UHSAS/PCASP estimated size distribution is reasonably good for the dry season 492 flight when the accumulation mode dominated the aerosol particle size distribution. During the 493 494 wet season, there was a strong vertical gradient in the particle size spectrum above central 495 Amazonia under clean conditions. Thus, we can observe an increase of underestimation of the small size particle concentration both for the size ranges less than 50 nm and less than 100 nm, as 496 497 the filled markers move away from each other with the increase of altitude. Because of the negligible mass contribution to the total aerosol loading, those ultrafine aerosol particles (< 50 nm) 498 are conventionally considered too small to affect cloud formation. However, the new observational 499 500 evidence and numerical simulation of deep convective clouds outlined a new mechanism, which suggests an energetic anthropogenic invigoration of deep convective clouds by those ultrafine 501 502 aerosol particles in previously pristine regions of the world (Fan et al., 2018). Two newly published 503 studies (Fan et al., 2018; Wang et al., 2016) emphasize the importance of the airborne observation and suggest the ultrafine aerosol particles (<50 nm) measurement should be included as a baseline 504 505 routine measurement in future airborne experiments.

For field studies without the deployment of FIMS, one option to assess the accuracy of UHSAS/PCASP estimated size distribution is to compare the total number concentration based on the integration of the UHSAS/PCASP estimated size distribution to the total number concentration from CPC. For field study focusing on the high concentration and variability of sub-50 or sub-100 nm aerosol particles, such as new particle formation events, it is highly recommended to request





511 the deployment of FIMS. Due to the limited availability of FIMS, one option is to use several 512 well-characterized CPCs, which operate at the different cut-off sizes, to measure the ambient aerosol simultaneously, and then use the data-inversion technique to estimate the aerosol size 513 514 distribution of sub-50 or sub-100 nm aerosol particles. Another reasonable substitute to the FIMS might be a Scanning Mobile Particle Sizer (SMPS), but it should be noted that on an airborne 515 platform an SMPS does not nearly have the same time resolution as a FIMS. To better adapt the 516 spatial change in aerosol concentration, a residence chamber similar to a system described in 517 518 another study (Kotchenruther and Hobbs, 1998) should be deployed with SMPS.

519 3.3.4 Aerosol particle chemical composition

520 Figure 12(a) shows vertical profiles of the total mass concentrations measured by the two AMS instruments on September 21. Above 2500 m altitude, the agreement between the two 521 instruments is excellent (mean difference less than 5%). Between 2000 and 2500 m, the agreement 522 523 is within the uncertainty range. Below 2000 m altitude, however, the aerosol particle mass concentrations measured by the AMS operated on HALO are lower than the concentrations 524 525 measured by the AMS on the G1. The aerosol volume concentrations from G1 AMS was converted 526 from the mass concentration from AMS, by assuming the organic compound density was 1.5 g 527 cm-3 (Pöschl et al., 2010). The converted aerosol volume concentration agreed well with the volume concentration calculated based on UHSAS data below 2500 m, as shown in Figure 12(b). 528 The agreement at lower altitudes suggests that the lower concentration in HALO AMS is due to 529 the transmission efficiency issue in the constant pressure inlet used by HALO AMS. This inlet was 530 a prototype, designed and built at MPIC Mainz, and works by changing the size of the critical 531 orifice that regulates the flow into the aerodynamic lens. The design and transmission 532 characteristics will be described in an upcoming publication (Molleker, S., in prep.). The AMS 533 aboard the G-1 used a constant pressure inlet based on the design in Bahreini et al., 2008. Thus, 534 535 we conclude that data above 2500 m altitude measured by the AMS aboard HALO in 2014 are valid, while data below 2500 m need to be corrected using correction factors derived from 536 laboratory characterization before further study. After 2014, the HALO inlet design was improved 537 to address the inlet transmission issues specific to this field campaign. 538

539 3.3.5 CCN number concentration





540 These measurements provide valuable information about the aerosol's ability to form cloud 541 droplets and modify the microphysical properties of clouds. Numerous laboratory and field studies have improved the current understanding of the connections between aerosol particle size, 542 chemical composition, mixing states and CCN activation properties (Bhattu and Tripathi, 2015; 543 Broekhuizen et al., 2006b; Chang et al., 2010; Duplissy et al., 2008; Lambe et al., 2011; Mei et al., 544 2013a; Mei et al., 2013b; Pöhlker et al., 2016; Thalman et al., 2017). In addition, based on the 545 simplified chemical composition and internal mixing state assumption, various CCN closure 546 547 studies have achieved success within $\pm 20\%$ uncertainty for ambient aerosols (Broekhuizen et al., 548 2006a; Mei et al., 2013b; Rissler et al., 2004; Wang et al., 2008).

According to earlier studies (Gunthe et al., 2009; Pöhlker et al., 2016; Roberts et al., 2001; Roberts et al., 2002; Thalman et al., 2017), the hygroscopicity (κ_{CCN}) of CCN in the Amazon basin is usually dominated by organic components (κ_{Org}). Long-term ground-based measurements at the Amazon Tall Tower Observatory also suggested that there were low temporal variability and no pronounced diurnal cycles in hygroscopicity only under natural rainforest background conditions (Pöhlker et al., 2018; Pöhlker et al., 2016).

555 Using FIMS and CCN data from both the G1 and HALO collected during the coordinated flight leg on September 9, the critical dry diameter (D_{50}) was determined by integrating FIMS size 556 distribution to match the CCN total number concentration. Then, D_{50} was combined with the CCN-557 558 operated supersaturation to derive the effective particle hygroscopicity by applying the k-Köhler 559 theory. The histogram plots based on the density of the estimated hygroscopicity (κ_{est}) from both aircraft were compared for the flight leg above T3. For the G1 and HALO data, the κ_{est} value 560 derived from the flight leg above the T3 site is 0.186±0.067 and 0.189±0.083 separately. That 561 value is also slightly higher than the overall mean kappa derived from long-term measurements 562 563 from the Amazon Tall Tower Observatory, which is 0.17±0.06 (Pöhlker et al., 2016; Thalman et al., 2017). 564

An example of a comparison of vertical profiles of the CCN concentration at 0.5% supersaturation on September 21 is shown in Figure 13. The difference between CCN measurements on the two aircraft is about 20% on average. However, the linear regression slope increases to 0.9120 if we focused on the data above 2500 m. The main contributions to the





569 difference include the difference in aerosol inlet structure, aerosol particle loss correction in the 570 main aircraft inlet and the constant pressure inlet, the systematic inlet difference below 2500 m as

- shown in AMS data, and the error propagation of CCN measurements.
- 572 3.4 Comparison of cloud measurements

573 In situ cloud measurements help to capture the diversity of different cloud forms and their 574 natural temporal and spatial variability. The G1 CDP and FCDP were deployed under the different wing pylons, and also on the different side of the aircraft. The G1 2DS was deployed on the same 575 side of FCDP. The HALO cloud combination probe (CCP-CDP and CCP-CIPgs) and NIXE-CAPS 576 577 (NIXE-CAS and NIXE-CIPgs) were deployed under the different wing pylons but on the same side of the aircraft. On September 21, 2014, based on the aircraft location and elevation 578 information as shown in Figure 1(b) and Figure 3, two aircraft were sampling above T3 site and 579 passing through the same cloud field at ~1600 m flight leg and ~1900 m flight leg as shown in 580 Figure S8 and Figure S9. We used the cloud probes data from ~1900 m flight leg for the cloud 581 droplet number concentration comparison. Two size ranges were considered: 3-20 µm from light 582 583 scattering probes (CDP vs. FCDP on the G1, CCP-CDP vs. NIXE-CAS on HALO) and 2-960 µm 584 from combined cloud probes.

585 3.4.1 Comparison of cloud droplet number concentration between 3-20 μm

The primary cloud layer was observed by both the G1 and HALO between 1000-2500 m above 586 ground. Although the two aircraft have sampled along the same flight path, the instruments 587 probably observed different sets of the cloud due to cloud movement with the prevailing wind or 588 different cloud evolution stages. Thus, an initial comparison focuses on the redundant instruments 589 on the same aircraft, that measured truly collocated and synchronous on board of HALO and of 590 591 the G1, respectively. In Figure 14 (a), the data of the CCP-CDP and of the NIXE-CAS are juxtaposed sampled over about 13 minutes for particles detection size ranges which were 592 considered as most equivalent. The comparison reveals two ranges of particle number 593 concentrations at which densification of agreeing measurements become visible. At very low 594 number concentrations (about $10^{-1} - 10$ per cm³) the presence of inactivated (interstitial) aerosols 595 596 in the clear air space between the very few cloud elements should be considered. Over specific 597 ranges, however, the fine structure of varying cloud droplet number concentration may cause the





598 regression's scattering, indicated by cloud particle measured by one instrument whilst respective 599 antagonist seems to measure within almost clear air - and vice versa. At higher number concentrations, i.e., between 10^2 and 10^3 per cm³, the comparison of the highly resolved data 600 601 constitutes increasing compactness with respect to the 1:1 line. The overall data scatter of this 602 comparison, however, may indicate the highly variable structure within clouds as those investigated over the Amazon basin. The data of the G1 CDP and the FCDP are juxtaposed as the 603 604 same as HALO cloud probes. However, the sampled cloud period was much shorter – about 3 605 minutes. Similar as the HALO cloud probes comparison, we observe two ranges of particle number concentrations at which a densification of agreeing measurements become visible, especially for 606 607 the lower number concentrations, in Figure 14(b). At higher number concentrations, only a few cloud elements were observed by the G1 cloud probes. That is because the G1 was about 7-23 608 609 minutes later to pass the same location as HALO, and experienced much fewer cloud elements.

610 3.4.2 Comparison of cloud droplet size distribution between 2-960 μ m from both aircraft

611 Comparing the cloud probes from the G1 and HALO, although it seems that the size distributions never match better than for the cloud particle diameter size range below 10 µm, the 612 size distributions from two aircraft are in remarkably good agreement, considering the instance 613 that the cloud detection on board the G1 occurred 7-23 minutes after the cloud probing on board 614 of HALO, as shown in Figure 15. On HALO, the CCP and NIXE-CAPS probes agreed very well 615 between 2-960 µm and both peaked around 10 µm. On the G1, although CDP and FCDP has a 616 more significant difference in the size range less than 8 µm, both of them showed the peak of the 617 size distribution was around 15 µm. The difference between the G1 CDP and FCDP may be due 618 to the data post-processing. Additional coincidence correction and shattering correction were 619 applied to FCDP, but not to CDP. For cloud elements larger than 10 µm, the difference between 620 621 the obtained cloud particle size distributions from two aircraft becomes substantial (up to two orders of magnitude) which may be indicative for observations of two different stages within the 622 progressing development of a precipitation cloud which is particularly expressed in elevated 623 number concentrations of larger cloud elements observed during the G1 measurement that 624 625 happened later. We also observed that the general cloud characteristic is similar at different altitude levels as shown in Figure S10. The first two of three averaged periods were chosen during the 626 flight leg of ~1600 m and the last average period is for the flight leg ~1900 m compared in Figure 627





628 15. Due to the averaging, the fine in-cloud structure gets suppressed. The small scale variabilities 629 inside a cloud which are illustrated by the scattering of the highly resolved measurement data from 630 the instrument comparison (cf. Figure 14) and the temporal evolution of in-cloud microphysics are 631 not ascertainable and furthermore are beyond the scope of this study.

632 3.5 Comparison of radiation measurements

633 In this study, the downward irradiance measured by the SPN-1 unshaded center detector was compared with the integrated downward irradiance from the SMART-Albedometer between 634 300–1,800 nm wavelengths in Figure 16. Only measurements from flight legs, where the G1 and 635 636 HALO flew near side-by-side and at the same altitude were taken into consideration for analysis. 637 In Figure 16, the top panel shows the time series of SPN-1 measurements, and the bottom panel shows the time series of SMART-Albedometer measurements. The black dots represented all data, 638 and the blue circles identified data when the navigation condition was within +/- 1 degree from the 639 horizontal level. The large scatter in the data between 15:12-15:28 and 15:35-15:40 is mainly due 640 to the different sensor trajectories during the maneuvering of the aircraft to get to the coordinated 641 642 flight position. Because of the difference of each aircraft position from horizontal, the measured signal varied from the signal of the direct component of sunlight. Each sensor might look at 643 644 different directions of the sky or different parts of the clouds. In addition, both aircraft flew under scattered clouds, and this uneven sunlight blocking is another contribution to the "drop-off" 645 646 behavior in the time series plots of the downward irradiance.

647 Comparing the G1 and HALO measurements between 15:15-1:55 using the restricted 648 navigation criteria in Figure 17, we observed that the G1 SPN1 irradiance is slightly higher than 649 the integrated irradiance from the SMART-Albedometer. However, the difference in the averaged 650 irradiance is less than 10 %. That result could be due to the difference in radiometer spectral ranges: 651 400–2700 nm (SPN1 radiometer) vs. 300–1800 nm (SMART).

652 4 Uncertainty assessment

As mentioned in the introduction, a low-flying G1 and a high-flying HALO cover the sampling area from the atmospheric boundary layer, low clouds to the free troposphere, and the sampling period from the dry and wet seasons (Martin et al., 2016a). This spatial coverage provides the user community with abundant atmospheric-related data sets for their further studies, such as for remote





657 sensing validation and modeling evaluation. However, one critical step to bridge the proper usage 658 of the observation with further atmospheric science study is to understand the measurement 659 uncertainty in this data set, especially the variation between the coexisting measurements due to 660 the temporal and spatial difference.

661 For the major measurements during this field study, three primary sources contribute to the measurement variation between the two aircraft: the temporal and spatial variations, the difference 662 in the inlet characterization, and the limitation of the instrument capability. The difference in the 663 inlet characterization and the instrument error are the same between the coordinated flights on 664 September 9 and September 21. Thus, we can examine the sensitivity of each measurement to the 665 spatial variation by comparing two flights. For the majority of the comparisons of the September 666 21 flight, there are no significant spatial and temporal variation between two aircraft 667 measurements. However, we noticed the comparison uncertainty is more significant between 2000 668 -3000 m altitude than the other altitudes in the aerosol and trace gas profile, especially for the 669 670 aerosol particles smaller than 100 nm. This additional difference occurring between 2000-3000 m 671 indicates the spatial variation contribution, while the G1 and HALO were sampling different air masses. The G1 had one flight leg around 2500 m above T3 site, while HALO continued climbing 672 673 through 2000-3000 m range to reach the next flight leg around 4500 m. Thus, for the G1 674 measurements, the data show two modes in the histogram distribution. The large mode was typically from the data when the G1 passed through the pollution plume, and the small mode 675 676 represented the background value. Because the flight path of HALO did not pass through the plume, their data shows only one mode in the histogram plots, as shown in S6 and S7 in the 677 678 supplemental material.

For atmospheric meteorological variables, the overall uncertainty is relatively smaller (less than 1 %) comparing to the other airborne measurements. The main contribution for the three-dimensional wind measurement is more sensitive to the spatial variation than the ambient temperature and pressure due to the complex turbulence structure in the boundary layer (see Figure S5). The other measurement affected mainly by the spatial and temporal variation is the cloud measurements, which is consistent with a previous study (Andreae et al., 2004). The considerable variation in the comparison between 2-960 μm indicates the evolution of cloud droplet size





distributions (DSDs) over time and space has a more significant influence on the large droplet size,and it serves as the major contributor for the DSDs comparison.

The inlet also significantly affects the aerosol and gas phase measurements. Inlet 688 689 characterizations are inherently challenging. However, comparisons as shown here can be used to assess the performance of the inlets indirectly. In this study, reasonable agreement of the total 690 number concentration of aerosol particles between two CPCs indicated the uncertainty caused by 691 the main aerosol inlet difference is less than 15%. In addition to the main aerosol inlet, the particle 692 losses caused by AMS aerosol constant pressure inlet also affects the AMS comparison below 693 2500 m altitude. Based on a literature survey, this study, for the first time, compares the non-694 695 refractory particle mass concentration between two aircraft measurements. Although two AMS sampled different air masses during the majority of the campaign, the excellent agreement between 696 the two measurements from the comparison flight linked the aerosol chemical composition from 697 the wet to dry season and from the atmospheric boundary layer to the upper troposphere. 698

We also noticed that the CCN and UHSAS comparisons are associated with more substantial 699 uncertainties because of the more complex instrument designs. The aerosol flow fluctuation, the 700 CCN column temperature fluctuation, and the stabilization of the optical particle counter all 701 contribute to the accurate estimation of the CCN concentration. In a similar sense, the aerosol flow 702 703 fluctuation, the difference in the inlet efficiency at different platform speeds, the laser temperature fluctuation, and the signal-to-noise ratio at lower size range all contribute to the considerable 704 705 uncertainty of the UHSAS concentration measurement. However, the CCN hygroscopicity estimation on the near side-by-side comparison on September 9, 2014, shows very encouraging 706 707 agreement. Thus, the spatial variance and the instability of the CCN and UHSAS performance 708 both led to the variance between two aircraft of up to 50% based on the comparison scenario on 709 September 21, 2014. This remains the most significant variance we observed during these two 710 aircraft measurement comparisons.

The summary of the major measurement uncertainty contributed by the spatial differencebetween the two aircraft is listed in Table 4.





714 **5 Summary**

715 In situ measurements made by well-characterized instruments installed on two research aircraft (the G1 and HALO) during the GoAmazon 2014/5 and ACRIDICON-CHUVA campaigns were 716 compared. Overall, the analysis shows good agreement between the G1 and HALO measurements 717 718 for a relatively broad range of atmospheric-related variables in a challenging strophic environment. 719 Measured variables included atmospheric state parameters, aerosol particles, trace gases, clouds, and radiation properties. This study outlines the well-designed coordinated flights for achieving a 720 meaningful comparison between two moving platforms. The high data quality was ensured by the 721 722 most sophisticated instruments aboard two aircraft used the most advanced techniques, assisted with the best-calibrated/characterized procedures. The comparisons and the related uncertainty 723 estimations quantify the current measurement limits, which provide the guidance to the modeler 724 725 to realistically quantify the modeling input value and evaluate the variation between the 726 measurement and the model output. The comparison also identified the measurement issues, outlined the associated reasonable measurement ranges, and evaluated the measurement 727 sensitivities to the temporal and spatial variance. 728

729 The comparisons presented here were mainly from two coordinated flights. The flight on September 9 was classified as a cloud-free flight. During this flight, the G1 and HALO flew near 730 731 side-by-side within a "polluted" leg, which was above the T3 site and across the downwind pollution plume from Manaus, and a "background" leg, which was outbound from Manaus to the 732 733 west and could be influenced by the regional biomass burning events during the dry season. Both legs were at 500 m altitude and showed linear regression slopes of ambient temperature and 734 735 pressure, horizontal wind speed and dew point temperature near to 1 between the G1 and HALO measurements. These comparisons provide a solid foundation for further evaluation of aerosol, 736 trace gas, cloud, and radiation properties. The total aerosol concentration from CPC and UHSAS 737 were compared for the 500 m flight leg above the T3 site. The UHSAS measurement had a better 738 agreement than the CPC measurement. That is because of the minor difference in the inlet structure 739 and instrument design between two UHSAS aboard the two aircraft. The average size distribution 740 741 from both UHSAS and one FIMS in the G1 suggests that UHSAS had an over-counting issue at the size range between 60-90 nm, which was probably due to electrical noise and small signal-to-742 noise ratio in that size range. Good agreement in the aerosol size distribution measurement 743 provides a "sanity" check for AMS measurements. A CCN closure study suggested that FIMS 744





745 provides valuable size coverage for better CCN number concentration estimation. Based on the κ -Köhler parameterization, κ_{eff} observed at 500 m above the T3 site is 0.18±0.09, which is similar to 746 the overall mean kappa from long-term ATTO measurements - 0.17±0.06 (Pöhlker et al., 2016). 747 748 This similarity suggests that there is no significant spatial variability along the downwind transect, 749 although the freshly emitted aerosol particles may have much less hygroscopicity. The difference in the ozone measurement comparison is about 4.1 ppb, which suggests that the bias due to the 750 sampling line loss inside of the G1 gas inlet. The irradiance from the SPN1 unshaded center 751 752 detector in the G1 was compared with the HALO integrated downward irradiance between 300-753 1800 nm and achieved a very encouraging agreement with a variance of less than 10%.

During the second type of the coordinated flights on September 21 (with cloudy conditions), 754 755 HALO followed the G1 after take-off from Manus airport; then the two aircraft flew stacked legs relative to each other at different altitudes above the T3 site. For atmospheric state parameters, 756 757 nearly linear correlations between the G1 and HALO were observed for ambient pressure, 758 temperature, and dew point temperature measurements at an altitude range from ground to around 759 5000 m. Cloud presence affected the measurements of dew point temperature in the G1, resulting 760 in a large discrepancy in the dew point temperature measurement and the derived relative humidity 761 between 2000–3000 m. The horizontal wind had more variation than the rest of the meteorological properties, which is mainly due to the temporal and spatial variability. The aerosol number 762 concentration comparison had an excellent agreement (<15 %) for aerosol particles larger than 10 763 nm counted by the CPC below 2500 m. While the integrated aerosol number concentration from 764 765 UHSAS showed consistent discrepancy at different altitudes, that suggests the significant temporal and spatial variation of smaller aerosol particles (<100 nm). Although the aircraft-based UHSAS 766 is a challenging instrument to operate, a reasonable size distribution profile comparison was made 767 between both UHSAS and FIMS in the G1. The aerosol concentration measured by AMS 768 instruments showed a mean difference less than 5% above 2500 m during the flight on September 769 770 21, although due to the ongoing study, the correction factors allowing for correction of data below 2500 m are not available yet. The difference between CCN number concentration measured on the 771 772 two aircraft was on average 20%, and these data show the same altitude behavior as the AMS data. 773 The main contributions to this difference include the difference in aerosol inlet structure, the aerosol loss correction in a constant pressure inlet, and the error propagation of CCN 774 measurements. The ozone and CO vertical profile comparisons show a variation of less than 10%. 775





The ozone measurement variation is mainly from the systematic bias between two instrumentswith different operation principles, especially at altitudes higher than 4000 m.

778 Cloud probe comparisons were made for the cloud droplet number concentration between 3– 779 20 µm for the initial comparison between the redundant instruments on the same aircraft. Then the comparison of cloud droplet size distribution between 2-960 µm for a flight leg around 1900 m 780 showed a remarkably good agreement. The major cloud appearance was captured by both aircraft, 781 although the cloud elements observed were affected by the cloud movement with the prevailing 782 wind and the different cloud evolution stages. Furthermore, the relatively short time delay of 7-23 783 784 minutes between the independent measurements may give a hint for the time scales in which the 785 cloud droplet spectra develop within a convective cloud over the Amazon basin.

The above results provide additional information about the reasonableness of measurements for each atmospheric variable. This study confirms the high-quality spatial and temporal dataset with clearly identified uncertainty ranges had been collected from two aircraft and builds a good foundation for further studies on the remote sensing validation and the spatial and temporal evaluation of modeling representation of the atmospheric processing and evolution.

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- Table 1. List of compared measurements and corresponding instruments deployed aboard the G1 and HALO during GoAmazon2014/5. The acronyms are defined in a table at the end of this
- 1100 paper. Dp indicates the particle diameter. \Box Dp refers to the size resolution.

Measurement	Instruments deployed on the G1	Instruments deployed on HALO
Variables	(Martin et al., 2016b; Schmid et al., 2014)	(Wendisch et al., 2016)
Static Pressure	Rosemount (1201F1), 0-1400 hPa	Instrumented nose boom tray (DLR development), 0-1400 hPa
Static air temperature	Rosemount E102AL/510BF	Total Air Temperature (TAT) inlet (Goodrich/Rosemount type 102) with an
	-50 to +50 °C	open wire resistance temperature sensor (PT100), -70 to +50 °C
Dewpoint temperature	Chilled mirror hygrometer 1011B -40 to +50 °C	Derived from the water-vapor mixing ratio, which is measured by a tunable diode laser (TDL) system (DLR development), 5-40000 ppmv
3-D wind	Aircraft Integrated Meteorological Measurement System 20 (AIMMS-20)	Instrumented nose boom tray (DLR development) with an air data probe (Goodrich/Rosemount) 858AJ and high- precision Inertial Reference System (IGI IMU-IIe)
Particle number concentration	CPC, cut off size $(D_p) = 10 \text{ nm}$	CPC, cut off size $(D_p) = 10 \text{ nm}$
Size	UHSAS-A, 60-1000 nm.	UHSAS-A, 60-1000 nm.
distribution*	FIMS, 20 nm – 500 nm	
Non-Refractory	HR-ToF-AMS: Organics, Sulfate, Nitrate,	C-ToF-AMS: Organics, Sulfate, Nitrate,
particle chemical composition	Ammonium, Chloride, 60-1000 nm	Ammonium, Chloride, 60-1000 nm
CCN concentration	CCN-200, SS= 0.25, 0.5%	CCN-200, SS= 0.13-0.53%
Gas phase	N2O/CO and Ozone Analyzer, CO, O ₃	N2O/CO and Ozone Analyzer, CO, O ₃
concentration	concentration, precision 2 ppb	concentration, precision 2 ppb
Cloud	CDP, 2-50 μm, ΔD _p =1-2 μm	CCP-CDP, 2.5-46 mm, $\Delta D_p=1-2 \ \mu m$
properties*	FCDP, 2-50 μ m, ΔD_{p} =1-2 μ m	NIXE-CAS: 0.61 -52.5 μm
	2DS, 10-1000 μm	NIXE-CIPgs, 15-960 µm
		ССР-СІРдs: 15-960 µm
Radiation	SPN1 downward irradiance, 400-2700 nm	SMART Albedometer, downward spectral irradiance, 300-2200 nm

1101 *for an individual flight, the size range may vary.





1103

1104 Table 2. Summary of the total data points compared between the G1 and HALO instruments.

	SEP 9, 2014		SEP 21, 2014	
	G1	HALO	G1	HALO
Atmospheric parameters	2815	2815	7326	12065
Gas phase, CO	N/A	N/A	7326	12065
Gas phase, Ozone	2815	2815	7110	11766
CPC	2043	2043	8466	11646
UHSAS (FIMS)	2031	2031	5841 (9405)	828
AMS	N/A	N/A	587	818
CCNc	663	531	7982	4546
G1: CDP(FCDP)	N/A	N/A	3627(4439)	2051(2260)
HALO: CCP-CDP				
(NIXE-CAS)				
G1: 2DS	N/A	N/A	2280	2261 (2260
HALO: CCP-CIPgs				
(NIXE-CIPgs)				
RAD	1355	1355	N/A	N/A





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Table 3. Summary of basic statistics of data between in situ measurements

		Con	parison o	of the coo	ordinate	ed flight o	on Sep. 9			
Variables	G1			HALO						
	min	max	mean	std	min	max	mean	std	slope	\mathbb{R}^2
Т, К	297.7	300.2	298.9	0.5	297.2	299.4	298.4	0.4	1.002	Neg.
P, hPa	955	965	960.1	1.5	958	964.9	961.8	0.9	0.998	Neg.
WSpd, m/s	0.3	8.9	3.4	1.2	0.3	7.7	3.8	1.1	0.998	Neg.
T _{dew} , k	293	296.5	295.0	0.5	292.9	294.9	294.0	0.3	0.996	Neg.
Ozone, ppb	0.5	58.8	22.2	9.3	18.3	50.8	26.3	6.6	1.082	0.9401
CPC, cm ⁻³	696.0	3480.6	1591.3	568.7	687.4	2639.4	1313.8	473.5	0.819	0.8508
UHSAS, cm ⁻³	78.2	1118.	645.5	116.3	504.1	1622.2	756.3	138.6	1.165	0.8193
<i>CCNc (к</i>)	0.010	0.347	0.1855	0.067	0.012	0.394	0.1890	0.083	0.8937	Neg.

1109





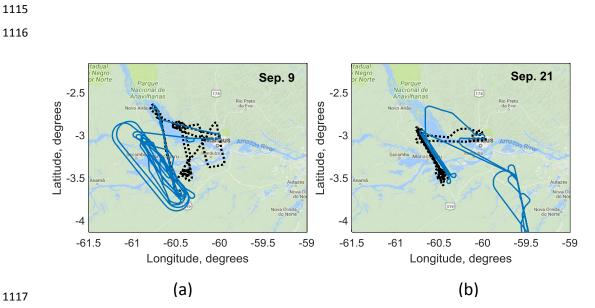
- 1111 Table 4. List of compared measurements ranges and measurement variances caused by the spatial
- 1112 variation during the field campaign.

Measurement	Measured Range during the Field	Measurement Variances between the
Variables	Campaign	Two Aircraft
Static Pressure	500 – 1010 hPa	< 1 %
Static air	272 – 310 K	< 1%
temperature		
Dewpoint	230 -300 K	Without clouds, <1%
temperature		With clouds, the measurement from the G1 can be up to 5% lower than that of HALO
3-D wind	1-15 m/s	< 40%
Particle number	500 – 15,000 cm ⁻³	< 20% for CPC, <50% for UHSAS (size
concentration		dependent)
Non-Refractory	$< 10 \ \mu g \cdot m^{-3}$	< 10% above 2500 m
particle chemical		
composition		Up to 50% below 2500 m
CCN	SS=0.25%, 100 - 2000 cm ⁻³	< 10% above 2500 m
concentration		
		Up to 50% below 2500 m
Gas phase	Ozone: 15-75 ppb	Ozone: < 25%
concentration	CO: 50-200 ppb	CO: < 15%
Cloud droplet	3- 20 μm	<50 %
number		
concentration		
Downward	200 -1500 W·m ⁻²	< 10%
irradiance		

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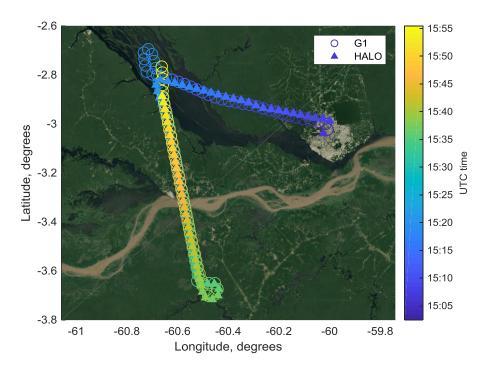


1118 Figure 1. Coordinated flight tracks for September 9 (a) and September 21 (b). The black dotted

1119 line is the flight track of the G1, and the blue line is the flight track of HALO.





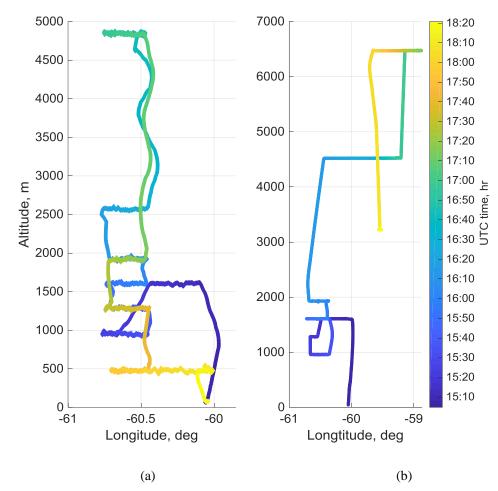


1122 Figure 2. Time-colored flight track of the G1 (circle) and HALO (triangle) on September 9 during

a cloud-free coordinated flight at 500 m above sea level (50 m apart as the closest distance).







1126 Figure 3. Time-colored flight profile of the G1 (a) and HALO (b) on September 21, during a

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¹¹²⁷ coordinated flight.





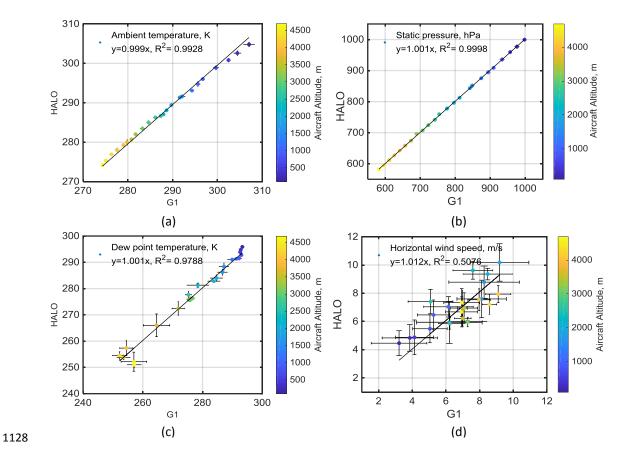
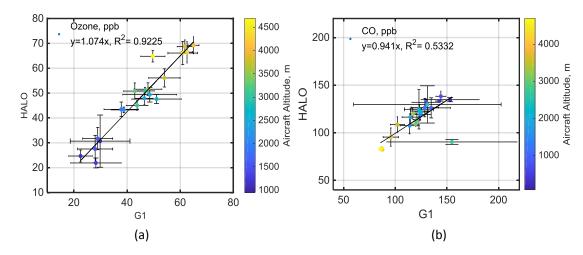


Figure 4. Aircraft altitude-colored plots of (a) ambient temperature, (b) static pressure, (c) dew
point temperature, and (d) horizontal wind speed observed by the G1 and HALO on September
21.



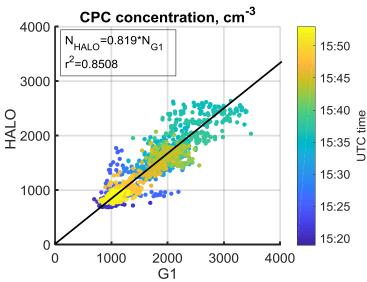




1134 Figure 5. Aircraft altitude-colored plots of trace gas (a) Ozone, (b) CO, for the coordinated flight

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on September 21.



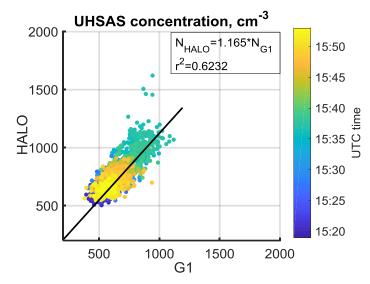


1137 Figure 6. The G1 and HALO comparison for aerosol number concentration measured by CPC

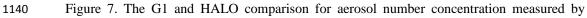
1138 (>10 nm) on September 9.



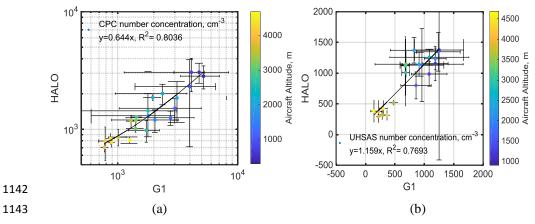








1141 UHSAS (90-500 nm) on September 9.

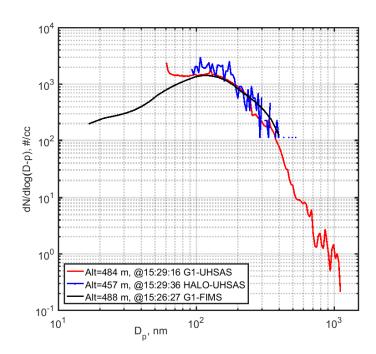


1144 Figure 8. The G1 and HALO comparison for aerosol number concentration profiling measured

1145 by (a) CPC and (b) UHSAS (100-700 nm) on September 21.







1146

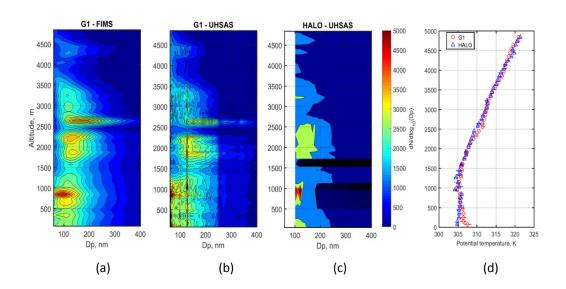
1147 Figure 9. The G1 and HALO comparison for aerosol size distribution measured by UHSAS

1148 (from both aircraft) and FIMS (on the G1) on September 9.











1152 Figure 10. Aerosol size distribution vertical profiles measured by (a) the G1 FIMS, (b) The G1

1153 UHSAS, (c) the HALO UHSAS, (d) Potential temperature aboard the G1 and HALO on September1154 21.





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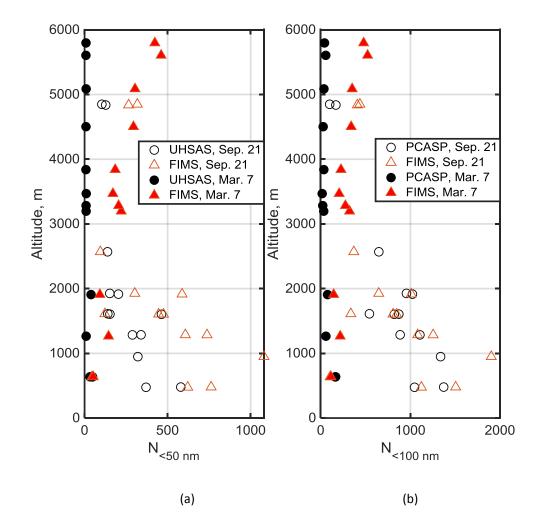
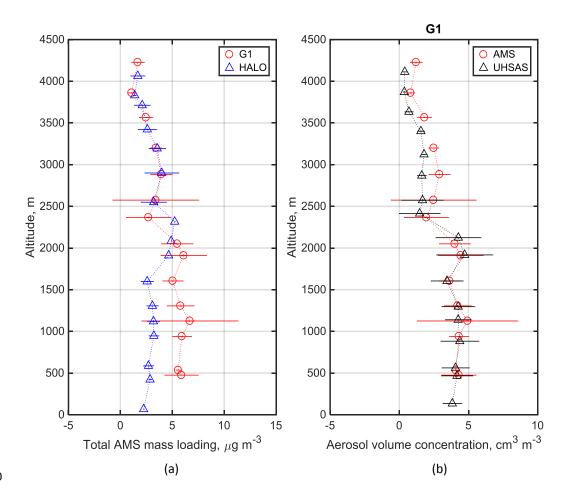


Figure 11. Comparison of the number concentration for (a) sizes less than 50 nm and (b) sizes
less than 100 nm. FIMS-measured size distribution and UHSAS/PCASP estimated size
distribution are used.







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Figure 12. (a) Comparison of aerosol mass loading measured by the G1 and HALO AMS on
September 21; (b) aerosol volume concentration comparison from AMS and the integrated
UHSAS on the G1.





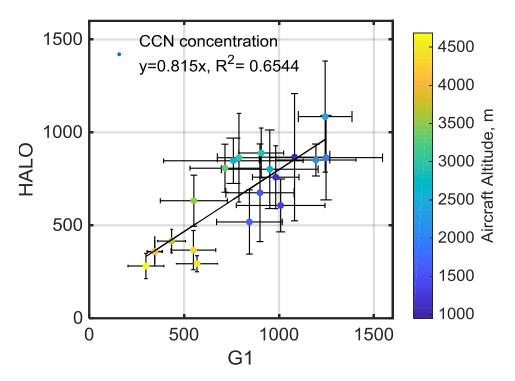
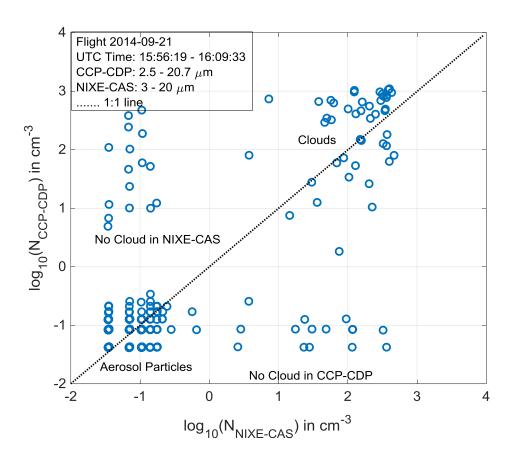


Figure 13. The G1 and HALO comparison of aerosol CCN concentration (S=0.5%) measuredon September 21.







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(a)





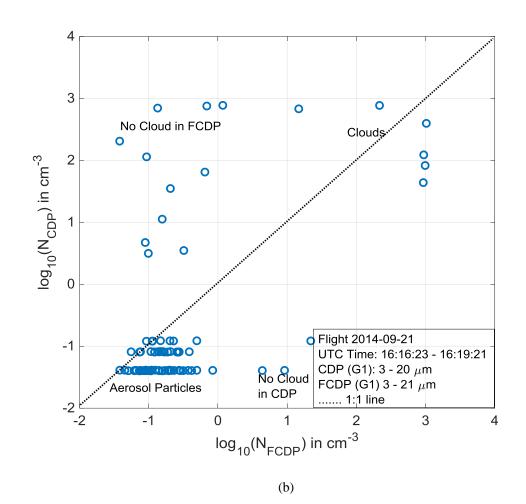
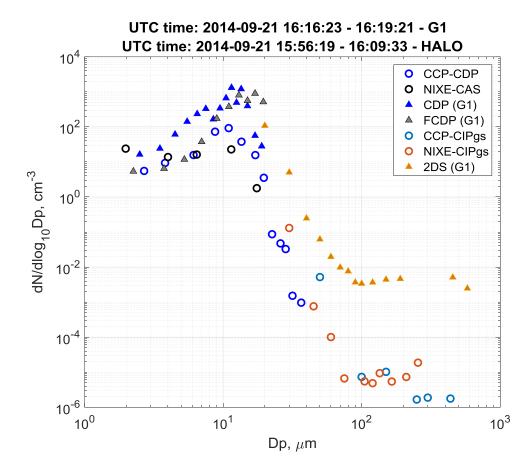




Figure 14 The comparison of cloud droplet concentrations in the same aircraft (a) betweenNIXE-CAS and CCP-CDP on board HALO; (b) between CDP and FCDP on board the G1.







1175 Figure 15. The cloud droplet size distribution from the cloud probes on the G1 and HALO.

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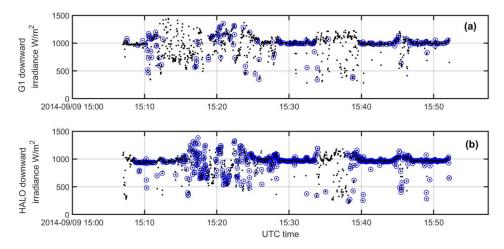
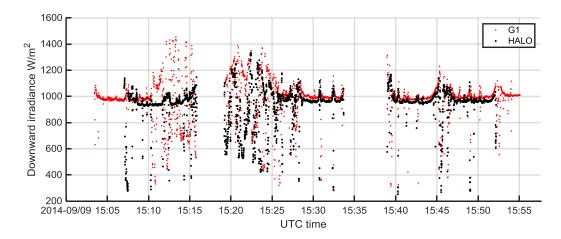




Figure 16. Time series of the G1 and HALO downward irradiance on September 9. The (a)
by SPN-1 and (b) by SMART-Albedometer. Black dots represent all data under the general intercomparison criteria. The blue circles represent the restricted navigation criteria.



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Figure 17. Time series comparison of the G1 (SPN-1) and HALO (SMART-Albedometer)radiation measurements on September 9.

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