1	Comparison of Aircraft Measurements during GoAmazon2014/5 and
2	ACRIDICON-CHUVA
3 4 5 6 7 8	Fan Mei <sup>1</sup> , Jian Wang <sup>2, 16</sup> , Jennifer M. Comstock <sup>1</sup> , Ralf Weigel <sup>3</sup> , Martina Krämer <sup>4, 3</sup> , Christoph Mahnke <sup>3, 5</sup> , John E. Shilling <sup>1</sup> , Johannes Schneider <sup>5</sup> , Christiane Schulz <sup>5</sup> , Charles N. Long <sup>6</sup> , Manfred Wendisch <sup>7</sup> , Luiz A. T. Machado <sup>8</sup> , Beat Schmid <sup>1</sup> , Trismono Krisna <sup>7</sup> , Mikhail Pekour <sup>1</sup> , John Hubbe <sup>1</sup> , Andreas Giez <sup>9</sup> , Bernadett Weinzierl <sup>10</sup> , Martin Zoeger <sup>9</sup> , Mira L. Pöhlker <sup>5</sup> , Hans Schlager <sup>9</sup> , Micael A. Cecchini <sup>11</sup> , Meinrat O. Andreae <sup>5,12</sup> , Scot T. Martin <sup>13</sup> , Suzane, S. de Sá <sup>13</sup> , Jiwen Fan <sup>1</sup> , Jason Tomlinson <sup>1</sup> , Stephen Springston <sup>2</sup> , Ulrich Pöschl <sup>5</sup> , Paulo Artaxo <sup>14</sup> , Christopher Pöhlker <sup>5</sup> , Thomas Klimach <sup>5</sup> , Andreas Minikin <sup>15</sup> , Armin Afchine <sup>4</sup> , Stephan Borrmann <sup>3, 5</sup>
9	1. Pacific Northwest National Laboratory, Richland, WA, United States.
10	2. Brookhaven National Laboratory, Upton, NY, United States.
11	3. Institute for Physics of the Atmosphere, Johannes Gutenberg University, Mainz, Germany
12	4. Research Centre Jülich, Institute for Energy and Climate Research 7: Stratosphere (IEK-7), Jülich, Germany
13	5. Max Planck Institute for Chemistry, Mainz, Germany
14	6. NOAA ESRL GMD/CIRES, Boulder, CO, United States
15	7. University of Leipzig, Leipzig, Germany
16	8. National Institute for Space Research (INPE), São Paulo, Brazil
17	9. Deutsches Zentrum für Luft- und Raumfahrt (DLR), Oberpfaffenhofen, Germany
18	10. University of Vienna, Vienna, Austria
19	11. University of São Paulo (USP), São Paulo, Brazil
20	12. Scripps Institution of Oceanography, University of California San Diego, La Jolla, California, USA
21	13. Harvard University, Cambridge, MA, United States
22	14. Instituto de Física, Universidade de São Paulo, São Paulo, Brazil
23	15. DLR Oberpfaffenhofen, Flight Experiments Facility, Wessling, Germany
24 25	16. Center for Aerosol Science and Engineering, Department of Energy, Environmental and Chemical Engineering, Washington University in St. Louis, St. Louis, Missouri, USA
26	Correspondence to: Fan Mei (fan.mei@pnnl.gov)
27	Abstract. The indirect effect of atmospheric aerosol particles on the Earth's radiation balance
28	remains one of the most uncertain components affecting climate change throughout the industrial
29	period. The large uncertainty is partly due to the incomplete understanding of aerosol-cloud
30	interactions. One objective of the GoAmazon2014/5 and ACRIDICON-CHUVA projects was to

understand the influence of emissions from the tropical megacity of Manaus (Brazil) on the 31 surrounding atmospheric environment of the rainforest and to investigate its role in the life cycle 32 33 of convective clouds. During one of the intensive observation periods (IOPs) in the dry season from September 1 to October 10, 2014, comprehensive measurements of trace gases and aerosol 34 properties were carried out at several ground sites. In a coordinated way, the advanced suites of 35 sophisticated in situ instruments were deployed aboard both the U.S. Department of Energy 36 Gulfstream-1 (G1) aircraft and the German High Altitude and Long-Range Research Aircraft 37 38 (HALO) during three coordinated flights on September 9, 21, and October 1. Here we report on the comparison of measurements collected by the two aircraft during these three flights. Such 39 comparisons are challenging but essential for assessing the data quality from the individual 40 platforms and quantifying their uncertainty sources. Similar instruments mounted on the G1 and 41 42 HALO collected vertical profile measurements of aerosol particle number concentrations and size distribution, cloud condensation nuclei concentrations, ozone and carbon monoxide mixing ratios, 43 cloud droplet size distributions, and downward solar irradiance. We find that the above 44 measurements from the two aircraft agreed within the measurement uncertainties. The relative 45 46 fraction of the aerosol chemical composition measured by instruments on HALO agreed with the corresponding G1 data, although the total mass loadings only have a good agreement at high 47 48 altitudes. Furthermore, possible causes of the discrepancies between measurements on the G1 and HALO are examined in this paper. Based on these results, criteria for meaningful aircraft 49 50 measurement comparisons are discussed.

51

### 52 1. Introduction

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

conducted in 2014 and 2015 (Martin et al., 2017; Martin et al., 2016b). The primary objective of
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).

68 A feature of the GoAmazon 2014/5 field campaign was the design of the ground sites' location, which uses principles of Lagrangian sampling to align the sites with the Manaus pollution 69 plume (Figure 1: Source location – Manaus (T1 site), and downwind location – Manacapuru (T3 70 site)). The ground sites were overflown with the low-altitude U.S. Department of Energy (DOE) 71 72 Gulfstream-1 (G1) aircraft and the German High Altitude and Long Range Research Aircraft (HALO). These two aircraft are among the most advanced in atmospheric research, deploying 73 74 suites of sophisticated and well-calibrated instruments (Schmid et al., 2014; Wendisch et al., 2016). The pollution plume from Manaus was intensively sampled during the G1 and HALO 75 76 flights and also by the DOE Atmospheric Radiation Measurement (ARM) program Mobile 77 Aerosol Observing System and ARM Mobile Facility located at one of the downwind surface sites 78 (T3 site-70 km west of Manaus). The routine ground measurements with coordinated and intensive observations from both aircraft provided an extensive data set of multi-dimensional observations 79 80 in the region, which serves i) to improve the scientific understanding of the influence of the emissions of the tropical megacity of Manaus (Brazil) on the surrounding atmospheric 81 environment of the rainforest and ii) to understand the life cycle of deep convective clouds and 82 study open questions related to their influence on the atmospheric energy budget and hydrological 83 84 cycle.

As more and more data sets are merged to link the ground-based measurements with aircraft observations, and as more studies focus on the spatial variation and temporal evolution of the atmospheric properties, it is critical to quantify the uncertainty ranges when combining the data collected from the different platforms. Due to the challenges of airborne operations, especially when two aircraft are involved in data collection in the same area, direct comparison studies are rare. However, this type of research is critical for further combining the datasets between the ground sites and aircraft. Thus, the main objectives of the study herein are to demonstrate how to 92 achieve meaningful comparisons between two moving platforms, to conduct detailed comparisons 93 between data collected by two aircraft, to identify the potential measurement issues, to quantify 94 reasonable uncertainty ranges of the extensive collection of measurements, and to evaluate the 95 measurement sensitivities to the temporal and spatial variance. The comparisons and the related 96 uncertainty estimations quantify the current measurement limits, which provide realistic 97 measurement ranges to climate models as initial conditions to evaluate their output.

The combined GoAmazon2014/5 and ACRIDICON-CHUVA field campaigns not only 98 provide critical measurements of aerosol and cloud properties in an under-sampled geographic 99 region but also offer a unique opportunity to understand and quantify the quality of these 100 measurements using carefully orchestrated comparison flights. The comparisons between the 101 measurements from similar instruments on the two research aircraft can be used to identify 102 103 potential measurement issues and quantify the uncertainty range of the field measurements, which include primary meteorological variables (Section 3.1), trace gases concentrations (Section 3.2), 104 aerosol particle properties (number concentration, size distribution, chemical composition, and 105 microphysical properties) (Section 3.3), cloud properties (Section 3.4), and downward solar 106 107 irradiance (Section 3.5). We evaluate the consistency between the measurements aboard the two 108 aircraft for a nearly full set of gas, aerosol particle, and cloud variables. Results from this 109 comparison study provide the foundation not only for assessing and interpreting the observations from multiple platforms (from the ground to low altitude, and then to high altitude) but also for 110 111 providing high-quality data to improve the understanding of the accuracy of the measurements related to the effects of human activities in Manaus on local air quality, terrestrial ecosystems in 112 rainforest, and tropical weather. 113

#### 114 **2. Measurements**

### 115 2.1 Instruments

The ARM Aerial Facility deployed several in situ instruments on the G1 to measure atmospheric state parameters, trace gas 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. Details on maintenance and calibration of the involved instrumentation canbe found in the supplement (Table S1 and Table S2).

#### 124 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 a 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  $\pm 0.1$  K, and the uncertainty of the field data is  $\pm 0.5$  K. The static pressure had a measurement uncertainty of 0.5 hPa. The standard measurement uncertainties were  $\pm 2$  K for the chilled mirror hygrometer and 0.5 ms<sup>-1</sup> for wind speed.

On HALO, primary meteorological data were obtained from the Basic HALO Measurement 132 133 and Sensor System (BAHAMAS) at a one-second time resolution. The system acquired data from airflow and thermodynamic sensors and from the aircraft avionics and a high-precision inertial 134 reference system to derive the basic meteorological parameters like pressure, temperature, the 3D 135 wind vector, aircraft position, and attitude. The water vapor mixing ratio and further derived 136 humidity quantities were measured by the Sophisticated Hygrometer for Atmospheric Research 137 (SHARC) based on direct absorption measurement by a tunable diode laser (TDL) system. The 138 absolute accuracy of the primary meteorological data was 0.5 K for air temperature, 0.3 hPa for 139 air pressure, 0.4-0.6 ms<sup>-1</sup> for wind, and 5% ( $\pm 1$  ppm) for water vapor mixing ratio. All sensors 140 were routinely calibrated and traceable to national standards (Giez et al., 2017; Krautstrunk and 141 Giez, 2012). 142

## 143 2.1.2 Gas phase

Constrained by data availability, the comparison of trace gas measurements is focused on carbon monoxide (CO) and ozone (O<sub>3</sub>) concentrations. Those measurements were made aboard the G1 by a CO/N<sub>2</sub>O/H<sub>2</sub>O instrument (Los Gatos Integrated Cavity Output Spectroscopy instrument model 907-0015-0001), and an Ozone Analyzer (Thermo Scientific, Model 49i), respectively. The G1 CO analyzer was calibrated for response daily by NIST-traceable commercial standards before the flight. Due to the difference between laboratory and field conditions, the uncertainty of the CO measurements is about  $\pm$ 5% for one-second sampling periods. An ultra-fast

carbon monoxide monitor (Aero Laser GmbH, AL5002) was deployed on HALO. The detection 151 of CO is based on a vacuum-ultraviolet-fluorimetry, employing the excitation of CO at 150 nm, 152 and the precision is 2 ppb, and the accuracy is about 5%. The ozone analyzer measures ozone 153 concentration based on the absorbance of ultraviolet light at a wavelength of 254 nm. The ozone 154 analyzer (Thermo Scientific, Model 49c) in the HALO payload is very similar to the one on the 155 G1 (Model 49i), with an accuracy greater than 2 ppb or about  $\pm 5\%$  for four-second sampling 156 periods. The G1 ozone monitor was calibrated at the New York State Department of 157 158 Environmental Conservation testing laboratory at Albany.

#### 159 2.1.3 Aerosol

Aerosol number concentration was measured by different condensation particle counters 160 (CPCs) on the G1 (TSI, CPC 3010) and HALO (Grimm, CPC model 5.410). Although two CPCs 161 162 were from different manufacturers, they were designed using the same principle, which is to detect particles by condensing butanol vapor on the particles to grow them to a large enough size that 163 they can be counted optically. Both CPCs were routinely calibrated in the lab and reported the data 164 at a one-second time resolution. The HALO CPC operated at 0.6-1 L min<sup>-1</sup>, with a nominal cutoff 165 of 4 nm. Due to inlet losses, the effective cutoff diameter increases to 9.2 nm at 1000 hPa, and 166 11.2 nm at 500 hPa (Andreae et al., 2018; Petzold et al., 2011). The G1 CPC operated at 1 L min<sup>-</sup> 167 <sup>1</sup> volumetric flow rate and the nominal cut-off diameter  $D_{50}$  measured in the lab was ~10 nm. 168 169 During a flight, the cut-off diameter may vary due to tubing losses, which contributes less than 10 % uncertainty to the comparison between two CPC concentrations. 170

Two instruments deployed on the G1 measured aerosol particle size distribution. a Fast 171 172 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 173 ambient aerosol particles were charged after entering the FIMS inlet and then separated into 174 175 different trajectories in an electric field based on their electrical mobility. The spatially separated 176 particles grow into super-micrometer droplets in a condenser where supersaturation of the working fluid is generated by cooling. At the exit of the condenser, a high-speed charge-coupled device 177 178 camera captures the image of an illuminated grown droplet at high resolution. In this study, we used the FIMS 1 Hz data for comparison. The size distribution data from FIMS were smoothed. 179 Aside from the FIMS, the airborne version of the Ultra High Sensitivity Aerosol Spectrometer 180

(UHSAS) was deployed on G1 and HALO. The G1 and HALO UHSAS were manufactured by
the same company, and both were mounted under the wing on a pylon. UHSAS is an opticalscattering, laser-based particle spectrometer system. The size resolution is around 5% of the
particle size. The G1 UHSAS typically covered a size range of 60 nm to 1000 nm. HALO UHSAS
covered 90 nm to 500 nm size range for the September 9 flight.

Based on operating principles, FIMS measures aerosol electrical mobility size, and UHSAS 186 measures the aerosol optical equivalent size. Thus, the difference in the averaged size distributions 187 from those two types of instruments might be linked to differences in their underlying operating 188 principles, such as the assumption in the optical properties of aerosol particles. The data processing 189 in the G1 UHSAS assumed that the particle refractive index is similar to ammonium sulfate (1.55), 190 which is larger than the average refractive index (1.41-0.013i) from a previous Amazon study 191 192 (Guyon et al., 2003). The HALO UHSAS was calibrated with polystyrene latex spheres, which have a refractive index of about 1.572 for the UHSAS wavelength of 1054 nm. The uncertainty 193 due to the refraction index can lead to up to 10% variation in UHSAS measured size (Kupc et al., 194 2018). Also, the assumption of spherical particles affects the accuracy of UHSAS sizing of ambient 195 196 aerosols.

197 The chemical composition of submicron non-refractory (NR-PM<sub>1</sub>) organic and inorganic 198 (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; 199 200 Shilling et al., 2018; Shilling et al., 2013). Based on the standard deviation of observed aerosol mass loadings during filter measurements, the HR-ToF-AMS detection limits for the average time 201 of thirteen seconds are approximately 0.13, 0.01, 0.02, 0.01 (3 $\sigma$  values) µg m<sup>-3</sup> for organic, sulfate, 202 nitrate, and ammonium, respectively (DeCarlo et al., 2006). A Compact Time-of-flight Aerosol 203 Mass Spectrometer (C-ToF-AMS) was operated aboard HALO to investigate the aerosol 204 composition. Aerosol particles enter both the C-ToF-AMS and HR-ToF-AMS via constant 205 pressure inlets controlling the volumetric flow into the instrument, although the designs of the 206 207 inlets are somewhat different (Bahreini et al., 2008). The details about the C-ToF-AMS operation and data analysis are reported in Schulz's paper (Schulz et al., 2018). The overall accuracy has 208 209 been reported as ~30 % for both AMS instruments (Alfarra et al., 2004; Middlebrook et al., 2012). 210 Data presented in this section were converted to the same condition as the HALO AMS data, which is 995 hPa and 300 K. Both AMS instruments were calibrated before and after the field deploymentand also once a week during the field campaign.

213 The number concentration of cloud condensation nuclei (CCN) was measured aboard both 214 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 215 216 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 217 and heat. After the supersaturated water vapor condenses on the CCN in the sample air, droplets 218 are formed, counted, and sized by an Optical Particle Counter (OPC). The sampling frequency is 219 220 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 221 222 vapor supersaturation was  $\pm 5\%$ . (Rose et al., 2008)

### 223 2.1.4 Clouds

Aircraft-based measurements are an essential method for in situ samplings of cloud properties 224 (Brenguier et al., 2013; Wendisch and Brenguier, 2013). Over the last 50–60 years, hot-wire probes 225 have been the most commonly used devices to estimate liquid water content (LWC) in the cloud 226 from research aircraft. Since the 1970s, the most widely used technique for cloud droplet spectra 227 measurements has been developed based on the light-scattering effect. This type of instrument 228 229 provides the cloud droplet size distribution as the primary measurement. By integrating the cloud droplet size distribution, additional information, such as LWC can be derived from the high-order 230 data product. 231

Three cloud probes from the G1 are discussed in this manuscript. The Cloud Droplet Probe 232 233 (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 state-of-the-art electro-optics 234 and electronics, Stratton Park Engineering (SPEC Inc.) developed a Fast Cloud Droplet Probe 235 (FCDP), which also uses forward-scattering to determine cloud droplet distributions and 236 concentrations in the same range as CDP with up to 100 Hz sampling rate. The G1 also carried a 237 238 two-dimensional stereo probe (2DS, SPEC Inc.), which has two 128-photodiode linear arrays working independently. The 2DS electronics produce shadowgraph images with 10 µm pixel 239

resolution. Two orthogonal laser beams cross in the middle of the sample volume, with the sample 240 cross section for each optical path of  $0.8 \text{ cm}^2$ . The manufacturer claims the maximum detection 241 size is up to 3000 µm for the 2DS. However, due to the counting statistic issue, the data used in 242 this study is from 10–1000 µm only (Lawson et al., 2006). 2DS was upgraded with modified probe 243 tips, and an arrival time algorithm was applied to the 2DS data processing. Both efforts effectively 244 245 reduced the number of small shattered particles (Lawson, 2011). For G1 cloud probes, the 246 laboratory calibrations of the sample area and droplet sizing were performed before the field deployment. During the deployment, weekly calibrations with glass beads were performed with 247 248 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 249 250 LWC measurement was made to estimate/eliminate the coincidence errors in cloud droplet concentration measurements (Lance et al., 2010; Wendisch et al., 1996) 251

252 On board of HALO, two cloud probes were operated and discussed in this manuscript, each 253 consisting of a combination of two instruments: Cloud Combination Probe (CCP) and a Cloud Aerosol Precipitation Spectrometer (CAPS, denoted as NIXE-CAPS; NIXE: Novel Ice 254 Experiment). The CCP is a combination of a CDP (denoted as CCP-CDP) with a CIPgs (Cloud 255 256 Imaging Probe with greyscale, DMT, denoted as CCP-CIPgs). NIXE-CAPS consists of a CAS-257 Dpol (Cloud and Aerosol Spectrometer, DMT, denoted as NIXE-CAS) and a CIPgs (denoted as NIXE-CIPgs). CIPgs is an optical array probe comparable to the 2DS operated on the G1. CIPgs 258 obtains images of cloud elements using a 64-element photodiode array (15µm resolution) to 259 generate two-dimensional images with a nominal detection diameter size range from 15 to 960 µm 260 (Klingebiel et al., 2015; Molleker et al., 2014). The CCP-CDP detects the forward-scattered laser 261 light by cloud particles in size range of 2.5 to 46 µm. The sample area of the CCP- CDP was 262 determined to be  $0.27\pm0.025$  mm<sup>2</sup> with an uncertainty of less than 10% (Klingebiel et al., 2015). 263 CAS-Dpol (or NIXE-CAS) is a light scattering probe comparable to the CDP but covers the size 264 range of 0.6 to 50 µm in diameter, thus including the upper size range of the aerosol particle size 265 spectrum (Luebke et al., 2016). Furthermore, CAS-Dpol measures the polarization state of the 266 267 particles (Costa et al., 2017). Correspondingly to the G1 CDP, the performance of the CCP-CDP and NIXE-CAS were frequently examined by glass beads calibrations. Prior to or after each HALO 268 flight, CCP-CIPgs and NIXE-CIPgs calibrations were performed by using a mainly transparent 269 spinning disc that carries opaque spots of different but known size. The data of the CCP measured 270

particle concentration on board of HALO are corrected to gain ambient conditions using a
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.

## 274 2.1.5 Solar radiation

The G1 radiation suite included shortwave (SW, 400 - 2,700 nm) broadband total upward and 275 276 downward irradiance measurements using Delta-T Devices model SPN-1 radiometers. The radiation data were corrected for aircraft tilt from the horizontal reference plane. A methodology 277 278 has been developed (Long; et al., 2010) for using measurements of total and diffuse shortwave 279 irradiance and corresponding aircraft navigation data (latitude, longitude, pitch, roll, heading) to 280 calculate and apply a correction for platform tilt to the broadband hemispheric downward SW measurements. Additionally, whatever angular offset there may be between the actual orientation 281 282 of each radiometer's detector and what the navigation data say is level has also been determined 283 for the most accurate tilt correction.

The Spectral Modular Airborne Radiation measurement sysTem (SMART-Albedometer) was 284 installed aboard HALO. Depending on the scientific objective and the configuration, the optical 285 inlets determining the measured radiative quantities can be chosen. The SMART-Albedometer has 286 287 been utilized to measure the spectral upward and downward irradiances; thereby, it is called as an albedometer, as well as to measure the spectral upward radiance. The SMART-Albedometer is 288 designed initially to cover measurements in the solar spectral range between 300 and 2,200 nm 289 (Krisna et al., 2018; Wendisch et al., 2001; Wendisch et al., 2016). However, due to the decreasing 290 291 sensitivity of the spectrometer at large wavelengths, the use of the wavelengths was restricted to 300 – 1,800 nm. The spectral resolution is defined by the full width at half maximum (FWHM), 292 which is between 2 and 10 nm. In this case, the instruments were mounted on an active horizontal 293 stabilization system for keeping the horizontal position of the optical inlets during aircraft 294 movements (up to +/-6 degrees from the horizontal plane). 295

## 296 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 (section
S1, Figure S1, S2, S7, and S8).

For the cloud-free coordinated flight on September 9, 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<sup>-1</sup>, and the normal HALO average sampling speed is 200 m s<sup>-1</sup>. During the coordinated flight on September 9, both aircraft also adjusted their normal sampling speed by about 50 m s<sup>-1</sup> so that they could fly side by side.

For the second type of coordinated flights, the G1 and HALO flew the stacked pattern at their 310 311 own typical airspeed. On September 21, the G1 also took off from the airport first, followed by HALO 15 minutes later. Then, both aircraft flew above the T3 ground site and subsequently flew 312 313 several flight legs stacked at different altitudes. The two aircraft were vertically separated by about 330 m and sampled below, inside, and above clouds. Due to the different aircraft speeds, the time 314 315 difference between two aircraft visiting the same part of the flight paths varied, increasing up to 1 hour at the end of the flight path, as shown in Figure 3. On October 1, the G1 focused on the cloud 316 317 microphysical properties and contrasting polluted versus clean clouds. HALO devoted the flight to the cloud vertical evolution and life cycle and also probed the cloud processing of aerosol 318 319 particles and trace gases. The G1 and HALO coordinated two flight legs between 950-1250 m above the T3 site under cloud-free conditions. Following that, HALO flew to the south of 320 321 Amazonia, and the G1 continued sampling plume-influenced clouds above the T3 site, and then flew above the Rio Negro area. 322

323 In this study, to perform a meaningful comparison of in situ measurements, all the data from 324 instruments were time-synchronized with the aircraft (G1 or HALO) navigation system. For AMS and CPC data, the time shifting due to tubing length and instrument flow had been corrected. For 325 the coordinated flight on Sep. 9, the data compared was from the same type of measurements with 326 327 the same sampling rate. For the measurements with the different sampling rate, the data were 328 binned to the same time interval for comparison. For the flight with the cloud present (Sep.21 and Oct. 1), the following criteria are used: 1) the data collected by the two aircraft must be less than 329 30 mins apart from each other; 2) the comparison data were binned to 200 m altitude intervals; 3) 330

the cloud flag was applied to the aerosol measurements, and the data affected by the cloud shattering are eliminated from the comparisons of aerosol measurements. Moreover, additional comparison criteria are specified for individual measurements in the following section. Table 2 shows the total number of points used for the comparison.

335 3. Results

## 336 3.1 Comparison of the G1 and HALO measurements of atmospheric state parameters

The atmospheric state parameters comprise the primary variables observed by the research 337 aircraft. The measurements provide essential meteorological information not only for 338 understanding the atmospheric conditions but also for providing the sampling conditions for other 339 340 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 341 at around 500 m, as shown in Figure 2. Table 3 shows the basic statistics of the data for primary 342 343 atmospheric state parameters, assuming that two measurements from the G1 and HALO have a proportional relationship without any offset (Y=m0\*X). In general, the atmospheric state 344 parameters observed from both aircraft were in excellent agreement. The linear regression 345 achieved a slope that was near 1 for four individual measurements. The regression is evaluated 346 using the equation below. 347

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

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).

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 humidity, the G1 data were slightly higher than the HALO data. The main contributions to the observed differences include the error propagation in the derivation of the ambient temperature 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<sup>-1</sup> higher than

the average horizontal wind speed measured by the G1. The uncertainty source of wind estimation 360 is mainly due to the error propagation from the indicated aircraft speed measurement and the 361 362 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). 363 Part of the reason for this difference is a more substantial variation of the G1 altitude during level 364 flight legs when the G1 flew at around 50 m/s faster than its normal airspeed. Thus, any biases 365 caused by their near side-by-side airspeeds being different from their typical airspeeds would be 366 undetected during these coordinated flights. 367

For the coordinated flights under cloudy conditions, we used the criteria from Section 2 to 368 compare ambient conditions measured by the G1 and HALO aircraft. In addition to the ordinary 369 linear regression, we also used the orthogonal regression to minimize the perpendicular distances 370 371 from the data points to the fitted line. The ordinary linear regression assumes only the response (Y) variable contains measurement error but not the predictor (X), which remains unknown when 372 373 we start the comparison between the measurements from the G1 and HALO. Thus, the additional orthogonal regression examines the assumption in the least square regression and makes sure the 374 375 roles of the variables have little influence on the results. In Table 4, two equations were used for the orthogonal regression. One assumes that two measurements have a proportional relationship 376 377 (Y=m1\*X). The other one assumes a linear relationship, which can be described with the slopeintercept equation Y=m\*X+b. Two regression results in Table 4 don't show a significant 378 379 difference. The regression using the slope-intercept equation shows a different level of improvement in each individual measurement and will be discussed in the corresponding sections. 380 As shown in Figure 4, the linear regression slopes for ambient temperature (Figure 4(a)), 381

pressure (Figure 4 (b)), and dew point temperature were also close to 1 between the G1 and HALO 382 measurements during the September 21 coordinated flight. The  $R^2$  value is also close to 1. These 383 384 results suggest that the G1 and HALO measurements achieved excellent agreements. Note that the dew point temperature from the G1 measurement was erroneous and removed from the comparison 385 the data points between 2200–2700 m and above 3700 m (Figure 4(c)) because the G1 sensor was 386 skewed by wetting in the cloud. The HALO dew point temperature was calculated from the total 387 388 water mixing ratio measured by TDL, and that measurement in the cloud was more accurate than the measurement made by the chilled mirror hygrometer aboard the G1. 389

The lower value of the  $R^2$  value in horizontal wind speed means the ratio of the regression error 390 and total error in wind measurement is much higher than the temperature and pressure 391 392 measurements. The main contributions to this difference are the error propagation during the 393 horizontal wind speed estimation and the temporal and spatial variance between two aircraft sampling locations. We observed differences between the two aircraft data of up to 2 m s<sup>-1</sup>, caused 394 by the increasing sampling distance as the two aircraft were climbing up. For example, the G1 395 flew a level leg above T3 around 2500 m between 16:20-16:30, while HALO stayed around 2500 396 m for a short period and kept climbing to a higher altitude. Due to strong vertical motion, 397 turbulence, and different saturations (evaporation-condensation processes), the variances in the 398 399 horizontal wind speed (Figure 4(d)) were also more significant compared to the variances of temperature and pressure measurements. 400

#### 401 3.2 Comparison of trace gas measurements

For the cloud-free coordinated flight on September 9, ozone is the only trace gas measurement 402 available on both aircraft. The linear regression slope shows that the HALO ozone concentration 403 was about 8% higher than the G1 concentration. The difference between the averaged ozone 404 concentrations was 4.1 ppb. As mentioned in section 2.1.2, each instrument has a 2 ppb accuracy 405 406 (or 5%) on the ground based on a direct photometric measurement measuring the ratio between 407 a sample and an ozone-free cell. The in-flight calibration suggested that the accuracy of each instrument could raise to 5-7% (or 2-3.5 ppb). Thus, the difference between the averaged ozone 408 409 concentrations – 4.1 ppb is within the instrument variation. The primary source of bias is probably 410 the different ozone loss in the sampling and transfer lines.

The comparison made on September 21 flight in Figure 5 shows good agreement for the 411 412 vertically averaged ozone measurements. Comparing the statistics data from September 9, the ozone measurement is not sensitive to the temporal and spatial changes. Although we do not have 413 the comparison data on September 9, the G1 and HALO CO measurements comparison show a 414 higher correlation than the ozone data comparison at different altitudes on September 21. Note 415 that the data points with more substantial variance in CO concentration were excluded because the 416 G1 and HALO were sampling different air masses between 2000-3000 m, as indicated in Figure 417 S3-S5. The CO plot in Figure (5b) shows the real atmospheric variability. Around 4000 m, the CO 418 reading from the G1 and HALO has the minimum variation and is averaged around 85 ppb, which 419

is at the atmospheric background level. At lower altitudes and higher CO concentrations, the local
contribution is not well-mixed, and the inhomogeneity expresses as the more substantial variations
observed in the plot.

#### 423 3.3 Comparison of aerosol measurements

424 Aerosol particles exhibited substantial spatial variations, both vertically and horizontally, due 425 to many aerosol sources and complex atmospheric processes in the Amazon basin, especially with 426 the local anthropogenic sources in Manaus. Thus, spatially resolved measurements are critical to 427 characterizing the properties of the Amazonian aerosols. The cloud-free coordinated flights allow us to compare the G1 and HALO aerosol measurements and thus will facilitate further studies that 428 utilize the airborne measurements. The vertical profiles obtained using the G1 and HALO in 429 different aerosol regimes of the Amazon basin have contributed to many studies (Fan et al., 2018; 430 431 Martin et al., 2017; Wang et al., 2016).

The design and performance of the aircraft inlets can strongly influence measured aerosol 432 particle number concentration, size distribution, and chemical composition (Wendisch et al., 433 2004). Therefore, they need to be taken into consideration when comparing the measurements 434 aboard two aircraft. The G1 aerosol inlet is a fully automated isokinetic inlet. Manufacturer wind 435 tunnel tests and earlier studies show that this inlet operates for aerosol particles with a diameter up 436 to 5 µm, with transmission efficiency around 50 % at 1.5 µm (Dolgos and Martins, 2014; Kleinman 437 et al., 2007; Zaveri et al., 2010). The HALO sub-micrometer Aerosol Inlet (HASI) was explicitly 438 designed for HALO. Based on the numerical flow modeling, optical particle counter 439 measurements, and field study evaluation, HASI has a cut-off size of 3 µm, with transmission 440 efficiency larger than 90 % at 1 µm (Andreae et al., 2018; Minikin et al., 2017). 441

442 3.3.1 Aerosol particle number concentration

For the cloud-free coordinated flight on September 9, the linear regression of CPC and UHSAS between the G1 and HALO measurements are also included in Table 3. The total number concentration measured by HALO CPC was about 20 % lower than that by the G1 CPC, as shown in Figure 6 (a). The CPC measurement is critically influenced by the isokinetic inlet operation and performance. During the flights, the aircraft attitude, such as the pitch and roll angles, will cause the isokinetic sampling under non-axial conditions. The non-axial flow at the probe inlet may result

in flow separation, turbulence, and particle deposition. Therefore, quantitative particle 449 measurements have more substantial uncertainty. As shown in Figure 6 (b), we compared the CPC 450 451 data by applying three different data quality criteria. The first criterion is the same criterion described in the previous section that makes sure all the compared measurements happen in less 452 than 30 minutes apart, and the linear regression is included in Table 3. The second criterion 453 454 constrains the data under the isokinetic and iso-axial condition and the plot in Figure 6(b) shows the iso-axial criteria reduced the broadness of the scattered data, but no significant change to the 455 456 linear regression. We further constrained the data with the averaging. Based on the average wind speed and distance between two aircraft, we averaged the data into 10 seconds interval and found 457 that the regression  $R^2$  increase to 0.9392. The typical uncertainty between two CPCs is 5-10% on 458 the well-controlled environment (Gunthe et al., 2009; Liu and Pui, 1974). Although both CPCs 459 460 from the G1 and HALO were characterized in the lab to be within 10% with its respective lab standard, we observed a 20% variance during the flight. This result suggests the challenging 461 462 condition of airborne measurement can significantly increase the systematic uncertainties of CPC measurements, such as systematic instrument drifts, different aerosol particle losses inside of the 463 464 two CPCs, and different inlet transmission efficiencies in the two aircraft.

The CPC data in Figure 6 are color-coded with UTC time. The general trend is that the aerosol 465 466 number concentration increased with time through the Manaus plume between 15:30 and 15:40. A similar trend was observed in aerosol particle number concentration (Figure 7) measured by the 467 468 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 overlapping 469 470 size range (90 – 500 nm for the September 9 flight) for both the G1 and HALO UHSAS. The linear regression shows that the total aerosol particle number concentration from HALO UHSAS is about 471 472 16.5% higher than that from the G1 UHSAS. The discrepancy between the two UHSAS 473 measurements is mainly due to the error propagation in the sampling flow, the differential pressure transducer reading, the instrument stability, and calibration repeatability, consistent with the other 474 475 UHSAS study (Kupc et al., 2018). In the airborne version of UHSAS, mechanical vibrations have 476 a more significant impact on the pressure transducer reading than the case for the bench version of UHSAS. 477

For the coordinated flight on September 21, the G1 and HALO data are averaged to 200 m
vertical altitude intervals (Figure 8). The data points with an altitude between 2000 – 3000 m

altitude were excluded from the comparisons, because the G1 and HALO sampled different 480 airmasses, as evidenced from trace gas and aerosol chemical composition data (detailed in Section 481 482 3.2 and 3.3.3). The UHSAS size range was integrated from 100 to 700 nm on September 21. The variation of the size range was because the overlap of size distributions from both UHSAS 483 instruments was changed. Both the CPC and UHSAS measurement comparisons show stronger 484 variation at the low altitude, especially below 2000 m. Above 3500 m, the variations on the CPC 485 and UHSAS measured concentration became significantly smaller than the variation at the lower 486 altitude. This result is consistent with the observation from the trace gas measurement and confirms 487 that the variability of aerosol properties changes significantly with time and space. It is noticeable 488 that the discrepancy observed in the UHSAS measurements comparison is larger than that from 489 the CPC comparison. That is because the aerosol flow control inside the UHSAS can't respond 490 491 quickly enough to the rapid change of the altitude and caused significant uncertainty in the data.

### 492 3.3.2 Aerosol particle size distribution

For the cloud-free coordinated flight on September 9, the averaged aerosol size distributions 493 measured by FIMS, G1 UHSAS, and HALO UHSAS during one flight leg are compared in Figure 494 9. For particle diameter below 90 nm, the G1 UHSAS overestimated the particle concentration, 495 which is due to the uncertainty in counting efficiency correction. The UHSAS detection efficiency 496 is close to 100% for particles larger than 100 nm and concentrations below 3000 cm<sup>-3</sup> but decreases 497 considerably for both smaller particles and higher concentrations (Cai et al., 2008). The aerosol 498 counting efficiency correction developed under the lab conditions does not necessarily apply under 499 the conditions during the flight. Between 90 nm and 250 nm, FIMS agreed well with the G1 500 501 UHSAS, whereas HALO UHSAS was about 30 % higher than the two instruments. For the size range of 250-500 nm, FIMS had good agreement with HALO UHSAS and was about 30-50 % 502 higher than the G1 UHSAS depending on the particle size. Because the UHSAS has a simplified 503 "passive" inlet, the large size aerosol particle loss in the UHSAS inlet was expected to increase 504 505 with the increasing of the aircraft speed. Thus, the lower G1 UHSAS concentrations at a larger 506 aerosol particle size are likely related to the particle loss correction.

507 For the September 21 flight, the vertical profiles of aerosol size distributions are averaged into 508 100 m altitude intervals (Figure 10). Overall, all size distribution measurements captured the mode 509 near 100 nm between 800–1000 m at the top of the convective boundary layer, as indicated by the potential temperature (Figure 10(d)), which starts from a maximum near the ground and then becomes remarkably uniform across the convective boundary layer. The peak of the aerosol size distribution shifted from 100 nm to 150 nm with increasing altitude. Note that due to data availability, the aerosol size distribution data from the HALO UHSAS has a reduced vertical resolution.

515 3.3.3 Aerosol particle chemical composition

516 Figure 11 shows the vertical profiles of the aerosol mass concentrations measured by the two 517 AMS on September 21. The upper panel shows the medians and interquartile ranges of the different species (organics, nitrate, sulfate, ammonium) and the total mass concentration for the G1 (circles) 518 519 and HALO (triangles). The lower panel shows the difference between the medians of G1 and 520 HALO. The error bars were calculated using error propagation from the error of the median 521 (interquartile range divided by 2\*sqrt(N)). The data were grouped into 400 m altitude bins. The total mass concentration is the highest in the lower altitudes between 100 m and 2000 m with a 522 median value of 5  $\mu$ g m<sup>-3</sup> (G1-AMS). At altitudes between 2000 m and 3800 m, the aerosol mass 523 concentration decreased to a median value of  $1.2 \,\mu g \, m^{-3}$  (G1-AMS). 524

The most significant difference was observed at altitudes below 1800 m. The aerosol mass concentration measured by HALO-AMS is less than that measured by G1 AMS, likely due to particle losses in the constant pressure inlet (CPI) used on the HALO AMS. Between 1800 m and 3000 m, the mass concentrations measured by the HALO AMS exceed those measured by the G1-AMS. This is most likely because the G1 was sampling different air masses than the HALO, as indicated by the differences in CO mixing ratios and the CPC concentrations for this altitude region (see Fig. 5 and 8). Above 3000 m altitude, both instruments agree within the uncertainty range.

Among individual species, the largest difference above 2000 m is observed for ammonium. The deployed G1 AMS is a high-resolution mass spectrometer (HR-ToF), whereas the HALO AMS has a lower resolution (C-ToF). The higher resolution of the G1-AMS allows for a better separation of interfering ions at m/z 15, 16, and 17 (NH<sup>+</sup>, NH<sup>2+</sup>, NH<sup>3+</sup>) and thereby a more reliable calculation of the ammonium mass concentration.

537 Overall the aerosol chemical composition is dominated by organics, as is evident from the 538 vertical profiles of the relative fractions (Fig. 12). Both AMS show a dominant contribution of

organics to the total mass concentration with values around 70 %. This contribution is constant at 539 altitudes between 100 m and 3500 m and decreases to 50 % at 3800 m altitude. The inorganic 540 fraction has the highest contribution from sulfate (20%), followed by ammonium (7%) and nitrate 541 (2 - 4%). For organics, ammonium, and sulfate, both instruments give similar relative fractions, 542 only for nitrate where a discrepancy is observed between 1000 and 3000 m. Although the absolute 543 aerosol mass concentration measured by the HALO-AMS was affected by the constant pressure 544 inlet below 1800 m altitude, the relative fractions of both instruments generally agree well. Similar 545 546 results were found for a second comparison flight on October 1, 2014 (see supplemental section 2 and plots S7, S8). 547

548 3.3.4 CCN number concentration

These measurements provide key information about the aerosol's ability to form cloud droplets 549 550 and thereby modify the microphysical properties of clouds. Numerous laboratory and field studies have improved the understanding of the connections among aerosol particle size, chemical 551 composition, mixing states and CCN activation properties (Bhattu and Tripathi, 2015; 552 Broekhuizen et al., 2006b; Chang et al., 2010; Duplissy et al., 2008; Lambe et al., 2011; Mei et al., 553 554 2013a; Mei et al., 2013b; Pöhlker et al., 2016; Thalman et al., 2017). In addition, based on the simplified chemical composition and internal mixing state assumption, various CCN closure 555 studies have achieved success within ±20% uncertainty for ambient aerosols (Broekhuizen et al., 556 2006a; Mei et al., 2013b; Rissler et al., 2004; Wang et al., 2008). 557

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 ( $\kappa_{CCN}$ ) of CCN in the Amazon basin is usually dominated by organic components ( $\kappa_{Org}$ ). Long-term ground-based measurements at the Amazon Tall Tower Observatory also suggest low temporal variability and lack of pronounced diurnal cycles in hygroscopicity only under natural rainforest background conditions (Pöhlker et al., 2018; Pöhlker et al., 2016).

Using FIMS and CCN data from both the G1 and HALO collected during the coordinated flight leg on September 9, the critical activation dry diameter ( $D_{50}$ ) was determined by integrating FIMS size distribution to match the CCN total number concentration (section S3). Then, the effective particle hygroscopicity was derived from  $D_{50}$  and the CCN-operated supersaturation using the  $\kappa$ - Köhler theory. The histogram plots based on the density of the estimated hygroscopicity ( $\kappa_{est}$ ) from both aircraft were compared for the flight leg above T3. The  $\kappa_{est}$  values derived from the G1 and HALO measurements during the flight leg above the T3 site are  $0.19\pm0.07$  and  $0.19\pm0.08$ , respectively. Those values agree very well with the overall mean value of  $0.17\pm0.06$  derived from long-term measurements at the Amazon Tall Tower Observatory (Pöhlker et al., 2016; Thalman et al., 2017).

A comparison of the vertical profiles of the CCN concentrations at 0.5% supersaturation on September 21 is shown in Figure 13 as an example. The difference between the CCN measurements on the two aircraft is about 20% on average. The linear regression slope would increase to 0.9120 if we focused on the data above 2500 m. The main contributions to the difference include the difference in aerosol inlet structure, aerosol particle loss correction in the main aircraft inlet, and the constant pressure inlet, the systematic inlet difference below 2500 m as shown in AMS data, as well as the error propagation of CCN measurements.

#### 581 3.4 Comparison of cloud measurements

582 In situ cloud measurements help to capture the diversity of different cloud forms and their natural temporal and spatial variability. The G1 CDP and FCDP were deployed under the different 583 wing pylons, and also on the different side of the aircraft. The G1 2DS was deployed on the same 584 side of FCDP. The HALO cloud combination probe (CCP-CDP and CCP-CIPgs) and NIXE-CAPS 585 (NIXE-CAS and NIXE-CIPgs) were deployed under the different wing pylons but on the same 586 side of the aircraft. On September 21, 2014, based on the aircraft location and elevation 587 information as shown in Figure 1(b) and Figure 3, both aircraft were sampling above T3 site and 588 passing through the same cloud field at ~1600 m flight leg and ~1900 m flight leg as shown in 589 Figure S11 and Figure S12. We used the cloud probes data from ~1900 m flight leg for the cloud 590 droplet number concentration comparison. Two size ranges were considered: 3-20 µm from light 591 scattering probes (CDP vs. FCDP on the G1, CCP-CDP vs. NIXE-CAS on HALO) and 2-960 µm 592 593 from combined cloud probes.

## 3.4.1 Comparison of cloud droplet number concentration between 3-20 $\mu m$

For underwing cloud probes, such as the CDP and the CAS, Lance (Lance, 2012) suggests an 595 undercounting bias of measured particle number concentration by up to 44% due to coincidence 596 597 as soon as the ambient cloud particle density rises to 1000 per cm<sup>3</sup>. At identical cloud particle densities, an earlier study (Baumgardner et al., 1985) estimates the coincidence bias for underwing 598 cloud probes to the range at 20%. Factually, the coincidence correction depends on the 599 600 instruments' individual detection volume, the air's volume flow rate through the detector, and the cloud particles' residence time within the detection volume (Hermann and Wiedensohler, 2001; 601 602 Jaenicke, 1972). For this comparison, coincidence bias remained unconsidered for each of the 603 cloud probe measurements to avoid deviations that are caused by the application of different corrections. 604

605 The primary cloud layer was observed by both the G1 and HALO between 1000-2500 m above ground. Although the two aircraft have sampled along the same flight path, the instruments 606 607 probably observed different sets of the cloud due to cloud movement with the prevailing wind or different cloud evolution stages. Thus, an initial comparison focuses on the redundant instruments 608 609 on the same aircraft, that measured truly collocated and synchronous on board of HALO and of the G1, respectively. In Figure 14 (a), the data of the CCP-CDP and of the NIXE-CAS are 610 juxtaposed sampled over about 13 minutes for particle detection size ranges which were considered 611 as most equivalent. The comparison reveals two ranges of particle number concentrations at which 612 densification of agreeing measurements become visible. At very low number concentrations (about 613  $10^{-1} - 10$  per cm<sup>3</sup>), the presence of inactivated (interstitial) aerosols in the clear air space between 614 the very few cloud elements should be considered. Over specific ranges, however, the fine 615 616 structure of varying cloud droplet number concentration may cause the regression's scattering, 617 indicated by cloud particle measured by one instrument whilst respective antagonist seems to measure within almost clear air – and vice versa. At higher number concentrations, i.e., between 618 619  $10^2$  and  $10^3$  per cm<sup>3</sup>, the comparison of the highly resolved data constitutes increasing compactness with respect to the 1:1 line. The overall data scatter of this comparison, however, may indicate the 620 621 highly variable structure within clouds as those investigated over the Amazon basin. The data of 622 the G1 CDP and the FCDP are juxtaposed as the same as HALO cloud probes. However, the sampled cloud period was much shorter - about 3 minutes. Similar to the HALO cloud probes 623 624 comparison, we observe two ranges of particle number concentrations at which densification of agreeing measurements become visible, especially for the lower number concentrations (Figure 625

626 14(b)). At higher number concentrations, only a few cloud elements were observed by the G1
627 cloud probes. That is because the G1 was about 7-23 minutes later to pass the same location as
628 HALO, and experienced much fewer cloud elements.

629 3.4.2 Comparison of cloud droplet size distribution between 2-960 μm from both aircraft

Comparing the cloud probes from the G1 and HALO, the size distributions from the HALO 630 CCP and NIXE-CAPS probes are in remarkably good agreement between 2-960 µm, and both 631 peaked around 10 µm, as shown in Figure 15. That is because the potential effects of cloud 632 elements shattering on the probe measurements were considered similarly for the HALO-deployed 633 634 CCP and NIXE instruments. On the G1, the CDP and FCDP had a more significant difference in the size range of less than 8 µm, although both of them peaked between 10-20 µm. The difference 635 between the G1 CDP and FCDP is mainly due to the data post-processing. The G1 CDP used an 636 637 old data acquisition system from the Science Engineering Associates, which limited its capability 638 to store the particle-by-particle (PBP) data for further processing. The CDP had placed an 800µm-diameter pinhole in front of the sizing detector to minimize the coincidence up to 1850 cm<sup>-3</sup>. 639 640 On the other side, the FCDP was equipped with new electronics and PBP data was locally stored 641 in a flash drive onboard the Linux machine. For the G1 flights, a constant probe-dependent 642 adjustment factor was applied to FCDP to adjust the coincidence further in the final data product. The G1 CDP and FCDP operated with a redesigned probe tip to minimize the shattering effect. An 643 644 additional algorithm was applied to the FCDP data to eliminate particles with short interarrival 645 times.

646 For cloud droplets larger than 20 µm, the difference between the obtained cloud particle size distributions from two aircraft becomes substantial (up to two orders of magnitude), which 647 648 indicated the observations of two different stages within the progressing development of a precipitation cloud. The precipitation cloud developing process is evidently expressed in elevated 649 650 number concentrations of larger cloud elements observed during the G1 measurement that happened later. We also observed that the general cloud characteristic is similar at different 651 altitude levels, as shown in Figure S13. The first two of three averaged periods were chosen during 652 the flight leg of ~1600 m and the last average period is for the flight leg ~1900 m compared in 653 654 Figure 15. Due to the averaging, the fine in-cloud structure gets suppressed. The small scale variabilities inside a cloud which are illustrated by the scattering of the highly resolved measurement data from the instrument comparison (cf. Figure 14) and the temporal evolution of in-cloud microphysics are not ascertainable and furthermore are beyond the scope of this study.

#### 658 3.5 Comparison of radiation measurements

659 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 660 661 300–1,800 nm wavelengths in Figure 16. Only measurements from flight legs, where the G1 and 662 HALO flew near side-by-side and at the same altitude were taken into consideration for analysis. In Figure 16, the top panel shows the time series of SPN-1 measurements, and the bottom panel 663 664 shows the time series of SMART-Albedometer measurements. The black dots represented all data, and the blue circles identified data when the navigation condition was within +/- 1 degree from the 665 666 horizontal level. The large scatter in the data between 15:12-15:28 and 15:35-15:40 is mainly due to the different sensor trajectories during the maneuvering of the aircraft to get to the coordinated 667 flight position. Because of the difference of each aircraft position from horizontal, the measured 668 signal varied from the signal of the direct component of sunlight. Each sensor might look at 669 670 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" 671 behavior in the time series plots of the downward irradiance. 672

673 Comparing the G1 and HALO measurements between 15:15-1:55 using the restricted 674 navigation criteria in Figure S14, we observed that the G1 SPN1 irradiance is slightly higher than 675 the integrated irradiance from the SMART-Albedometer. We used the NCAR tropospheric 676 ultraviolet and visible (TUV) radiation model estimated the weighted irradiance at 15:42:00 on 677 Sep 9, 2014, and confirm that the spectral variation in the instruments is the main contribution 678 to the difference in the comparison.

679

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

For the majority of the measurements during this field study, three primary sources contribute 688 689 to the measurement variation between the two aircraft: the temporal and spatial variations, the 690 difference in the inlet characterization, and the limitation of the instrument capability. We used 691 both ordinary least squares (OLS) linear regression and the orthogonal distance regression (ODR) to correlate the measurements from the G1 and HALO and confirmed that the slope and  $R^2$  are 692 693 very similar for the measurements made on September 9. The results from Table 2 confirmed that the G1 and HALO measurements should be in a linear relationship without an offset if there is no 694 695 altitude variation. It also shows the minimum discrepancy between two aerosol instruments (CPC or UHSAS) could be around 20%, which will include the error caused by the difference in the inlet 696 697 characterization and the limitation of the instrument capability. If we assume those two measurement variation sources are not affected by the altitude, then by comparing the linear 698 699 regression data from Table 3 to Table 2, we can estimate the temporal and spatial variation between two aircraft in a stack flight pattern. Three linear regression approaches were assessed, and the 700 701 results are listed in Table 3. If we assume that two measurements from the G1 and HALO should 702 not have any offsets, the OLS and ODR regressions show similar results. For the meteorological parameters, this assumption is valid. In addition, good correlations also indicate that there is no 703 704 significant temporal or spatial variation during the stack pattern flight. As expected, the wind speed 705 and the aerosol measurements show that the correlations between the measurements from the G1 706 and HALO significantly improved with the offset assumption. This result suggests that the 707 temporal and spatial variation in a half-hour will add an additional 20% variance to the measured 708 aerosol properties. This will lead to considerable uncertainty when we combine the observation 709 data between the ground station and the airborne platform. Thus, to evaluate or constrain 710 atmospheric modeling work, more routine and long-term airborne measurements should be used to provide statistically sufficient observation. 711

#### 713 **5** Summary

714 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 715 compared (Table S3). Overall, the analysis shows good agreement between the G1 and HALO 716 717 measurements for a relatively broad range of atmospheric-related variables in a challenging lower 718 troposphere environment. Measured variables included atmospheric state parameters, aerosol particles, trace gases, clouds, and radiation properties. This study outlines the well-designed 719 720 coordinated flights for achieving a meaningful comparison between two moving platforms. The 721 high data quality was ensured by the most sophisticated instruments aboard two aircraft used the 722 most advanced techniques, assisted with the best-calibrated/characterized procedures. The 723 comparisons and the related uncertainty estimations quantify the current measurement limits, 724 which provide guidance to the modeler to realistically quantify the modeling input value and evaluate the variation between the measurement and the model output. The comparison also 725 726 identified the measurement issues, outlined the associated reasonable measurement ranges, and evaluated the measurement sensitivities to the temporal and spatial variance. 727

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

744 that FIMS provides valuable size coverage for better CCN number concentration estimation. Based on the  $\kappa$ -Köhler parameterization,  $\kappa_{est}$  observed at 500 m above the T3 site is 0.19±0.08, which is 745 similar to the overall mean kappa from long-term ATTO measurements - 0.17±0.06 (Pöhlker et 746 747 al., 2016). This similarity suggests that there is no significant spatial variability along the 748 downwind transect, although the freshly emitted aerosol particles may have much less hygroscopicity. The difference in the ozone measurement comparison is about 4.1 ppb, which 749 750 suggests that the bias due to the sampling line loss inside of the G1 gas inlet. The irradiance from 751 the SPN1 unshaded center detector in the G1 was compared with the HALO integrated downward 752 irradiance between 300–1800 nm and achieved a very encouraging agreement with a variance of less than 10%. 753

During the second type of 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. 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 concentration and 760 761 the trace gas measurements both suggest inhomogeneous aerosol distribution between 2000-3000 m altitude. The integrated aerosol number concentration from UHSAS showed consistent 762 763 discrepancy at different altitudes. This considerable uncertainty in the UHSAS measurements is 764 caused by the significant aerosol sample flow variations due to the slow and unstable flow control. Although the aircraft-based UHSAS is a challenging instrument to operate, a reasonable size 765 distribution profile comparison was made between both UHSAS and FIMS on the G1. Overall the 766 767 chemical composition of the aerosol is dominated by organics. Around 70% of the AMS measured 768 mass is organic, and this fractional contribution is maintained from the surface to 3500 m, then decreases to 50% at higher altitudes. The most substantial difference among all the species is 769 770 observed for ammonium due to the different mass resolution of the AMS instruments, and more reliable ammonium mass concentration can be achieved with high resolution mass spectrometer. 771 772 Although the absolute aerosol mass concentration measured by the HALO AMS was affected 773 below 1800 m altitude by the constant pressure inlet, the relative fractions of both instruments 774 from the G1 and HALO agree well.

Cloud probe comparisons were made for the cloud droplet number concentration between 3– 775 776  $20 \,\mu m$  for the initial comparison between the redundant instruments on the same aircraft. Then the 777 comparison of cloud droplet size distribution between 2-960 µm for a flight leg around 1900 m 778 showed a remarkably good agreement. The major cloud appearance was captured by both aircraft, although the cloud elements observed were affected by the cloud movement with the prevailing 779 780 wind and the different cloud evolution stages. Furthermore, the relatively short time delay of 7-23 minutes between the independent measurements may give a hint for the time scales in which the 781 782 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.

Several efforts made by both airborne measurement teams have significantly contributed to theoverall success of this comparison study, and we recommend them for future field operations.

- Characterize instruments following the same established guideline. For example, the
   aerosol instruments can follow the guideline from the World Calibration Centre for
   Aerosol Physics (WCCAP).
- Periodically compare measurements from different instruments for consistency in the
  field. For example, we found that comparing the integrated aerosol volume distribution
  from the aerosol sizer with the converted total aerosol mass from the AMS
  measurement can help check both the instrument performances and the inlet operation
  condition. Additionally, measurements from different cloud probes should be
  compared in the overlapping size ranges.
- 3) Daily calibration would be valuable but likely unrealistic to perform in the field. One
  alternative is to daily even hourly monitor the variation of the critical instrument
  parameters, such as the aerosol sample flow of the individual aerosol instruments.
- 4) For the cases with minor variations in the calibration results, the typical practice is to
  use the average calibration results for the variation period. However, we also
  recommend documenting the corresponding uncertainty with the data product.

S) A side-by-side comparison among the similar instruments deployed at different
platforms, including those at ground sites, is highly recommended and will provide a
comprehensive view of the data reliability.

Acknowledgments: This study was supported by the U.S. DOE, Office of Science, Atmospheric 808 System Research Program, and used data from Atmospheric Radiation Measurement Aerial 809 Facility, a DOE Office of Science User Facility. The Pacific Northwest National Laboratory 810 811 (PNNL) is operated for DOE by Battelle under contract DE-AC05-76RL01830. This work was also supported by the Max Planck Society, the DFG (Deutsche Forschungsgemeinschaft, German 812 813 Research Foundation) HALO Priority Program SPP 1294, the German Aerospace Center (DLR), the FAPESP (São Paulo Research Foundation) Grants 2009/15235-8 and 2013/05014-0, and a 814 wide range of other institutional partners. The contributions from Micael A. Cecchini were funded 815

816 by FAPESP grant number 2017/04654-6.

## 817 **References:**

- Alfarra, M. R., Coe, H., Allan, J. D., Bower, K. N., Boudries, H., Canagaratna, M. R., Jimenez,
- J. L., Jayne, J. T., Garforth, A. A., and Li, S.-M.: Characterization of urban and rural organic

particulate in the lower Fraser valley using two aerodyne aerosol mass spectrometers, Atmos
Environ, 38, 5745-5758, 2004.

- Andreae, M. O., Acevedo, O. C., Araujo, A., Artaxo, P., Barbosa, C. G. G., Barbosa, H. M. J.,
- Brito, J., Carbone, S., Chi, X., Cintra, B. B. L., da Silva, N. F., Dias, N. L., Dias, C. Q., Ditas, F.,
- Ditz, R., Godoi, A. F. L., Godoi, R. H. M., Heimann, M., Hoffmann, T., Kesselmeier, J.,
- 825 Konemann, T., Kruger, M. L., Lavric, J. V., Manzi, A. O., Lopes, A. P., Martins, D. L.,
- 826 Mikhailov, E. F., Moran-Zuloaga, D., Nelson, B. W., Nolscher, A. C., Nogueira, D. S., Piedade,
- M. T. F., Pohlker, C., Poschl, U., Quesada, C. A., Rizzo, L. V., Ro, C. U., Ruckteschler, N., Sa,
- L. D. A., Sa, M. D., Sales, C. B., dos Santos, R. M. N., Saturno, J., Schongart, J., Sorgel, M., de
- Souza, C. M., de Souza, R. A. F., Su, H., Targhetta, N., Tota, J., Trebs, I., Trumbore, S., van
- 830 Eijck, A., Walter, D., Wang, Z., Weber, B., Williams, J., Winderlich, J., Wittmann, F., Wolff, S.,
- and Yanez-Serrano, A. M.: The Amazon Tall Tower Observatory (ATTO): overview of pilot
- measurements on ecosystem ecology, meteorology, trace gases, and aerosols, Atmos Chem Phys,
- 833 15, 10723-10776, 2015.
- Andreae, M. O., Afchine, A., Albrecht, R., Holanda, B. A., Artaxo, P., Barbosa, H. M. J.,
- Borrmann, S., Cecchini, M. A., Costa, A., Dollner, M., Fütterer, D., Järvinen, E., Jurkat, T.,
- 836 Klimach, T., Konemann, T., Knote, C., Krämer, M., Krisna, T., Machado, L. A. T., Mertes, S.,
- 837 Minikin, A., Pöhlker, C., Pöhlker, M. L., Pöschl, U., Rosenfeld, D., Sauer, D., Schlager, H.,
- 838 Schnaiter, M., Schneider, J., Schulz, C., Spanu, A., Sperling, V. B., Voigt, C., Walser, A., Wang,
- 339 J., Weinzierl, B., Wendisch, M., and Ziereis, H.: Aerosol characteristics and particle production
- in the upper troposphere over the Amazon Basin, Atmos. Chem. Phys., 18, 921-961, 2018.

- Artaxo, P., Rizzo, L. V., Brito, J. F., Barbosa, H. M. J., Arana, A., Sena, E. T., Cirino, G. G.,
- Bastos, W., Martin, S. T., and Andreae, M. O.: Atmospheric aerosols in Amazonia and land use
- change: from natural biogenic to biomass burning conditions, Faraday Discuss, 165, 203-235,
  2013.
- Bahreini, R., Dunlea, E. J., Matthew, B. M., Simons, C., Docherty, K. S., DeCarlo, P. F.,
- Jimenez, J. L., Brock, C. A., and Middlebrook, A. M.: Design and operation of a pressure-
- controlled inlet for airborne sampling with an aerodynamic aerosol lens, Aerosol Sci Tech, 42,
- 848 465-471, 2008.
- 849 Baumgardner, D., Strapp, W., and Dye, J. E.: Evaluation of the Forward Scattering Spectrometer
- Probe. Part II: Corrections for Coincidence and Dead-Time Losses, 2, 626-632, 1985.
- 851 Bhattu, D. and Tripathi, S. N.: CCN closure study: Effects of aerosol chemical composition and
- 852 mixing state, J Geophys Res-Atmos, 120, 766-783, 2015.
- Brenguier, J. L., Bachalo, W. D., Chuang, P. Y., Esposito, B. M., Fugal, J., Garrett, T., Gayet, J.
- F., Gerber, H., Heymsfield, A., and Kokhanovsky, A.: In situ measurements of cloud and
- precipitation particles, Airborne Measurements for Environmental Research: Methods and
- 856 Instruments, 2013. 225-301, 2013.
- 857 Broekhuizen, K., Chang, R.-W., Leaitch, W., Li, S.-M., and Abbatt, J.: Closure between
- 858 measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol
- compositions in downtown Toronto, Atmos Chem Phys, 6, 2513-2524, 2006a.
- Broekhuizen, K., Chang, R. Y. W., Leaitch, W. R., Li, S. M., and Abbatt, J. P. D.: Closure
- between measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol
- compositions in downtown Toronto, Atmos Chem Phys, 6, 2513-2524, 2006b.
- Cai, Y., Montague, D. C., Mooiweer-Bryan, W., and Deshler, T.: Performance characteristics of
- the ultra high sensitivity aerosol spectrometer for particles between 55 and 800 nm: Laboratoryand field studies, J Aerosol Sci, 39, 759-769, 2008.
- 866 Chang, R. Y. W., Slowik, J. G., Shantz, N. C., Vlasenko, A., Liggio, J., Sjostedt, S. J., Leaitch,
- W. R., and Abbatt, J. P. D.: The hygroscopicity parameter (kappa) of ambient organic aerosol at
  a field site subject to biogenic and anthropogenic influences: relationship to degree of aerosol
  arridation. Atmos Chem Phys. 10, 5047 5064, 2010.
- 869 oxidation, Atmos Chem Phys, 10, 5047-5064, 2010.
- 870 Costa, A., Meyer, J., Afchine, A., Luebke, A., Günther, G., Dorsey, J. R., Gallagher, M. W.,
- 871 Ehrlich, A., Wendisch, M., and Baumgardner, D.: Classification of Arctic, midlatitude and
- tropical clouds in the mixed-phase temperature regime, Atmos Chem Phys, 17, 12219-12238,
- 873 2017.
- Davidson, E. A., de Araujo, A. C., Artaxo, P., Balch, J. K., Brown, I. F., Bustamante, M. M. C.,
- 875 Coe, M. T., DeFries, R. S., Keller, M., Longo, M., Munger, J. W., Schroeder, W., Soares, B. S.,
- 876 Souza, C. M., and Wofsy, S. C.: The Amazon basin in transition (vol 481, pg 321, 2012), Nature,
- **877 483**, 232-232, 2012.
- B78 DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin,
- 879 M., Fuhrer, K., Horvath, T., Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.: Field-
- deployable, high-resolution, time-of-flight aerosol mass spectrometer, Anal Chem, 78, 8281-
- 881 8289, 2006.
- 882 Dolgos, G. and Martins, J. V.: Polarized Imaging Nephelometer for in situ airborne
- measurements of aerosol light scattering, Optics express, 22, 21972-21990, 2014.
- B84 Duplissy, J., Gysel, M., Alfarra, M. R., Dommen, J., Metzger, A., Prevot, A. S. H., Weingartner,
- E., Laaksonen, A., Raatikainen, T., Good, N., Turner, S. F., McFiggans, G., and Baltensperger,

- 886 U.: Cloud forming potential of secondary organic aerosol under near atmospheric conditions,
- 887 Geophys Res Lett, 35, 2008.
- 888 Faber, S., French, J. R., and Jackson, R.: Laboratory and in-flight evaluation of measurement
- uncertainties from a commercial Cloud Droplet Probe (CDP), Atmos Meas Tech, 11, 3645-3659,
  2018.
- Fan, J., Rosenfeld, D., Zhang, Y., Giangrande, S. E., Li, Z., Machado, L. A., Martin, S. T., Yang,
- Y., Wang, J., and Artaxo, P.: Substantial convection and precipitation enhancements by ultrafine
   aerosol particles, Science, 359, 411-418, 2018.
- Giez, A., Mallaun, C., Zöger, M., Dörnbrack, A., and Schumann, U.: Static pressure from aircraft
- trailing-cone measurements and numerical weather-prediction analysis, J Aircraft, 54, 17281737, 2017.
- 897 Gunthe, S., King, S., Rose, D., Chen, Q., Roldin, P., Farmer, D., Jimenez, J., Artaxo, P.,
- Andreae, M., and Martin, S.: Cloud condensation nuclei in pristine tropical rainforest air of
- 899 Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and
- 900 CCN activity, Atmos Chem Phys, 9, 7551-7575, 2009.
- 901 Guyon, P., Boucher, O., Graham, B., Beck, J., Mayol-Bracero, O. L., Roberts, G. C., Maenhaut,
- 902 W., Artaxo, P., and Andreae, M. O.: Refractive index of aerosol particles over the Amazon
- tropical forest during LBA-EUSTACH 1999, J Aerosol Sci, 34, 883-907, 2003.
- Hermann, M. and Wiedensohler, A. J. J. o. A. S.: Counting efficiency of condensation particle
- counters at low-pressures with illustrative data from the upper troposphere, 32, 975-991, 2001.
- Jaenicke, R. J. J. o. A. S.: The optical particle counter: cross-sensitivity and coincidence, 3, 95-111, 1972.
- Jayne, J. T., Leard, D. C., Zhang, X. F., Davidovits, P., Smith, K. A., Kolb, C. E., and Worsnop,
- D. R.: Development of an aerosol mass spectrometer for size and composition analysis of
- submicron particles, Aerosol Sci Tech, 33, 49-70, 2000.
- 911 Keller, M., Bustamante, M., Gash, J., and Dias, P. S. (Eds.): Amazonia and Global Change,
- 912 American Geophysical Union, Washington, DC, 2009.
- 913 Kleinman, L. I., Daum, P. H., Lee, Y. N., Senum, G. I., Springston, S. R., Wang, J., Berkowitz,
- 914 C., Hubbe, J., Zaveri, R. A., and Brechtel, F. J.: Aircraft observations of aerosol composition and
- 915 ageing in New England and Mid-Atlantic States during the summer 2002 New England Air
- 916 Quality Study field campaign, Journal of Geophysical Research: Atmospheres, 112, 2007.
- 917 Klingebiel, M., de Lozar, A., Molleker, S., Weigel, R., Roth, A., Schmidt, L., Meyer, J., Ehrlich,
- 918 A., Neuber, R., Wendisch, M., and Borrmann, S.: Arctic low-level boundary layer clouds: in situ
- 919 measurements and simulations of mono- and bimodal supercooled droplet size distributions at
- the top layer of liquid phase clouds, Atmos Chem Phys, 15, 617-631, 2015.
- Krautstrunk, M. and Giez, A.: The transition from FALCON to HALO era airborne atmospheric
   research. In: Atmospheric Physics, Springer, 2012.
- 923 Krisna, T. C., Wendisch, M., Ehrlich, A., Jäkel, E., Werner, F., Weigel, R., Borrmann, S.,
- 924 Mahnke, C., Pöschl, U., Andreae, M. O., Voigt, C., and Machado, L. A. T.: Comparing airborne
- and satellite retrievals of cloud optical thickness and particle effective radius using a spectral
- 926 radiance ratio technique: two case studies for cirrus and deep convective clouds, Atmos. Chem.
- 927 Phys., 18, 4439-4462, 2018.
- 928 Kuhn, U., Ganzeveld, L., Thielmann, A., Dindorf, T., Schebeske, G., Welling, M., Sciare, J.,
- 929 Roberts, G., Meixner, F. X., Kesselmeier, J., Lelieveld, J., Kolle, O., Ciccioli, P., Lloyd, J.,
- 930 Trentmann, J., Artaxo, P., and Andreae, M. O.: Impact of Manaus City on the Amazon Green

- 931 Ocean atmosphere: ozone production, precursor sensitivity and aerosol load, Atmos Chem Phys,
- 932 10, 9251-9282, 2010.
- 933 Kulkarni, P. and Wang, J.: New fast integrated mobility spectrometer for real-time measurement
- of aerosol size distribution I: Concept and theory, J Aerosol Sci, 37, 1303-1325, 2006a.
- 935 Kulkarni, P. and Wang, J.: New fast integrated mobility spectrometer for real-time measurement
- 936 of aerosol size distribution: II. Design, calibration, and performance characterization, J Aerosol
- 937 Sci, 37, 1326-1339, 2006b.
- 938 Kupc, A., Williamson, C., Wagner, N. L., Richardson, M., and Brock, C. A.: Modification,
- 939 calibration, and performance of the Ultra-High Sensitivity Aerosol Spectrometer for particle size
- 940 distribution and volatility measurements during the Atmospheric Tomography Mission (ATom)
- airborne campaign, Atmos. Meas. Tech., 11, 369-383, 2018.
- 242 Lambe, A. T., Onasch, T. B., Massoli, P., Croasdale, D. R., Wright, J. P., Ahern, A. T.,
- 943 Williams, L. R., Worsnop, D. R., Brune, W. H., and Davidovits, P.: Laboratory studies of the
- 944 chemical composition and cloud condensation nuclei (CCN) activity of secondary organic
- aerosol (SOA) and oxidized primary organic aerosol (OPOA), Atmos Chem Phys, 11, 8913-
- 946 8928, 2011.
- 947 Lance, S.: Coincidence Errors in a Cloud Droplet Probe (CDP) and a Cloud and Aerosol
- Spectrometer (CAS), and the Improved Performance of a Modified CDP, J Atmos Ocean Tech,29, 1532-1541, 2012.
- 950 Lance, S., Brock, C. A., Rogers, D., and Gordon, J. A.: Water droplet calibration of the Cloud
- Droplet Probe (CDP) and in-flight performance in liquid, ice and mixed-phase clouds during
- ARCPAC, Atmospheric Measurement Techniques, 3, 1683-1706, 2010.
- Lawson, R.: Effects of ice particles shattering on the 2D-S probe, Atmos Meas Tech, 4, 1361-1381, 2011.
- Lawson, R. P., O'Connor, D., Zmarzly, P., Weaver, K., Baker, B., Mo, Q., and Jonsson, H.: The
- 2D-S (stereo) probe: Design and preliminary tests of a new airborne, high-speed, high-resolution particle imaging probe, J Atmos Ocean Tech, 23, 1462-1477, 2006.
- 958 Liu, B. Y. and Pui, D. Y.: A submicron aerosol standard and the primary, absolute calibration of
- the condensation nuclei counter, Journal of Colloid and Interface Science, 47, 155-171, 1974.
- 960 Long;, C. N., Bucholtz;, A., Jonsson;, H., Schmid;, B., Vogelmann;, A., and Wood, J.: A Method
- 961 of Correcting for Tilt from Horizontal in Downwelling Shortwave Irradiance Measurements on
- 962 Moving Platforms, The Open Atmospheric Science Journal, 4, 78-87, 2010.
- 963 Luebke, A. E., Afchine, A., Costa, A., Grooss, J. U., Meyer, J., Rolf, C., Spelten, N., Avallone,
- L. M., Baumgardner, D., and Kramer, M.: The origin of midlatitude ice clouds and the resulting influence on their microphysical properties, Atmos Chem Phys, 16, 5793-5809, 2016.
- 966 Martin, S., Artaxo, P., Machado, L., Manzi, A., Souza, R., Schumacher, C., Wang, J., Andreae,
- 967 M., Barbosa, H., and Fan, J.: Introduction: observations and modeling of the Green Ocean
- 968 Amazon (GoAmazon2014/5), Atmos Chem Phys, 16, 2016a.
- 969 Martin, S. T., Artaxo, P., Machado, L., Manzi, A. O., Souza, R. A. F., Schumacher, C., Wang, J.,
- 970 Biscaro, T., Brito, J., Calheiros, A., Jardine, K., Medeiros, A., Portela, B., de Sa, S. S., Adachi,
- 971 K., Aiken, A. C., Albrecht, R., Alexander, L., Andreae, M. O., Barbosa, M. J., Buseck, P.,
- 972 Chand, D., Comstock, J. M., Day, D. A., Dubey, M., Fan, J., Fast, J., Fisch, G., Fortner, E.,
- 973 Giangrande, S., Gilles, M., Goldststein, A. H., Guenther, A., Hubbe, J., Jensen, M., Jimenez, J.
- 974 L., Keutsch, F. N., Kim, S., Kuang, C., Laskskin, A., McKinney, K., Mei, F., Miller, M.,
- 975 Nascimento, R., Pauliquevis, T., Pekour, M., Peres, J., Petaja, T., Pohlker, C., Poschl, U., Rizzo,
- 976 L., Schmid, B., Shilling, J. E., Dias, M. A. S., Smith, J. N., Tomlmlinson, J. M., Tota, J., and

- 977 Wendisch, M.: The Green Ocean Amazon Experiment (Goamazon2014/5) Observes Pollution
- Affecting Gases, Aerosols, Clouds, and Rainfall over the Rain Forest, B Am Meteorol Soc, 98,
  981-997, 2017.
- 980 Martin, S. T., Artaxo, P., Machado, L. A. T., Manzi, A. O., Souza, R. A. F., Schumacher, C.,
- 981 Wang, J., Andreae, M. O., Barbosa, H. M. J., Fan, J., Fisch, G., Goldstein, A. H., Guenther, A.,
- Jimenez, J. L., Poschl, U., Dias, M. A. S., Smith, J. N., and Wendisch, M.: Introduction:
- 983 Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5), Atmos Chem
- 984 Phys, 16, 4785-4797, 2016b.
- 985 Mei, F., Hayes, P. L., Ortega, A., Taylor, J. W., Allan, J. D., Gilman, J., Kuster, W., de Gouw, J.,
- Jimenez, J. L., and Wang, J.: Droplet activation properties of organic aerosols observed at an
- urban site during CalNex-LA, J Geophys Res-Atmos, 118, 2903-2917, 2013a.
- 988 Mei, F., Setyan, A., Zhang, Q., and Wang, J.: CCN activity of organic aerosols observed
- downwind of urban emissions during CARES, Atmos Chem Phys, 13, 12155-12169, 2013b.
- 990 Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of
- 991 composition-dependent collection efficiencies for the aerodyne aerosol mass spectrometer using
- 992 field data, Aerosol Sci Tech, 46, 258-271, 2012.
- 993 Minikin, A., Sauer, D., Ibrahim, A., Franke, H., Röschenthaler, T., Fütterer, D. A., and Petzold,
- 994 A.: The HALO Submicrometer Aerosol Inlet (HASI): Design concept and first characterization,
- 1st HALO symposium: Airborne Research with HALO: Achievements and Prospects,
- 996 Oberpfaffenhofen, Deutschland,, 2017. 2017.
- 997 Molleker, S., Borrmann, S., Schlager, H., Luo, B., Frey, W., Klingebiel, M., Weigel, R., Ebert,
- 998 M., Mitev, V., Matthey, R., Woiwode, W., Oelhaf, H., Dornbrack, A., Stratmann, G., Grooss, J.
- 999 U., Gunther, G., Vogel, B., Muller, R., Kramer, M., Meyer, J., and Cairo, F.: Microphysical
- 1000 properties of synoptic-scale polar stratospheric clouds: in situ measurements of unexpectedly
- large HNO3-containing particles in the Arctic vortex, Atmos Chem Phys, 14, 10785-10801,2014.
- 1003 Olfert, J. S., Kulkarni, P., and Wang, J.: Measuring aerosol size distributions with the fast 1004 integrated mobility spectrometer, J Aerosol Sci, 39, 940-956, 2008.
- 1005 Petzold, A., Marsh, R., Johnson, M., Miller, M., Sevcenco, Y., Delhaye, D., Ibrahim, A.,
- 1006 Williams, P., Bauer, H., Crayford, A., Bachalo, W. D., and Raper, D.: Evaluation of Methods for
- Measuring Particulate Matter Emissions from Gas Turbines, Environ Sci Technol, 45, 3562-3568, 2011.
- 1009 Pöhlker, M. L., Ditas, F., Saturno, J., Klimach, T., de Angelis, I. H., Araujo, A. C., Brito, J.,
- 1010 Carbone, S., Cheng, Y. F., Chi, X. G., Ditz, R., Gunthe, S. S., Holanda, B. A., Kandler, K.,
- 1011 Kesselmeier, J., Konemann, T., Kruger, O. O., Lavric, J. V., Martin, S. T., Mikhailov, E., Moran-
- 1012 Zuloaga, D., Rizzo, L. V., Rose, D., Su, H., Thalman, R., Walter, D., Wang, J., Wolff, S.,
- 1013 Barbosa, H. M. J., Artaxo, P., Andreae, M. O., Pöschl, U., and Pöhlker, C.: Long-term
- 1014 observations of cloud condensation nuclei over the Amazon rain forest Part 2: Variability and
- 1015 characteristics of biomass burning, long-range transport, and pristine rain forest aerosols, Atmos
- 1016 Chem Phys, 18, 10289-10331, 2018.
- 1017 Pöhlker, M. L., Pöhlker, C., Ditas, F., Klimach, T., de Angelis, I. H., Araujo, A., Brito, J.,
- 1018 Carbone, S., Cheng, Y. F., Chi, X. G., Ditz, R., Gunthe, S. S., Kesselmeier, J., Konemann, T.,
- 1019 Lavric, J. V., Martin, S. T., Mikhailov, E., Moran-Zuloaga, D., Rose, D., Saturno, J., Su, H.,
- 1020 Thalman, R., Walter, D., Wang, J., Wolff, S., Barbosa, H. M. J., Artaxo, P., Andreae, M. O., and
- 1021 Poschl, U.: Long-term observations of cloud condensation nuclei in the Amazon rain forest Part

- 1: Aerosol size distribution, hygroscopicity, and new model parametrizations for CCN 1022
- prediction, Atmos Chem Phys, 16, 15709-15740, 2016. 1023
- Poschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., 1024
- 1025 Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E.,
- Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S. 1026
- R., Artaxo, P., and Andreae, M. O.: Rainforest aerosols as biogenic nuclei of clouds and 1027
- precipitation in the Amazon, Science, 329, 1513-1516, 2010. 1028
- Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L., and Artaxo, P.: 1029
- Physical properties of the sub-micrometer aerosol over the Amazon rain forest during the wet-to-1030
- dry season transition-comparison of modeled and measured CCN concentrations, Atmos Chem 1031 1032 Phys, 4, 2119-2143, 2004.
- 1033 Roberts, G. C., Andreae, M. O., Zhou, J., and Artaxo, P.: Cloud condensation nuclei in the
- Amazon Basin:"Marine" conditions over a continent?, Geophys Res Lett, 28, 2807-2810, 2001. 1034
- Roberts, G. C., Artaxo, P., Zhou, J., Swietlicki, E., and Andreae, M. O.: Sensitivity of CCN 1035
- 1036 spectra on chemical and physical properties of aerosol: A case study from the Amazon Basin,
- Journal of Geophysical Research: Atmospheres, 107, LBA 37-31-LBA 37-18, 2002. 1037
- 1038 Rose, D., Gunthe, S., Mikhailov, E., Frank, G., Dusek, U., Andreae, M. O., and Pöschl, U.:
- Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei 1039
- counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol 1040
- 1041 particles in theory and experiment, Atmos Chem Phys, 8, 1153-1179, 2008.
- Salati, E. and Vose, P. B.: Amazon basin: a system in equilibrium, Science, 225, 129-138, 1984. 1042
- Schmid, B., Tomlinson, J. M., Hubbe, J. M., Comstock, J. M., Mei, F., Chand, D., Pekour, M. S., 1043
- 1044 Kluzek, C. D., Andrews, E., Biraud, S. C., and Mcfarquhar, G. M.: The Doe Arm Aerial Facility, 1045 B Am Meteorol Soc, 95, 723-+, 2014.
- 1046 Schulz, C., Schneider, J., Amorim Holanda, B., Appel, O., Costa, A., de Sá, S. S., Dreiling, V.,
- 1047 Fütterer, D., Jurkat-Witschas, T., Klimach, T., Knote, C., Krämer, M., Martin, S. T., Mertes, S.,
- Pöhlker, M. L., Sauer, D., Voigt, C., Walser, A., Weinzierl, B., Ziereis, H., Zöger, M., Andreae, 1048
- M. O., Artaxo, P., Machado, L. A. T., Pöschl, U., Wendisch, M., and Borrmann, S.: Aircraft-1049
- based observations of isoprene-epoxydiol-derived secondary organic aerosol (IEPOX-SOA) in 1050
- the tropical upper troposphere over the Amazon region, Atmos. Chem. Phys., 18, 14979-15001, 1051 2018. 1052
- Shilling, J. E., Pekour, M. S., Fortner, E. C., Artaxo, P., Sá, S. d., Hubbe, J. M., Longo, K. M., 1053
- 1054 Machado, L. A., Martin, S. T., and Springston, S. R.: Aircraft observations of the chemical
- composition and aging of aerosol in the Manaus urban plume during GoAmazon 2014/5, Atmos 1055
- Chem Phys, 18, 10773-10797, 2018. 1056
- Shilling, J. E., Zaveri, R. A., Fast, J. D., Kleinman, L., Alexander, M., Canagaratna, M. R., 1057
- Fortner, E., Hubbe, J. M., Jayne, J. T., and Sedlacek, A.: Enhanced SOA formation from mixed 1058
- anthropogenic and biogenic emissions during the CARES campaign, Atmos Chem Phys, 13, 1059 2091-2113, 2013.
- 1060
- 1061 Thalman, R., de Sa, S. S., Palm, B. B., Barbosa, H. M. J., Pohlker, M. L., Alexander, M. L.,
- Brito, J., Carbone, S., Castillo, P., Day, D. A., Kuang, C. G., Manzi, A., Ng, N. L., Sedlacek, A. 1062
- 1063 J., Souza, R., Springston, S., Watson, T., Pohlker, C., Poschl, U., Andreae, M. O., Artaxo, P.,
- 1064 Jimenez, J. L., Martin, S. T., and Wang, J.: CCN activity and organic hygroscopicity of aerosols
- downwind of an urban region in central Amazonia: seasonal and diel variations and impact of 1065
- 1066 anthropogenic emissions, Atmos Chem Phys, 17, 11779-11801, 2017.

- Wang, J.: A fast integrated mobility spectrometer with wide dynamic size range: Theoreticalanalysis and numerical simulation, J Aerosol Sci, 40, 890-906, 2009.
- 1069 Wang, J., Krejci, R., Giangrande, S., Kuang, C., Barbosa, H. M., Brito, J., Carbone, S., Chi, X.,
- 1070 Comstock, J., Ditas, F., Lavric, J., Manninen, H. E., Mei, F., Moran-Zuloaga, D., Pohlker, C.,
- 1071 Pohlker, M. L., Saturno, J., Schmid, B., Souza, R. A., Springston, S. R., Tomlinson, J. M., Toto,
- 1072 T., Walter, D., Wimmer, D., Smith, J. N., Kulmala, M., Machado, L. A., Artaxo, P., Andreae, M.
- 1073 O., Petaja, T., and Martin, S. T.: Amazon boundary layer aerosol concentration sustained by 1074 vertical transport during rainfall, Nature, 539, 416-419, 2016.
- 1075 Wang, J., Lee, Y.-N., Daum, P. H., Jayne, J., and Alexander, M.: Effects of aerosol organics on
- 1076 cloud condensation nucleus (CCN) concentration and first indirect aerosol effect, Atmos Chem1077 Phys, 8, 6325-6339, 2008.
- 1078 Webster, C. and Freudinger, L.: Interagency Working Group for Airborne Data and
- 1079 Telecommunications, 2018.
- 1080 Wendisch, M. and Brenguier, J.-L.: Airborne measurements for environmental research: methods
- and instruments, John Wiley & Sons, 2013.
- 1082 Wendisch, M., Coe, H., Baumgardner, D., Brenguier, J.-L., Dreiling, V., Fiebig, M., Formenti,
- 1083 P., Hermann, M., Krämer, M., Levin, Z., Maser, R., Mathieu, E., Nacass, P., Noone, K.,
- 1084 Osborne, S., Schneider, J., Schütz, L., Schwarzenböck, A., Stratmann, F., and Wilson, J. C.:
- 1085 Aircraft Particle Inlets: State-of-the-Art and Future Needs, B Am Meteorol Soc, 85, 89-92, 2004.
- 1086 Wendisch, M., Keil, A., and Korolev, A. V.: FSSP characterization with monodisperse water
- droplets, Journal of Atmospheric and Oceanic Technology, 13, 1152-1165, 1996.
- 1088 Wendisch, M., Müller, D., Schell, D., and Heintzenberg, J.: An airborne spectral albedometer
- 1089 with active horizontal stabilization, J Atmos Ocean Tech, 18, 1856-1866, 2001.
- 1090 Wendisch, M., Poschl, U., Andreae, M. O., Machado, L. A. T., Albrecht, R., Schlager, H.,
- 1091 Rosenfeld, D., Martin, S. T., Abdelmomonem, A., Afchine, A., Araujo, A. C., Artaxo, P.,
- 1092 Aufmhoff, H., Barbosa, H. M. J., Borrmann, S., Braga, R., Buchholz, B., Cecchini, M. A., Costa,
- 1093 A., Curtius, J., Dollner, M., Dorf, M., Dreiling, V., Ebert, V., Ehrlich, A., Ewald, F., Fisch, G.,
- 1094 Fix, A., Frank, F., Futterer, D., Heckl, C., Heidelberg, F., Huneke, T., Jakel, E., Jarvinen, E.,
- 1095 Jurkat, T., Kanter, S., Kastner, U., Kenntner, M., Kesselmeier, J., Klimach, T., Knecht, M., Kohl,
- 1096 R., Kolling, T., Kramer, M., Kruger, M., Krisna, T. C., Lavric, J. V., Longo, K., Mahnke, C.,
- 1097 Manzi, A. O., Mayer, B., Mertes, S., Minikin, A., Molleker, S., Munch, S., Nillius, B.,
- 1098 Pfeilsticker, K., Pohlker, C., Roiger, A., Rose, D., Rosenowow, D., Sauer, D., Schnaiter, M.,
- 1099 Schneider, J., Schulz, C., de Souza, R. A. F., Spanu, A., Stock, P., Vila, D., Voigt, C., Walser,
- 1100 A., Walter, D., Weigel, R., Weinzierl, B., Werner, F., Yamasoe, M. A., Ziereis, H., Zinner, T.,
- and Zoger, M.: ACRIDICON-CHUVA CAMPAIGN Studying Tropical Deep Convective
- 1102 Clouds and Precipitation over Amazonia Using the New German Research Aircraft HALO, B
- 1103 Am Meteorol Soc, 97, 1885-1908, 2016.
- 1104 Zaveri, R. A., Berkowitz, C. M., Brechtel, F. J., Gilles, M. K., Hubbe, J. M., Jayne, J. T.,
- 1105 Kleinman, L. I., Laskin, A., Madronich, S., and Onasch, T. B.: Nighttime chemical evolution of
- aerosol and trace gases in a power plant plume: Implications for secondary organic nitrate and
- 1107 organosulfate aerosol formation, NO3 radical chemistry, and N2O5 heterogeneous hydrolysis,
- 1108 Journal of Geophysical Research: Atmospheres, 115, 2010.
- 1109
- 1110

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

Measurement Variables	Instruments deployed on the G1 (Martin et al., 2016; Schmid et al., 2014)	Instruments deployed on HALO (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 -50 to +50 °C	Total Air Temperature (TAT) inlet (Goodrich/Rosemount type 102) with an 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 particle chemical composition	HR-ToF-AMS: Organics, Sulfate, Nitrate, Ammonium, Chloride, 60-1000 nm	C-ToF-AMS: Organics, Sulfate, Nitrate, Ammonium, Chloride, 60-1000 nm			
CCN concentration	CCN-200, SS= 0.25, 0.5%	CCN-200, SS= 0.13-0.53%			
Gas phase concentration	N2O/CO and Ozone Analyzer, CO, $O_3$ concentration, precision 2 ppb	N2O/CO and Ozone Analyzer, CO, O <sub>3</sub> concentration, precision 2 ppb			
Cloud	CDP, 2-50 $\mu$ m, $\Delta D_{\rm p}=1-2 \mu$ m	CCP-CDP, 2.5-46 mm, $\Delta D_{\rm p}$ =1-2 µm			
properties*	FCDP, 2-50 $\mu$ m, $\Delta D_{\rm p}$ =1-2 $\mu$ m	NIXE-CAS: 0.61 -52.5 µm			
	2DS, 10-1000 µm	NIXE-CIPgs, 15-960 µm			
		CCP-CIPgs: 15-960 µm			
Radiation	SPN1 downward irradiance, 400-2700 nm	SMART Albedometer, downward spectral irradiance, 300-2200 nm			

- 4 \*for an individual flight, the size range may vary.
- 5

6 7

SEP 9, 2014 SEP 21, 2014 HALO G1 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 531 7982 4546 663 G1: CDP(FCDP) N/A 3627(4439) 2051(2260) N/A HALO: CCP-CDP (NIXE-CAS) G1: 2DS N/A N/A 2280 2261 (2260) HALO: CCP-CIPgs (NIXE-CIPgs) N/A RAD 1355 1355 N/A

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

 Table 3. Summary of basic statistics of data between in situ measurements on Sep 9.

 Comparison of the coordinated flight on Sep 9.

Comparison of the coordinated flight 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 <sub>dew</sub> , k	293	296.5	295.0	0.5	292.9	294.9	294.0	0.3	0.996	Neg.
O3, ppb	10.5	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,	78.2	1118.	645.5	116.3	504.1	1622.2	756.3	138.6	1.165	0.8193
<i>cm</i> <sup>-3</sup>										
CCNc (к)	0.010	0.347	0.1855	0.067	0.012	0.394	0.1890	0.083	0.8937	Neg.

Comparison of the coordinated flight on Sep. 21							
	m	offset	$\mathbb{R}^2$	m0	$\mathbb{R}^2$	m1	$\mathbb{R}^2$
Т, К	0.929	20.0	0.9992	0.999	0.9928	0.999	0.9928
P, hPa	1.001	0.929	0.9998	1.001	0.9998	1.001	0.9998
WSpd, m/s	0.885	1.0	0.7875	1.012	0.5076	1.023	0.5049
T <sub>dew</sub> , k	0.989	3.8	0.9963	1.003	0.9904	1.003	0.9904
O <sub>3</sub> , ppb	1.134	-1.5	0.9598	1.075	0.9369	1.101	0.9208
CO, ppb	0.922	5.4	0.9654	0.966	0.9254	0.967	0.9254
$CPC, cm^{-3}$	0.571	199.4	0.9482	0.635	0.8738	0.641	0.8735
UHSAS,	1.126	178.0	0.8249	1.293	0.5070	1.384	0.4847
<i>cm</i> <sup>-3</sup>							
ССЛс (к)	0.766	55.3	0.8330	0.815	0.6544	0.829	0.6521

Table 4 Summary of three statistics analysis of data between in situ measurements on Sep 21





Figure 1. Coordinated flight tracks for September 9 (a) and September 21 (b). The black dotted line is the flight track of the G1, and the blue line is the flight track of HALO. (This figure was

created using Mapping Toolbox<sup>TM</sup> © COPYRIGHT 1997–2019 by The MathWorks, Inc.)





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). (This
figure was created using Mapping Toolbox<sup>™</sup> © COPYRIGHT 1997–2019 by The MathWorks,
Inc.)



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





Figure 5. Aircraft altitude-colored plots of trace gas (a) Ozone, (b) CO, for the coordinated flight
on September 21.







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

45 (>10 nm) on September 9: (a) with iso-kinetic inlet constrain; (b) with different criteria.



47 Figure 7. The G1 and HALO comparison for aerosol number concentration measured by48 UHSAS (90-500 nm) on September 9.



Figure 8. The G1 and HALO comparison for aerosol number concentration profiling measured
by (a) CPC and (b) UHSAS (100-700 nm) on September 21.



Figure 9. The G1 and HALO comparison for aerosol size distribution measured by UHSAS(from both aircraft) and FIMS (on the G1) on September 9.



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

- 59 UHSAS, (c) the HALO UHSAS, (d) Potential temperature aboard the G1 and HALO on September
- 60 21.
- 61



Figure 11. The vertical profile of aerosol mass concentration measured by the G1 and HALOAMS on September 21.



Figure 12. The vertical profile of relative mass fraction of major aerosol chemical speciesmeasured by the G1 and HALO AMS, respectively, on September 21



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







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



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



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 inter-

86 comparison criteria. The blue circles represent the restricted navigation criteria.

83

## Data availability

All ARM datasets used for this study can be downloaded at <u>https://iop.archive.arm.gov/arm-iop/2014/mao/goamazon/</u> (DOI: 10.5439/1346559). The full data set from the ACRIDICON-CHUVA campaign is archived and publicly accessible from the HALO database maintained by the German Aerospace Center (DLR) at <u>https://halo-db.pa.op.dlr.de/mission/5</u>.

## Competing interests

The authors declare that they have no conflict of interest.