Balloon-borne aerosol-cloud interaction studies (BACIS): New

3 observational techniques Field campaigns to understand and quantify aerosol effects

4—__on clouds

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Abstract.

- 34 Better A better understanding of aerosol-cloud interaction processes is an-important aspect to quantify the role of clouds and aerosols
- 35 in on the climate system. There have been significant efforts to explain the ways aerosols modulate cloud properties. However,
- 36—from the observational point of view, it is indeed challenging to observe and/or verify some of these processes because no
- 37—single instrument or platform is proven sufficient. <u>Discrimination between aerosol and cloud is vital for the quantification of aerosol-cloud interaction.</u> With this motivation, a unique—set of observational field campaigns named
- 38—Balloon—borne Aerosol Cloud Interaction Studies (BACIS) is proposed and conducted using balloon—borne in-situ
- 39—_measurements in addition to the ground-based (Lidars, MST radar, LAWP, MWR, Ceilometer) and space—borne (CALIPSO)
- 40—remote sensing instruments from Gadanki (13.45°N, 79.2°E).), India. So far, 15 campaigns have been conducted as a part of BACIS
- 41—campaigns from 2017 to 2020. This paper presents the concept of the observational approach, lists the major objectives of the
- 42—_campaigns, describes the instruments deployed, and discusses results from selected campaigns. Consistency in balloon_Balloon_borne
- 43— measurements is are assessed using the data from simultaneous observations of ground-based, space—borne remote sensing
- 44— instruments. A good agreement is found among Aerosol/cloud profiles obtained from the multi-instrumental observations. Balloon

- are found similar. Apart from this, balloon-borne in situ-profiling is found to
- 45 <u>complement the provides</u> information <u>provided missed</u> by ground-based and/or space—borne measurements. A combination of the Compact
- 46—Optical Backscatter AerosoL Detector (COBALD) and Cloud Particle Sensor (CPS) sonde is employed for the first time to
- 47—_discriminate cloud and aerosol in an in-situ profile. A threshold value of COBALD <u>color_colour</u> index (CI) for ice clouds is found to
- 48—be between 18 and 20 and CI values for coarse mode aerosol particle particles range between 11 and 15. Using the data from balloon
- __measurements, the relationship between cloud and aerosol is quantified for the liquid clouds. A statistically significant slope <u>(aerosol-cloud interaction index)</u> of 0.77 found between aerosol back scatter and cloud particle count reveals the role of aerosol in the cloud activation process. In a nutshell, the results presented here demonstrate the observational approach to quantifying aerosol-cloud interactions.
 - 50 (aerosol cloud interaction index) of 0.77 (0.86) found between aerosol back scatter from 300m (400m) below the cloud base
 - 51 and cloud particle count within the cloud indicates the role of aerosol in the cloud activation process. In a nutshell, the results
 - 52 presented here demonstrate the observational approach to quantify acrosol-cloud interactions and paves the way for further
 - 53 investigations using the approach.

1 Introduction

Understanding the fundamental process of aerosol cloud interactions remains to be a challenging issue in the seientific community, already for more than three decades (Scinfeld et al., 2016). First ever observational evidence from analysis of ship tracks using satellite imagery had open up a wide scope for further research in this area (Coakley et al., 60—1987; Radke et al., 1989). Since then, efforts are underway using different observational and modeling techniques and lead to a significant development in the process based understanding, quantification and modeling (Abbott and Cronin, 2021; Fan

et al., 2018; Haywood and Boucher, 2000; Koren et al., 2010; Lohmann, 2006; Lohmann and Feichter, 2004; Rosenfeld et al., 2008, 2014b). Despite of all these efforts, radiative forcing estimates due to aerosol cloud interactions still show large uncertainties (IPCC, 2013). Apart from this, climate model simulations have uncertainties due to the fact that parameterization schemes are inefficient in representing the ways aerosols interact with clouds (Fan et al., 2016; Rosenfeld et al., 2014b; Seinfeld et al., 2016). At process level, various hypothesis have been proposed subsequent to the first indirect effect which was proposed almost four decades ago (Twomey, 1977). All these effects are found to act specific to cloud -based on background meteorological, dynamical conditions. For example, the invigoration effect is proposed for convective -clouds (Rosenfeld et al., 2014a) under the influence of updrafts. First indirect effect (Twomey effect) and second indirect effect (Albrecht effect) for liquid clouds have been shown to be influenced by mixing (Costantino and Bréon, 2010), turbulence and entrainment (Jose et al., 2020; Schmidt et al., 2015; Small et al., 2009). Although the first indirect effect is reasonably well understood, observational limitation poses serious challenges in understanding and/or evaluating other 71 hypotheses. Among the various observational techniques that are currently available (ground based, space borne remote sensing and aircraft or unmanned ariel vehicle; UAV), none of the single observational technique has been proven self sufficient in aerosol cloud interaction studies. For example, ground based (and/or space borne) lidars suffer serious attenuation and even losses of observations due to the presence of optically thick cloud layers in the atmosphere. Thus, they may not be able to represent the complete vertical structure of cloud and aerosols. Note that information on aerosol/cloud profile is essential for the estimation of their climate effects. Similarly, satellite data analysed with different analytical methods such as by changing grid resolutions have shown different results and conclusions (Grosvenor et al., 2018; Koren et al., 2010; McComiskey and Feingold, 2012). Besides this, in situ measurements using aircraft and UAV have been remarkable in obtaining detailed information on the microphysics of cloud and aerosol (Corrigan et al., 2008; Kulkarni et al., 2012; Redemann et al., 2020; Weinzierl et al., 2017). However, there are serious limitations with respect to altitude coverage, feasibility of conducting aircraft or UAV campaigns and overall cost involved. Also, there is a chance that the aircraft

-perturb the atmosphere before it actually makes the measurement of cloud/aerosol.

Therefore, it is essential to examine the combined information obtained simultaneously using multi-instrumental techniques so as to obtain a comprehensive picture. A classic paper by Feingold et al. (2003) first time quantified the 'Twomey effect' using ground based remote sensing instruments such as a micro wave radiometer (MWR), cloud radar and a Raman Lidar. In an intensive operations program, Feingold et al. (2006) conducted airborne in situ measurements for obtaining the cloud effective radius using an aircraft in addition to the ground based and space borne remote sensing instruments. Pandithurai et al. (2009) also quantified the 'Twomey effect' using a suite of ground based remote sensing instruments (cloud radar, MWR, polarization Lidar) along with the surface aerosol measurements (aerosol size distribution, scattering coefficient and cloud condensation nuclei concentration). Similarly, Sena et al. (2016) utilized 14 years of coincident observations from cloud radar and a laser Ceilometer along with surface reaching shortwave radiation measurements from the Atmospheric Radiation Measurement (ARM) program over the Southern Great Plains, USA to investigate acrosol modifications on cloud macroscopic parameters and radiative properties rather than cloud microphysical parameters. In addition to simultaneous measurements of cloud/aerosol, concurrent measurements of thermodynamic and dynamic parameters of the atmosphere are also needed to thoroughly understand the process of aerosol cloud interactions. A step forward in this direction, McComiskey et al. (2009) used long term, statistically robust ground based remote sensing data from Pt. Reyes, California, USA to not only quantify the 'Twomey effect' but also examine the factors influencing the variability in aerosol indirect effects such as updraft velocity, liquid water path, scale and resolution of observations. Using a novel dual field of view Raman Lidar and a Doppler Lidar technique, Schmidt et al. (2014) analysed the data from Leipzig, Germany to explore linkages between aerosol and cloud properties, the influence of updrafts. Sarna and Russchenberg, (2016) used synergy of measurements from a Lidar (Ceilometer), Radar (cloud radar) and a Radiometer (MWR) collected at ARM Mobile facility at Graciosa Island, the Azores, Portugal and at the Cabaw Experimental Site for Atmospheric Research (CESAR) observatory, The Netherlands, to not only quantify the aerosol indirect effect but also attempted to disentangle the effect of vertical wind (Sarna and Russchenberg, 2017). All these studies contributed significantly to the knowledge on aerosol cloud interactions but are based on remote sensing techniques, limited to the low level, warm and non precipitating clouds only.

In view of Understanding the fundamental process of aerosol-cloud interactions remains to be a challenging issue in the scientific community, already for more than three decades (Seinfeld et al., 2016). First-ever observational evidence from analysis of ship tracks using satellite imagery had opened up a wide scope for further research in this area (Coakley et al., 1987; Radke et al., 1989). Since then, efforts are underway using different observational and modelling techniques and lead to a significant development in the process-based understanding, quantification, and modelling (Abbott and Cronin, 2021; Fan et al., 2018; Haywood and Boucher, 2000; Koren et al., 2010; Lohmann, 2006; Lohmann and Feichter, 2004; Rosenfeld et al., 2008, 2014b). Despite all these efforts, radiative forcing estimates due to aerosol-cloud interactions still show large uncertainties (IPCC, 2021). Apart from this, climate model simulations have uncertainties because parameterization schemes are inefficient in representing the ways aerosols interact with clouds (Fan et al., 2016; Rosenfeld et al., 2014b; Seinfeld et al., 2016). At the process level, various hypotheses have been proposed after the first indirect effect which was proposed almost four decades ago (Twomey, 1977). All aerosol-cloud effects are found to act specifically to cloud type, background meteorological, and dynamical conditions. For example, the invigoration effect is proposed for convective clouds (Rosenfeld et al., 2014a) under the influence of updrafts. The first indirect effect (Twomey effect) and the second indirect effect (Albrecht effect) for liquid clouds be influenced by mixing (Costantino and Bréon, 2010), turbulence, and entrainment (Jose et al., 2020; Schmidt et al., 2015; Small et al., 2009). Although the first indirect effect is reasonably well understood, observational limitation poses serious challenges in understanding and/or evaluating other hypotheses.

Among the various observational techniques that are currently available (ground-based, space-borne remote sensing, and aircraft or unmanned aerial vehicle; UAV), none of the techniques alone has been proven self-sufficient in aerosol-cloud interaction studies. For example, ground-based (and/or space-borne) lidars suffer serious attenuation and even losses of observations due to the presence of optically thick cloud layers in the atmosphere. Thus, they may not be able to represent the complete vertical structure of clouds and aerosols. Note that information on aerosol/cloud profiles is essential for the estimation of their climate effects. Similarly, satellite data sets have shown distinct results and conclusions (Grosvenor et al., 2018; Koren et al., 2010; McComiskey and Feingold, 2012) using different analytical methods for example changing grid resolutions, etc. Besides this, in-situ measurements using aircraft and UAVs have been remarkable in obtaining detailed information on the microphysics of cloud and aerosol (Corrigan et al., 2008; Girdwood et al., 2020, 2021; Kulkarni et al., 2012; Mamali et al., 2018; Redemann et al., 2020; Weinzierl et al., 2017). However, there are serious limitations concerning altitude coverage, the feasibility of conducting aircraft or UAV campaigns, and the overall cost involved. Also, there is a chance that the aircraft perturb the atmosphere before it measures cloud/aerosol.

Therefore, it is essential to examine the combined information obtained simultaneously using multi-instrumental techniques to obtain aerosol, cloud and associated environmental parameters to understand aerosol-cloud interaction. A classic paper by Feingold et al. (2003) first time quantified the 'Twomey effect' using ground-based remote sensing instruments such as a microwave radiometer (MWR), cloud radar, and a Raman Lidar. In an intensive operations program, Feingold et al. (2006) conducted airborne in-situ measurements for obtaining the cloud effective radius using an aircraft in addition to the ground-based and space-borne remote sensing instruments. Pandithurai et al. (2009) also quantified the 'Twomey effect' using a suite of ground-based remote sensing instruments (cloud radar, MWR, polarization Lidar) along with the surface aerosol measurements (aerosol size distribution, scattering coefficient, and cloud condensation nuclei concentration). Similarly, Sena et al. (2016) utilized 14 years of coincident observations from cloud radar and a laser Ceilometer along with surface-reaching shortwave radiation measurements from the Atmospheric Radiation Measurement (ARM) program over the Southern Great Plains, USA to investigate acrosol modifications on cloud macroscopic parameters and radiative properties rather than cloud microphysical parameters. In addition to simultaneous measurements of cloud/aerosol, concurrent measurements of thermodynamic and dynamic parameters of the atmosphere are also needed to thoroughly understand the process of aerosol-cloud interactions. A step forward in this direction, McComiskey et al. (2009) used long-term, statistically robust ground-based remote sensing data from Pt. Reves, California, the USA to not only quantify the 'Twomey effect' but also examine the factors influencing the variability in aerosol indirect effects such as updraft velocity, liquid water path, scale, and resolution of observations. Using a novel dual field of view Raman Lidar and a Doppler Lidar technique, Schmidt et al. (2014) analyzed the data from Leipzig, Germany to explore linkages between aerosol, cloud properties, and the influence of updrafts. Sarna and Russchenberg, (2016) used synergy of measurements from a Lidar (Ceilometer), Radar (cloud radar) and a Radiometer (MWR) collected at ARM Mobile facility at Graciosa Island, the Azores, Portugal, and the Cabaw Experimental Site for Atmospheric Research (CESAR) observatory, The Netherlands, to not only quantify the aerosol indirect effect but also attempted to disentangle the effect of vertical wind (Sarna and Russchenberg, 2017). All these studies contributed significantly to the knowledge on aerosol-cloud interactions but are based on remote sensing techniques, limited to the low-level, warm, and non-precipitating clouds only.

- Given the measurement limitations discussed above, a balloon--borne in-situ measurement is suggested to be the
- 89 best complimentary complementary technique as balloons can pass through the cloud (during their ascent/descent) representing the vertical
- 91— structure of the cloud as well as aerosol below and above the cloud near simultaneously (see Sect. 2 for details) without
- 93 __perturbing the atmosphere. <u>Information Although there is less information and data on balloon-based aerosol sampling artefacts</u> than on conventional aircraft, information from balloon—borne in-situ measurements in combination with the ground-based

and/or space—borne platforms will be of great help in constructing the complete vertical profiles of aerosol, cloud—and further, and further understanding the process of aerosol-cloud interactions. With this in mind, a balloon-borne field campaign named BACIS (Balloon-borne Aerosol Cloud Interaction Studies) was initiated in the year 2017 at National Atmospheric Research Laboratory (NARL), Gadanki (13.45° N, 79.2° E), India, with the multi-instrumental approach.

understanding the process of aerosol cloud interactions. With this in mind, a balloon borne field campaign named BACIS

(Balloon borne Aerosol Cloud Interaction Studies) was initiated in the year 2017 from National Atmospheric Research

Laboratory (NARL), Gadanki (13.45°N, 79.2°E), India, with multi instrumental approach. Gadanki located in Southern

Peninsular India is influenced by both the South West and North East Monsoon. Most importantly the location hosts a suit of

unique ground based instruments that cover optical, radio and in situ techniques. A combination of specialized sondes like

the Compact Optical Backscatter Aerosol Detector (COBALD; see https://iac.ethz.ch/group/atmospheric
chemistry/research/ballon-soundings.html, last access: 19 June 2021) and the Cloud Particle Sensor (CPS; Fujiwara et al.,

The concept of observational strategy, objectives of the campaign and details about the balloon sensors,

ground/space based instruments, data processing, interpretation of data are provided in the second section. The third section

discusses the results on consistency in multi-instrumental observations, interpretation of aerosol and cloud features in a

profile, multiple soundings to estimates statistics on threshold values of aerosol/cloud and finally illustrates the relationship

between aerosol-cloud relationships. The fourth section summarizes the results.

Balloon-borne measurement of aerosol/cloud was first reported in Rosen and Kjome, 1991 using a backscatter sonde developed by them. COBALD is similar to this but lightweight sonde (Brabec et al., 2012). Measurements of aerosol size distribution in the stratosphere were carried out using an optical particle counter developed at Wyoming university (Deshler et al., 2003). But Smith et al., 2019 developed a novel, low-cost, and lightweight open path configuration optical particle counter, UCASS (Universal Cloud Aerosol Sampling System) for a wide range of particle size measurements covering both aerosol and cloud. Kezoudi et al., 2021 and Mamali et al., 2018 used UCASS and reported balloon-borne in-situ measurement of dust aerosol and compared UCASS with ground-based, airborne instruments. However, BACIS campaigns are designed to understand and quantify aerosol-cloud interactions. For this, a combination of balloon-borne sondes, COBALD and CPS is used for the first time to separate/discriminate aerosol and cloud in a profile. Note that individually COBALD and CPS have been used in other studies (Brunamonti et al., 2018, 2020; Fujiwara et al., 2016a; Hanumanthu et al., 2020; Inoue et al., 2021; Vernier et al., 2015, 2018).

The purpose of this manuscript is to introduce the motivation and objectives of the BACIS Campaigns for quantifying aerosol-cloud interactions. In order to do this, we have discussed most related topics, such as the campaign approach, sensors/instruments employed, analytical methods and comparison of balloon features. Results from selected campaigns focus on discrimination of aerosol/cloud in a profile.

Overall, the methods presented in this paper for the data analysis/processing are novel. Using these methods aerosol-cloud interaction is estimated in liquid clouds.

- 2. Instruments and methodologymethods
- 2.1. Balloon-borne sensors
- **2.1.1. COBALD**
- The Compact Optical Backscatter AerosoL Detector (COBALD) deployed in BACIS campaigns is a lightweight
- 107 (540 g) balloon-borne sonde developed in the group of Professor Thomas Peter at ETH Zurich, Switzerland. It is essentially a
- _miniaturized version of the backscatter sonde developed by Rosen and Kjome (1991). A backscatter sonde is a balloon-borne sensor which measures the backscattered light from molecules, aerosol and clouds at multiple wavelengths in the vicinity of the sonde as it passes through the atmospheric column. The COBALD consists of two LED light
- 111— sources of approximately 500 mW power emitting 455 nm (blue) and 940 nm (termed 'infrared') wavelengths, respectively

- 112 (Brabec et al., 2012). (Brabec et al., 2012). The light emitted by the sonde illuminates the air in the vicinity, and backscattered light from an ensemble of particles is detected
- <u>114</u> using a silicon <u>photo detector. photodetector.</u> The emitted <u>beams beam's</u> divergence (<u>with a full-width half-maximum of 4</u> degrees <u>FWHM</u>), detector field of view (<u>of 6</u> degrees), and
- 116—geometrical alignment of optics yields the reception of backscatter light from a distance of 0.5 m (overlapping distance) from the
- 118—sonde. The region of up to 10 m from the instrument contributes to 90 % of the measured backscattering signal. The real-
- 120—time backscatter data, in units of counts per secondssecond (cps, originating from the internal data treatment) is included in the
- 122 radiosonde telemetry at a frequency of 1 Hz and sent to the ground station along with the pressure and temperature
- 124—measurements. In the present case, we have used an iMet radiosonde (InterMet, USA). The sondes were usually operated for
- 126—about 15 minutes at the surface (before launch) for thermal stabilization, verified by cross-checking the LED brightness
- <u>nonitor signals, and</u> also delivered in <u>epscounts per second</u>, with sonde specific reference values provided by the <u>manufacture</u>manufacturer. The sonde is <u>passed</u>
- $\frac{130}{100}$ launched when the return signal data at the surface is within $\pm 15\%$ of the reference value.

143—2.1.2. CPS

- Cloud Particle Sensor (CPS) sonde is a <u>light-weight_lightweight</u> balloon_borne sensor (~200 g) developed for the detection of
- cloud particle number and phase (Fujiwara et al., 2016). cloud particle number and phase (Fujiwara et al., 2016b). The latest version of the sonde (launched in the campaigns) is
- 149—supplied by Meisei Electric Corporation, Japan, along with a Meisei RS-11G radiosonde (Kobayashi et al., 2019; RS-
- 151 (Kobayashi et al., 2019; RS-11G(R3) is the model with an interface for CPS). CPS primarily consists of a column (~1 cm x 1 cm in cross-section and
- 153—_~12 cm in vertical length) for air passage, a diode laser (~790 nm, polarized)), and two silicon photo detectors.photodetectors. Cloud particles
- 155—entering the column due to the balloon ascent are illuminated by the laser. The scattered light from cloud particles is detected
- 157 _by the photo-detectors placed at an angle of 55° and 125° to the incident laser light. The detector at 125° comes with an
- 459 additional polarization plate positioned in front of it for the detection of cross-polarization whereas the detector at 55°
- 161 measures the intensity of plane-polarized scattered light. The intensities I55 and I125, for the detectors located at 55° and
- 163—125°, respectively, are provided in voltage, and I55 is related to particle size. The minimum size of a water droplet that can

- 165 __be detected by CPS is found to be 2 μm (1 μm particles are undetected in laboratory experiments using various standard
- 167 _spherical particles) and I55 was found to sometimes saturate (~7.5V) for particles ~80-140 μm (Appendix A of Fujiwara et
- 169 __al., 2016). Real_time data from CPS ishas been transferred to the ground station through RS-11G_(R3) radiosonde at a frequency of
- 171 __1Hz. CPS data include the number of particles counted in a sec, scatted light intensity (in Voltage) for the two detectors (I55 and

172—I125), as well as particle signal width for the first six particles for each second, and DC output voltage. The particle

174—information is transmitted to the ground station only for the first six particles for each second due to the limited downlink

176 rate of RS-11G which is 25 byte s⁻¹. Before launch, the sonde is tested by spraying water near to air passage column for particle detection.

178 particle detection.

2.2. Remote sensing instruments

----2.2.1. MPL/Ceilometer

- A Micro Pulse Lidar (MPL) was operated on 07-08 July 2017 during the first two campaigns. However, a laser
- 167 Ceilometer (make of Vaisala, Finland) was made available for the rest of the campaigns due to non availability of MPL.
- Complete technical details of MPL used in the campaign can be found in Cherian et al. (2014). Cherian et al. (2014). A low energy $(< 10 \ \mu j)$ green
- 171 (532 nm) pulsed laser of pulse width less than 10 ns werewas shot from MPL at a pulse repetition frequency of 2500 s⁻¹. A
- 173—Cassegrain type telescope of 150 mm diameter and a PMT have been deployed to collect the backscattered photons (co-
- 175—polarized) from particles and clouds in the atmosphere. The entire system is operated at a dwell time of 200 ns which would
- 177 __correspond to a range resolution of 30 m. The return signals were collected for 1500 bins which correspond to the total range
- 179 of 45 km. A profile of backscattered photons was obtained for every 300 μs and all profiles collected were averaged for
- 181 __every one minute. The telescope field of view and laser beam divergence coincides or overlap at above ~150 m. Using the
- data from MPL (from Gadanki and the nearby location at Sri Venkateswara University, Tirupati, India (13.62°N, 79.41°E;
- 185 ~35 km from Gadanki), Ratnam et al. (2018) ~35 km from Gadanki), Ratnam et al. (2018) reported the presence of an elevated aerosol layer in the lower troposphere (~3
- 487—km) during South-West Monsoon Season and discussed the possible causes for the formation and maintenance of this
- 189 __elevated layer. The low-_level jet (LLJ) between 2 and 3 km in the lower troposphere present during South Westthe southwest Monsoon
- $\underline{\textbf{191}}\underline{\textbf{c}}\text{auses}\ \underline{\textbf{the}}\ \text{formation of}\ \underline{\textbf{an}}\ \text{elevated layer. In addition, the presence of shear between LLJ and tropical easterly jet (TEJ) maintains}$
- the elevated layer restricting the upliftment of aerosol. Prasad et al. (2019) Prasad et al. (2019) also used the same dataset

td discuss nocturnal, seasonal, and intra-annual variations in the tropospheric aerosol.

- 194 seasonal and intra annual variation in the tropospheric aerosol. A laser Ceilometer (operated in rest of the campaigns) is
- 195 similar to a MPL but operates at 910 nm wavelength and provides round the clock measurements of cloud base heights,
- 196 boundary layer height apart from aerosol extinction under all weather conditions (Wiegner et al., 2014).

A Ceilometer (make from Vaisala, Finland) was used in the rest of the campaigns during non-available dates of MPL. It is similar to

an MPL but operates at a 910 nm wavelength and provides round-the-clock measurements of cloud base heights, and boundary layer height

apart from aerosol extinction under all weather conditions (Wiegner et al., 2014).

2.2.2. Mie Lidar

Mie lidar at Gadanki is a unique lidar system with capabilities to probe the atmosphere to higher altitudes (-30 km).

199 This lidar was operated in almost all the campaigns. A very high energy (600 mJ) pulsed laser of pulse width of less than 7

199 ns and a pulse repetition frequency of 50 s⁺ is operated at a wavelength of 532 nm. A 320 mm diameter Cassegrain type

200 telescope along with a couple of PMT has been used as a detection assembly to collect the co and cross polarized return

200 signal. However, the co-polarization channel (only) is analysed in the present study. The data is stored at a dwell time of 2 μs

200 which corresponds to the range resolution of 300 m and the profiles collected were averaged for every 250 sec (- 4 min).

200 The data is considered to be reliable from an altitude of 3 4 km as the field of view of Mie telescope and laser beam

200 divergence overlap at this height (Pandit et al., 2014). For the first time, sixteen years of Mie lidar data has been analysed to

200 determine the long term climatology of tropical cirrus clouds (Pandit et al., 2015). Gupta et al. (2021) reported the long term

200 observations of aerosol extinction profiles using combination of MPL, Mie lidar and space borne CALIPSO lidar.

Mie lidar at Gadanki is a unique lidar system with capabilities to probe the atmosphere to higher altitudes (~30 km). This lidar was operated in almost all the campaigns. A very high energy (600 mJ) pulsed laser with a pulse width of a few 7 ns and a pulse repetition frequency of 50 s⁻¹ is operated at a wavelength of 532 nm. A 320 mm diameter Cassegrain type telescope along with a couple of PMT has been used as a detection assembly to collect the co and cross-polarized return signal. However, the co-polarization channel (only) is analysed in the present study. The data is stored at a dwell time of 2 μs which corresponds to the range resolution of 300 m and the profiles collected were averaged every 250 sec (~ 4 min). The data is considered to be reliable from an altitude of 3-4 km as the field of view of the Mie telescope and laser beam divergence overlap at this height (Pandit et al., 2014). For the first time, sixteen years of Mie lidar data have been analysed to determine the long-term climatology of tropical cirrus clouds (Pandit et al., 2015). Gupta et al. (2021) reported the long-term observations of aerosol extinction profiles using a combination of MPL, Mie lidar, and a space-borne CALIPSO lidar.

2.2.3. CALIPSO

- 203 ——Cloud_Aerosol Lidar with Orthogonal Polarization (CALIOP) is the space_born lidar on board onboard the CALIPSO satellite
- 205 (L'Ecuyer, 2011). (L'Ecuyer, 2011). CALIOP consists of two pulsed diode lasers operating at 532 and 1064 nm wavelengths with pulse energy
- 207—of 110 mJ and a repetition rate of ~ 20 Hz. <u>A Backscattered signal is collected by an avalanche photodiode (APD)</u> at 1064 nm
- 209 __and photo multiplier tubes (PMT) at 532 nm. The signals at 532 nm are collected at both parallel and perpendicular to the

- 211—plane of polarization of the outgoing beam, while for 1064 nm channel polarization is parallel only. The range resolution of the 213—backscattered profile at 532 nm is 30 m for the altitude range from -0.5 to 8.2 km, 60 m for 8.2 to 20.2 km and 180 m for 215—20-30 km. Horizontal resolution is 0.33 km for -0.5 to 8.2 km and 1 km for 8.5-20.2 km. More details about CALIOP can be found in Winker et al. (2007).
 - 217 be found in Winker et al. (2007).

2.2.4. MST Radar

- 221 The Indian MST radar located at Gadanki is high power coherent backscatter VHF (Very High Frequency) radar
- 223 operating at 53MHz. The detailed description of MST radar can be found in Rao et al. (1995). Before the BACIS campaign,
- 225 it has been upgraded to a fully active phased array with dedicated 1 kW solid state transmitter receiver units (total power of
- The Indian MST radar located at Gadanki is a high-power coherent backscatter VHF (Very High Frequency) radar operating at 53MHz. A detailed description of MST radar can be found in Rao et al. (1995). Before the BACIS campaign, it has been upgraded to a fully active phased array with dedicated 1 kW solid-state transmitter-receiver units (total power of 1024 kW). This radar operates in Doppler Beam Swinging (DBS) mode to provide wind information covering the
- 229—troposphere, lower stratosphere and mesosphere. Atmospheric scatterers are advected with the background air motions and

230—the three-dimensional wind velocity vectors (zonal, meridional and vertical) can be directly deduced from the Doppler shifts

232—of the radar echoes received in three independent beam directions. Note that these radars are the only means of getting direct

234—vertical velocities presently and playsplay a crucial role in the understanding of aerosol-cloud interaction processes. For the

236 present study, data is obtained from five beam directions with 256 FFT (Fast Fourier Transform) points and coherent integrations, 4 incoherent integrations, Inter Pulse Period (IPP) of 160 ms, the pulse width of 8 μs coded covering the altitude region of 3 to 21 km with 150 m vertical resolution.

integrations, 4 incoherent integrations, Inter Pulse Period (IPP) of 160 ms, pulse width of 8 µs coded covering the altitude region of 3 to 21 km with 150 m vertical resolution.

2.3. The observational concept of the BACIS Campaign

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A unique observational approach is proposed here wherein a balloon borne in situ measurement is made simultaneously while the multiple remote sensing instruments are operational from the ground and space borne platforms. The schematic diagram shown in Figure 1 illustrates the entire concept. A meteorological balloon with specialized sondes such as COBALD (Brabec et al., 2012) and CPS (Fujiwara et al., 2016) along with a radiosonde is launched ~10.30 minutes -prior to CALIOP on board Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO; Winker et al., 2007) (night time) overpass close by Gadanki. Ground based remote sensing instruments at NARL, Gadanki such as a Micro Pulse Lidar (MPL; Cherian et al., 2014) and/or a laser Ceilometer (Wiegner et al., 2014), a Mie Lidar (subsequently referenced to as 'Mie'; Pandit et al., 2014), an Indian MST Radar (Rao et al., 1995) and/or a Lower Atmospheric Wind Profiler (LAWP; Srinivasulu et al., 2012) are also operated before, during and after the launch. Other observational facilities -such as ambient aerosol instruments at the Indian Climate Observatory Network (ICON), NARL, Gadanki and a MWR are operated during the launch period. Table 1 lists the ensemble of instruments used in the campaign, their purpose and the -physical quantity that can be obtained from each instrument. Temporal variation of remote sensing data on cloud and aerosol profiles is obtained from ground based (MPL/Mie) lidars. Space borne lidar (CALIPSO) also provides the same but for an along track (roughly meridional) distribution near the time of overpass over Gadanki. On the other hand, in situ 245 measurements of aerosol and cloud profiles along with background meteorological parameters (temperature, relative 245 humidity, wind speed and direction) are collected using the specialized balloon sounding (COBALD and CPS). Combined data from balloon, ground/space borne lidars are the basis for the identification of aerosol and cloud particles. Apart from

247 this, temporal variation in wind components obtained from the ground based radars (MST Radar and/or LAWP) aids in

entangling the effect of vertical winds and turbulence on aerosol cloud interactions. A MWR provides the cloud liquid water

248 and relative humidity profiles, etc., useful to constrain the cloud water content in a cloud layer to understand the aerosol

248 influence on cloud properties. In addition to these measurements, surface aerosol information obtained by the

249 instrumentation available at the ICON observatory, NARL helps in understanding the role of sources of aerosol from the

249 surface. Altogether, near simultaneous information on the aerosol, cloud and background meteorological conditions obtained

249 from the multi-instruments is aimed to understand the aerosol cloud interactions.

An observational approach is conceptualized here wherein a balloon-borne in-situ measurement is made simultaneously while the multiple remote sensing instruments are operated from the ground and spaceborne platforms. The schematic diagram shown in Figure 1 illustrates the observational approach. A meteorological balloon with specialized sondes such as COBALD (Brabec et al., 2012) and CPS (Fujiwara et al., 2016b) along with a radiosonde is launched ~10-30 minutes before CALIOP onboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO; Winker et al., 2007) (night time) overpass close by Gadanki. Ground-based remote sensing instruments at NARL, Gadanki such as a Micro Pulse Lidar (MPL; Cherian et al., 2014) and/or a laser Ceilometer (Wiegner et al., 2014), a Mie Lidar (subsequently referenced to as 'Mie'; Pandit et al., 2014), an Indian MST Radar (Rao et al., 1995) and/or a Lower Atmospheric Wind Profiler (LAWP; Srinivasulu et al., 2012) are also operated before, during and after the launch. Other observational facilities such as ambient aerosol instruments at the Indian Climate Observatory Network (ICON), NARL, Gadanki and an MWR are operated during the launch period. Table 1 lists the ensemble of instruments used in the campaign, their purpose and the physical quantity that can be obtained from each instrument. Temporal variation of remote sensing data on the cloud and aerosol profiles is obtained from ground-based (MPL/Mie) lidars. Spaceborne lidar (CALIPSO) also provides the same but for an along-track (roughly meridional) distribution near the time of overpass over Gadanki. On the other hand, in-situ measurements of aerosol and cloud profiles along with background meteorological parameters (temperature, relative humidity, wind speed and direction) are collected using the specialized balloon sounding (COBALD and CPS). Combined data from balloon and ground/spaceborne lidar is the basis for the identification of aerosol and cloud particles. Apart from this, temporal variation in wind components obtained from the ground-based radars (MST Radar and/or LAWP) aids in entangling the effect of vertical winds and turbulence on aerosol-cloud interactions. An MWR provides the cloud liquid water and relative humidity profiles, etc., useful to constrain the cloud water content in a cloud layer to understand the aerosol influence on cloud properties. In addition to these measurements, surface aerosol information obtained by the instrumentation available at the ICON observatory, NARL helps in understanding the role of sources of aerosol from the surface. Altogether, near-simultaneous information on the aerosol, cloud and background meteorological conditions obtained from the multi-instruments is aimed to understand the aerosol-cloud interactions.

Initially, when the experiment was being conceptualized, it was thought to conduct a launch once in one or two

252—_months. However, due to the limited number of stock of specialized sondes (available with us), it was decided to conduct

254—_instead two pilot campaigns to demonstrate the concept proposed. Apart from this, it was also required to have

256—_balloon/payload tracking equipment to ensure the safe recovery of the payloads. A low-cost GPS/GSM—based tracker was

258—_made available is used for this purpose. Subsequently, two pilot campaigns were conducted in the early hours of 6 June and 8 July

260—_2017. Table 2 lists the date and time of all balloon campaigns that have been conducted from Gadanki as a part of BACIS

262—_campaigns and the instruments operated during the corresponding campaign. As shown in Table 2, so far 15 launches have

been conducted from the year 2017 to 2020.

264 been conducted from the year 2017 to 2020.

- Figure 2 shows the photographs taken at the balloon facility, NARL just before the launch during one of the

 __campaigns. The balloon payload with specialized sondes (COBALD, CPS) and radiosonde (iMet and RS-11G) is shown in

 __Fig. 2(a) and the prelaunch activities at the field are shown in Fig. 2(b). Skilled personnel were deployed for the launch and

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- 286—tracker assembly, among others. Because of these reasons, campaigns were conducted on random dates. However, as it-can

287 be-seen from thein Table 2, we have managed to operate all the essential instruments proposed in the observational approach during other campaigns. In particular, the campaigns in the year 2019 were conducted once a month (March to June 2019) or for two months (July to December 2019). during other campaigns. In particular, the campaigns in the year 2019 were conducted once in a month (March to June, 2019) or two months (July to December, 2019). With the observational approach described above, the following scientific issues/objectives are being pursued/realized: i. Demonstration of the potential of the multi-instrumental observational approach in obtaining the information on the aerosol cloud, and associated environmental parameters, such as 3D-winds, relative humidity, and temperature near-simultaneously. ii. Show consistency Comparison of balloon—borne in-situ measurements among the combination of space bornespaceborne and/or ground-based instruments. — iii. Discrimination of aerosol and cloud in a balloon sounding using the combined observations of COBALD and CPS sondes. This is a prerequisite to use balloon information for aerosol cloud studies. 307 iv. Testing Verifying and quantification of aerosol-cloud interactions and understanding the influence of meteorological and dynamical parameters. 311 v. Find out the differences, if any, in the estimates of the magnitude of aerosol-cloud interaction using multi-instruments _and discuss the possible reasons for the observed discrepancies. 15√i. vi. Understanding of how do thesethe indirect effects of aerosols change radiative transfer through the atmosphere. 17y i. vii. Assessment of Weather and Climate model simulations using the multi-sensor data. 318 2.4. Methods of data processing and interpretation 319 2.4.1. COBALD data processing -Backscattered light received -by COBALD is contributed fromby molecules, aerosols and cloud -particles in the

321 __atmosphere. The molecular Rayleigh contribution to the raw signal (cps) is established during the post-processing of the data

322 __using the simultaneous temperature and pressure recordings of the radiosonde. It serves to normalize the total signal in terms

of backscattering ratio (BSR) according to

323 of backscattering ratio (BSR) according to

 $\frac{324}{\beta_{\text{molecular}}}$ $\frac{BSR = -\beta_{\text{total}}}{\beta_{\text{molecular}}}$ (1)

325 where β_{total} and $\beta_{\text{molecular}}$ are the backscatter coefficient corresponds to the contribution from particles plus molecules and

molecules, respectively. The sole particle contribution is obtained by BSR 1, which expresses the ratio of particle backscatter coefficient to the molecular one. The uncertainty in the COBALD BSR is estimated to be 1% and 5% at surface level and 10 km, respectively (Brabec et al., 2012; Vernier et al., 2015). The Color Index (CI), referring to the particle backscatter only, is calculated from Equation 2.

$$\frac{GI = \frac{BSR_{940} - 1}{BSR_{455} - 1}}{BSR_{455} - 1} \tag{2}$$

$$BSR = \frac{\beta_{\text{total}}}{\beta_{\text{molecular}}} \tag{1}$$

Where β_{total} and $\beta_{\text{molecular}}$ are the backscatter coefficients corresponding to the contribution from particles plus molecules and Molecules, respectively. The sole particle contribution is obtained by BSR-1, which expresses the ratio of particle backscatter coefficient to the molecular one. The uncertainty in the COBALD BSR is estimated to be 1% and 5% at the surface level and 10 km, respectively (Brabec et al., 2012; Vernier et al., 2015). The Color Index (CI), referring to the particle backscatter only, is calculated from Equation 2.

$$CI = \frac{BSR_{940} - 1}{BSR_{455} - 1} \tag{2}$$

- 331—By definition, CI is an independent quantity of particle number concentration and <u>is</u> hence useful in interpreting the size of a
- <u>332</u> particle. For analysis, COBALD raw data is binned in tointo 1 hPa pressure levels. This could minimize noise, and unwanted data
- _and smootheningsmoothen the profile. Figure 3 shows a typical example of COBALD data collected during the second campaign (8 July
- 2017). BSR at 455 nm and 940 nm wavelength channels <u>are</u> represented by blue and red-<u>colored_coloured</u> lines, respectively, while CI
- 335 (derived using Equation 2) is shown in the green-colored line. From Fig. 3, a sharp increase in all parameters (BSR at two
- channels, CI) found around 5 km associated with a thermal inversion (see temperature profile in Fig.3 in black colorcolour) may be
- attributed to the presence of a low-level cloud or elevated aerosol layer. Below ~5 km, the BSR profile indicates tropospheric
- 338 __aerosol distribution. Within this altitude, BSR values around 2 km indicate boundary layer confinement. Note no significant
- changes in CI within this 2 km height. Significant values in all parameters between 10 and 16 km are indicative of multiple
- _high__level cloud layers. In the rest of the campaigns, we have noticed that COBALD has captured profile information that was

missing in the lidar data.

341 was missing in lidar data.

2.4.2. CPS data processing

The phase of the cloud particle detected by CPS is determined using a quantity called degree of polarization (DOP) given by the following relation:

344 given by the following relation:

$$DOP = \frac{155 - 1125}{155 + 1125} \tag{3}$$

$$DOP = \frac{I55 - I125}{I55 + I125} \tag{3}$$

346—Since the spherical particles (water droplets) do not provide significant voltage in the cross-polarization (I125 close to 0), the

347 DOP values for such particles would be close to 1. On the other hand, the DOP for non-spherical particles (for example ice

__crystals) would take values between -1 and 1 randomly as I125 is non-zero and may or may not be greater than I55. Apart __from this, CPS can also detect the non-spherical particles in the lower troposphere whose DOP values may vary between -1_and 1.

350 and 1.

The volume of the particle detection area within CPS is non zero and estimated as ~0.5 cm³ (see section 2.3 of 352 Fujiwara et al., 2016 for the details). Therefore, when the particle number concentration is greater than ~2 cm³, more than one particle would exist simultaneously in the detection area, resulting in particle overlap and multiple scattering and thus a counting loss. The counting loss occurrence can be identified using a house keeping parameter called 'particle signal width' defined as the time taken for detection of a single particle. A simple correction of particle count using the particle signal width information is proposed by Fujiwara et al. (2016, see their section 2.3 for the details) using a factor 'f' which is (particle signal width in ms)/(1 ms) as follows. The raw counts from a CPS are corrected for multiple scattering and overlap effects using particle signal width data using Equation 4.

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$$N_{corr} = N_{meas} \times 4f^3$$
 (4)

Finally, the number of particles counted per second is converted to number concentration by assuming that the air flow at the 361—CPS detection area as 70% of the balloon ascent rate (see Appendices B and C of Fujiwara et al., 2016). The uncertainty of the number concentration when the above correction to the particle count is made (i.e., for the case of > -2 cm⁻³) has not been evaluated by Fujiwara et al., 2016. It would be safe to assume that the estimated number concentration is valid in the representation of variations in the cloud property rather than magnitude.

Finally, the number of particles counted per second is converted to number concentration by assuming that the airflow at the CPS detection area is 70% of the balloon ascent rate (see Appendices B and C of Fujiwara et al., 2016). The uncertainty of the number concentration when the above correction to the particle count is made (i.e., for the case of > ~2 cm⁻³) has not been evaluated by Fujiwara et al., 2016. It would be safe to assume that the estimated number concentration is valid in the representation of variations in the cloud property rather than magnitude.

265 ____CPS data were analyzed at their actual resolution of ~ 5m. Figure 4a shows the corrected cloud particle (number)

366 ____count (based on eq. 4) for the same day as shown in fig. Figure 3. Significant cloud particle count is found at around 5 km and from

367 __above 10 to 16 km. The number of particles counted per second at 5 km turnturns out to be high suggesting the presence of a

368 __dense (optically thick) -layer of low-level cloud. -The corresponding cloud -particle number -concentration (#/cm³) also

369 __represents (Fig. 4b) the cloud layers at the same altitudes. The DOP is estimated as per Equation 3. In Fig. 4c, DOP values are

370 __found to be clustered in the region close to 1 at ~5 km, indicating that the dense (low) cloud layer is a liquid cloud. On the

371 __other hand, the DOP values are randomly distributed between -1 and 1 in the altitude region of >10 to 16 km, indicating that

372 __these are ice clouds. In Fig. 4d and 4e, particle signal width is often greater than 1 ms and 155 is sometimes ~7.5 V for the

373 __ice cloud region between 11 and 14 km suggesting particle overlap and multiple scattering which might have led to signal saturation. This portion of the profile is more vulnerable to the data correction which has been performed and shown in Fig. 4a.

374 saturation. This portion of the profile is more vulnerable for the data correction which has been performed and shown in Fig. 331—4a.

2.4.3. Lidar data processing

Though the backscattered data at very high altitudes (>30 km) are not significant, it is used as a background signal for 334—noise correction. Range corrected signal (RCS) from MPL/Mie is calculated from noise corrected backscattered signal 335—nultiplied with range square. In general, the RCS indicates the intensity of light backscattered from molecules, aerosols and 336—clouds in the atmospheric column. However, inversion techniques are commonly applied to the RCS with an assumption of 337—lidar ratio (the ratio of extinction coefficient to backscattering coefficient) to obtain the profiles to total backscatter 338—coefficient, and extinction coefficient of cloud/aerosol separately. Ground—based lidar data were analyzed at their actual vertical as a resolutions. However, CALIPSO data were interpolated and processed at every 30 m resolution. This information is used in the discussion (sec 3.1).

340 discussion (sec 3.1).

2.4.4. Estimation of saturation relative humidity

Two dedicated radiosondes from iMet and Meisei were employed in the balloon campaigns for measurement of
meteorological parameters (temperature, pressure, relative humidity, and horizontal winds, with respect to height) as well as
to act as interface with specialized sondes COBALD and CPS, respectively. As mentioned, temperature and pressure profiles
from the radiosonde were used in post processing of COBALD sonde to scale the signal to the molecular Rayleigh
scattering. In addition to this, radiosonde temperature, relative humidity is useful in understanding the state of saturation of
water vapor in the column. By convention, relative humidity reported from radiosonde is always over the plane surface of
liquid water (because radiosonde relative humidity sensors are factory calibrated) even below 0°C. This is because water
droplets may exist even below 0°C and down to 30 to 40°C (in the form of super cooled liquid) in the atmosphere.
Saturation relative humidity (SRH) defined in Fujiwara et al. (2016) (see also Fujiwara et al., 2003) as the ratio of saturation
vapor pressure over the plane surface of ice (e_{si}) to water (e_{si}) expressed in units of percentage can be a good metric to
describe the state of water vapor in the atmosphere such as sub-saturation, saturation and/or super-saturation in particular at
air temperatures below 0°C (with respect to ice). In this study, both e_{si} and e_{si} are calculated using Hyland and Wexler
formulation (see Appendix A of Murphy and Koop, 2005) by using radiosonde temperature data. For the temperatures
warmer than 0°C, water vapor saturation is indicated by 100% RH. For temperatures colder than 0°C, water vapor is said to

356 be saturated if RH ~= SRH and super saturated when RH > SRH. This information is used in discussion (sec 3.2).

Two dedicated radiosondes from iMet and Meisei were employed in the balloon campaigns for the measurement of meteorological parameters (temperature, pressure, relative humidity, and horizontal winds with height) as well as to act as an interface with specialized sondes COBALD and CPS, respectively. As mentioned, temperature and pressure profiles from the radiosonde were used in the post-processing of the COBALD sonde to scale the signal to the molecular Rayleigh scattering. In addition to this, radiosonde temperature, and relative humidity is useful in understanding the state of saturation of water vapour in the column. By convention, relative humidity reported from radiosonde is always over the plane surface of liquid water (because radiosonde relative humidity sensors are factory calibrated) even below 0° C. This is because water droplets may exist even below 0° C and down to -30 to -40 $^{\circ}$ C (in the form of supercooled liquid) in the atmosphere. Saturation relative humidity (SRH) is defined in Fujiwara et al. (2016) (see also Fujiwara et al., 2003) as the ratio of saturation vapour pressure over the plane surface of ice (e₈) to water (e₅) expressed in units of percentage can be a good metric to describe the state of water vapour in the atmosphere such as sub-saturation, saturation and/or super-saturation in particular at air temperatures below 0° C (with respect to ice). In this study, both e₈I and e₈I are calculated using Hyland and Wexler formulation (see Appendix A of Murphy and Koop, 2005) by using radiosonde temperature data. For temperatures warmer than 0° C, water vapour saturation is indicated by 100% RH. For temperatures colder than 0° C, water vapour is said to be saturated if RH ~= SRH and super-saturated when RH > SRH. This information is used in the discussion (see 3.2).

2.4.5. Discrimination of cloud and aerosol in a balloon profile

COBALD measurement always represents backscatter light from the combination of aerosol and cloud. Obtaining information on aerosol (only) is not possible (for COBALD) in the presence of clouds, and the corresponding regions have to be identified and rejected. This cloud clearing has been established previously for studies related to the UTLS region (Vernier et al., 2015, 2018). Contrary, for cloud investigation, the COBALD was used in combination with the Cryogenic Frost point Hygrometer (CFH) to identify super saturation (with respect to ice) below, above and within the cirrus clouds to improve the understanding of microphysical processes in cirrus clouds (Cirisan et al., 2014). This sonde in addition detected volcanic aerosol tracers in the stratosphere (Vernier et al., 2020). The Asian Tropopause Aerosol Layer (ATAL) is a well-documented phenomenon occurring in the UTLS region during the Summer Monsoon Season over South Asia. Vernier et al. 266—(2015) proposed two cloud clearing methods for discrimination of aerosol from cirrus clouds in ATAL region using the physical quantities Color Index (CI), relative humidity over ice (RHi) and backscatter ratio (BSR) at 940 or 532 nm (the latter was interpolated from the 455 nm data for inter-comparison with CALIOP). In the presence of CFH data, the RHi

acloud filtering approach classifies ATAL/UTLS aerosol layers by the criterion BSR (at 532 nm) < 1.3 and RHi < 70%. For measurements of COBALD alone, the CI method indicates clouds with CI < 7 and BSR (at 940 nm) < 2.5. It was shown that both methods effectively discriminate ATAL aerosol from upper tropospheric thin clouds. Brunamonti et al. (2018) also applied the cloud clearing criteria (BSR at 940 nm < 2.5, CI < 7 and RHi < 70%) following Vernier et al. (2015) and found a clear signal of enhanced BSR (at 455 nm) between 1.04 and 1.12 indicative of the aerosol population in the ATAL region. However, it is noted that the methods proposed by Vernier et al. (2015) and Brunamonti et al., (2018) were developed for the UTLS aerosol and their applicability to COBALD measurements of boundary layer and/or mid tropospheric aerosol needs to be validated.

In the present study, we made use of a CPS sonde in tandem with COBALD. As already mentioned, CPS is sensitive to particles in the size range of >2 µm and hence detects clouds particles (both liquid droplets and ice crystals) and sometimes coarse mode aerosol particles (such as dust) of these sizes. Fujiwara et al. (2016) has demonstrated in detail the potential of a CPS sonde using balloon sounding carried out at mid latitude (Japan) as well as tropical sites (Indonesia).

Narendra Reddy et al. (2018) have used a CPS measurement from Gadanki to validate their method of retrieving cloud

vertical structure based on radiosonde measurements. Therefore, to better segregate the clouds from aerosols in the COBALD measurements, CPS sonde has added advantage to the methods using simultaneous RH data described by Vernier et al. (2015) and Brunamonti et al. (2018). This implies wherever the cloud present in a profile, CPS identifies it (along with its phase) and the corresponding COBALD particle backscatter data refers to the cloud. The rest of the particle signals in the COBALD profile should correspond to aerosol. However, it may correspond to the (thin) cloud also which might have been missed or undetected by a CPS. So identification of aerosol and cloud in an altitude profile is key measurement of this paper.

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2.4.6. Estimation of Aerosol-cloud-interaction Index

-Balloon data from all campaigns can be pooled to explore the aerosol-cloud relationship. For this purpose, a simple 391—scheme is developed to carry out the required computations. CPS profile data is looked for a cloud layer presence present in the 392 — altitude regime of liquid or low-level clouds (below 5 km). As already discussed, CPS also identifies particle particles of non-393 spherical nature. In order to To separate cloud particles from non-spherical particles, the following conditions have been — imposed on various CPS measured parameters. Cloud particle count should be >10 #/s, cloud droplet number concentration 395 >10⁻³#/cc, DOP>0.6, relative humidity >95% and temperature >0 degC. As there is a chance of randomly distributed data 396—points in the measurement column satisfying the above conditions, we considered only those points present continuously up 397—to a thickness minimum of 100 m (with at least one point for every 40 m). Further, COBALD data of blue backscatter from 398—100m, 200m, 300m, 400m and 500m below the cloud base has been picked up separately (for the same profile) as a proxy of 399— aerosol to check its influence on the cloud above. As already mentioned, post—processed data of backscatter ratio from - COBALD sonde represents the contribution from both molecule and particle (cloud and/or aerosol). Hence, the particle backscatter 401 — ratio is obtained by subtracting the backscatter ratio from one. In order to To avoid high values of particle (blue) backscatter ratio 402 possibly originating from the interaction with high relative humidity usually expected near to cloud base (boundaries), we — have adopted two methods. First, high values of particle (blue) backscatter below the cloud base are removed if beyond a 404—threshold value of 3.15. The threshold is arrived at using a box plot (figure not shown) drawn for all the particle backscatter 405—data set (for sounding with clouds) from cloud base to 500 m below and found that 3.15 corresponds to the upper whisker

406 _(Q3+1.5*(Q3-Q1)). Further, the particle backscatter data is corrected for relative humidity in case a statistically significant (p-value <=0.05) and good correlation (>0.71) is found among relative humidity and particle backscatter ratio. A typical example from the scheme is shown in Fig. 5 for the launch conducted on 01 November 2018 which depicts cloud layers, blue particle backscatter ratio below the cloud along with shaded black dots (representative of aerosol backscatter ratio). The scheme is applied to the balloon sounding and the results were discussed in sec 3.4.

407 (p value <=0.05) and good correlation (>0.71) is found among relative humidity and particle back scatter ratio. A typical
408 example from the scheme is shown in Fig. 5 for the launch conducted on 01 November, 2018 that depicts cloud layers, blue
409 particle back scatter ratio below cloud along with shaded black dots (representative of aerosol back scatter ratio). The
410 scheme is applied to the balloon sounding and the results were discussed in sec 3.4.
411 Aerosol cloud interaction can be quantified based on an index (ACI) using three methods discussed in Feingold et
412 al., 2003, 2006. ACI is defined as slope of the linear fit between logarithm of cloud proxies such as cloud optical depth,
413 cloud particle radius and cloud droplet number with logarithm of aerosol proxy. ACI in this study has been estimated using
414 the equation (5).
415

416 ACI = \frac{4\log Ne}{1\log Ne} \frac{(5)}{2\log Ne}

where Aerosol-cloud interaction can be quantified based on an index (ACI) using three methods discussed in Feingold et al., 2003, 2006. ACI is defined as the slope of the linear fit between the logarithm of cloud proxies such as cloud optical depth, cloud particle radius and cloud droplet number with the logarithm of aerosol proxy. ACI in this study has been estimated using the equation (5).

$$ACI = \frac{dlogNc}{dlogBSRb}$$
 (5)

- 416—Where cloud droplet number count (Nc) is taken as cloud proxy whereas BSRb is the COBALD (blue) particle back scatter
- 417 <u>backscatter is (BSRb)</u> taken as aerosol proxy and interaction between both is indicated by index ACI. It is to be noted that cloud particle number
- 418 concentration is used here to represent cloud property instead of droplet number concentration as the former is a direct
- 419 __measurement (of CPS). The slope of the linear fit between the natural logarithm of Nc and BSRb indicates the magnitude of the aerosol-
- 420 cloud interaction (ACI) index) which should be between 0 and 1 (Feingold et al., 2003). (Feingold et al., 2003). Note this the condition (shown in eq.5) is
 - independent of the liquid water path as it verifies/quantifies the aerosol activation process.

2.4.7. Uncertainty in ACI estimation

- The uncertainty in ACI stems from both <u>uncertaintyuncertainties</u> in the COBALD backscatter ratio and CPS cloud particle counts.
- 424—The slope of the curve (linear fit of data on a log-log scale) can be written as a function of BSRb (blue back scatter backscatter

ratio) and Nc

_(cloud particle

count) as,

425

$$ACI = f(BSRb, Nc) = \frac{LogNc - C}{LogBSRb}$$
 (6)

where
$$ACI = f(BSRb, Nc) = \frac{LogNc - C}{LogBSRb}$$
 (6)

427 <u>Where</u> 'C' is the intercept of the curve. Partial The partial derivative of f(BSRb, Nc) with respect to BSRb and Ne indicates uncertainty in

_ACI with respect to uncertainty in individual parameters (Nc and BSRb). The combined uncertainty (UC) in ACI is given by the equation.

429 the equation,

430

$$UC = \sqrt{\frac{\theta f(BSRb,Nc)}{2} 2} \frac{2}{(uBSRb) + (uNc)} \frac{\theta f(BSRb,Nc)}{2} 2$$
(7)

 $\frac{431}{\text{where}}UC = \sqrt{\left(\frac{\partial f(BSRb,Nc)}{\partial BSRb}\right)^2 \left(uBSRb\right)^2 + \left(\frac{\partial f(BSRb,Nc)}{\partial Nc}\right)^2 \left(uNc\right)^2}$ (7)

Where uBSRb and uNc are individual uncertainties.

3. Results

- The multi instrument data from the BACIS campaigns are presented in this section. Consistency among multiple
- 435 measurements is discussed in Section 3.1, data from a particular balloon sounding (campaign) are interpreted in detail in
- 436 Section 3.2, statistics on cloud/acrosol features are given in Section 3.3, and finally, the relationship between acrosol and
- 437 cloud is illustrated in Section 3.4.
 - 3.1. Consistency. Comparison of balloon measurements
- 439 The combination of COBALD and CPS sondes is used for the first time for in situ measurement of aerosols and
 - 440 elouds. Therefore, it It is important to know the performance of these sondes in comparison to other measurement techniques.
- Here, we make use of data from two pilot campaigns to demonstrate the consistency of balloon-borne measurements with that of ground-based
- and spaceborne remote-sensing instruments. As mentioned previously, the first two (pilot) campaigns have been conducted in line with the
- proposed concept.
 - 441 Here, we make use of data from two pilot campaigns to demonstrate the consistency of balloon borne measurements with
 - 442 that of ground based and space borne remote sensing instruments. As mentioned previously, the first two (pilot) campaigns
 - 443 have been conducted in line with the proposed concept.
 - 3.1.1. Pilot campaign-1 (launch held on 06 June 2017 at 01:50 LT)
 - The CALIPSO satellite overpass time for the first pilot campaign was around 02:00 LT of on 06 June 2017 (starting
 - time of the track). The balloon was launched at 01:50 LT on the same day just before CALIPSO overpass time. Combined
- measurements from specialized balloon-borne sondes and ground-based and space-borne lidars obtained during the first launch of the campaign
- are shown in Figure 6.
 - 447 measurements from specialized balloon borne sondes, ground based and space borne lidars obtained during the first launch
 - 448 of the campaign are shown in Figure 6.

The BSR from COBALD sonde at 455 nm (950 nm) is plotted in Fig. 6d as <u>a</u> blue (red) line. BSR from both <u>the</u>

450—channels <u>areis</u> referenced to <u>the</u> same x-axis scale. Similarly, cloud particle number concentration (dN, #/cc) from CPS sonde is

451—plotted as black dots (Fig.6e). On the other hand, range corrected signal (RCS) from ground—based lidars (Mie, MPL) is

452—averaged over a short period <u>of time</u> during <u>the</u> CALIPSO overpass and plotted in magenta (averaged from 01:50 to 02:00LT),

453—orange <u>colorcolour</u> lines (averaged from 01:50 to 01:55 LT), respectively (Fig.6f). The total attenuated backscatter (km⁻¹ sr⁻¹) from

454—CALIPSO is also averaged for the profiles found nearest to the location and shown in <u>an</u> olive green <u>colorcolour</u> line (Fig.6f). The

- 456—responses from clouds and aerosols in the atmosphere. At this point of discussion, we have not distinguished their contributions.
- 457—balloon drifts away from the launch location with time, therefore, it is also required to check the degree of co-location of
- 458 __measurements with the lidars. In order to To facilitate this, a portion of nocturnal variation (representing the balloon launch
- 459—duration) in range corrected signal from both Mie and MPL is shown in Fig. 6b and 6c, respectively. The CALIPSO
- 460 overpass track consisting of 166 profiles is also plotted as a function of longitude (Fig. 6a). For the sake of easy
- identification of simultaneous lidar measurements, the balloon indices such as height and drift (radial distance from launch
- location) are overplotted as a function of time on contour maps as shown in black and red-coloured lines, respectively (Fig. 6b and 6c).
 - 462 location) are over plotted as a function of time on contour maps as shown in black and red colored lines, respectively (Fig.
 - 463 6b and 6c).
 - Balloon--borne in-situ measurements from COBALD and CPS show significant peaks in the lower tropospheric
 - 465 (below 4 km) and upper troposphere (between 13 and 17 km) at the same altitude regions. It can be seen from the Fig. 6d and 6c,
 - 466—that there is a good resemblance among between the in-situ and MPL measurements in the lower tropospheric (below 4km). This is
 - 467—because almost no change in the atmospheric conditions as the balloon took approximately 15 minutes to reach an altitude of
 - 468 4 km with a radial distance of 5 km away from the launch location. Mie lidar information is not reliable for this altitude
 - 469—region (below 4 km) as it is not in the overlapping region of the telescope viewing geometry and laser beam dispersion (see
 - section 2). CALIPSO signal also looks to be dispersed and noisy for this altitude region. This could be due to the attenuation
- the signal from the top side layers as seen in Fig. 6a at a longitude of 79.24° E (nearest profiles longitude).
 - 471 of the signal from top side layers as seen in Fig. 6a at longitude of 79.24° E (nearest profiles longitude).
 - Next to this is the sharp peak seen in COBALD red channel at slightly below 9 km (Fig. 6d). This again can be seen
 - 473—in Mie and MPL profiles also (Fig. 6b, 6c) but at 8.4 km (slightly below cloud detection height). However, it is to be noted
- 474—that these profiles are averaged for a short duration of time during the CALIPSO overpass. In fact, there There is another peak in
- 475— the Mie lidar profiles at ~7.2 km₅ (Fig. 6b₂), which is not seen in COBALD. It is approximately 45 min (around 02:45 LT)
- 476—from the time of launch when the balloon reached the altitude of ~9 km and 5.8 km away before detecting a sharp peak. As
- 477—there is no significant range corrected signal during this time and altitude in the ground—based lidar data (Fig. 6b and 6c), the
- 478—sharp layer detected by COBALD may be a localized cloud layer or a passing layer which might have ascended/descended.

Exact attribution can be made with a detailed study but it is beyond the scope of the current analysis.

-Further, the balloon drift was within a 10 km range until 03:00 LT when it reached heights of ~12 km. This implies 481 — weak horizontal winds and thus weakweakly associated wind drifts as well. Thereafter, the balloon started drifting rapidly due to 482 high wind speeds between 10 and 20 m/s. Both the in-situ measurements of COBALD and CPS show strong double peaks — from ~13-15.5 km and 16-16.5 km (Fig. 6d, e). Profiles from Mie, MPL and CALIPSO also showed similar peaks except 484 for MPL for which the upper side peak is missing (Fig. 6f). It may be once again noted that, these profiles are averaged for a 485— short duration of time during the CALIPSO overpass the and return signal from MPL at high altitudes (~16 km) during the same 486— time suffered severely due to the presence of a mid-tropospheric cloud layer (at ~7 km) as seen in Fig. 6c. This is not the — case for the return signal from Mie Lidar as the power and energy of the Mie laser isare relatively high (Fig. 6b). However, strong 488—double peak structures can be noticeable in the simultaneous observations of both ground—based lidars (Mie and MPL) at 489 — similar heights during the time corresponding to the balloon altitude of 13 km (post 03:00 LT). Therefore, the same upper - tropospheric cloud layers detected in the ground, space bornespaceborne and in-situ measurements suggest they are extended cloud - layers. Dynamical aspects of south west the southwest monsoon over the sub-continent refers refer to the presence of Tropical Easterly Jet (TEJ) 492 which is strong enough to swipe anvil clouds of meso scale mesoscale convective systems to thousands of kilo meters (Sathiyamoorthykilometres (Sathiyamoorthy et al., 2013).

493 et al., 2013).

3.1.2. Pilot campaign -2 (launch held on 08 July 2017 at 01:35 LT)

- The starting time of the CALIPSO <u>over passoverpass</u> track for the second pilot campaign was at 02:00 LT. The balloon was
- 496—launched at 01:35 LT nearly 30 minutes before the starting time of the CALIPSO overpass. Data from all the instruments are
- 497 __plotted in Figure 7, which is prepared the same as Figure 6. MPL₇ and Mie profiles were averaged from 01:50 to 02:00 LT (close to
 - the CALIPSO overpass time over Gadanki).
- The observations from COBALD and CPS are matching reasonably well (Fig. 7d, e) as significant peaks were
- 500—found in the lower troposphere (0-5km) and upper troposphere (10-16 km). The profiles from space bornespaceborne and ground-based

501—_lidars (Fig. 7f) also show a similar response as in-situ measurements (both in <u>the</u> lower and upper troposphere) except that lidar

502 __measurements exhibit additional peaks in the mid-troposphere (between 5 and 10 km). It is to be noted that profiles from <u>lidar</u>

measurements are averaged over a short period, as mentioned before.

503 lidar measurements are averaged over a short time period, as mentioned before.

504 -Simultaneous observations from both the space bornespaceborne (CALIPSO), and ground-based (Mie and MPL) lidars isare shown in 505 — Fig. 7 a, b &c respectively. Due to high wind speeds (10-20 m/s) the balloon drifted about 5 km away from the launch site 506 — while crossing boundary layer height (~2km). The features found within the boundary layer as measured by in-situ 507 instruments (Fig. 7d) are in agreement with that of MPL measurements (Fig. 7c) for the same altitude region. Note that, Mie 508—lidar measurements are not reliable at these low altitudes and CALIPSO has not yet started passing by the launch site. The 509 balloon continued to drift away but with thea reduced wind speedsspeed of 10 m/s. At around 4.3 and 4.7 km (10 km away of 510—from the launch site), the balloon had detected two layers (strong peaks). The time corresponding to this balloon height was around 01:50 511 LT and at this point of time, two layers can also be seen in both the ground—based lidars at the same altitudes (Fig. 7b and c) 512 indicating the presence of an extended layer (which is evident in both the in-situ and ground-based measurements). In fact, the The layer 513— at 4.7 km was also noticeable in the CALIPSO profile measurements (Fig. 7a). This is because the CALIPSO started coming 514 _close byto the site when the balloon was at this height and the CALIPSO profile corresponding corresponds to an average of (nearest) profiles at 515— around 79.32° E longitude (Fig. 7a). Further, the balloon started drifting towards the launch site until it reached a height of 516 ~ ~7.5 km at a distance of ~13 km away. While moving towards the site, the balloon started detecting the layers starting from 517—11 km. The time corresponding to the balloon height of 11 km is around 02:45 LT and at this point of time simultaneous 518 — MPL data show almost weak returns (Fig. 7c), whereas Mie lidar showshows a better return signal (Fig. 7b) than MPL. In 519 continuation toof this, the balloon started drifting further towardstoward the site until it reached as close as ~3.5 km at a height of ~12.5 520 km. Thereafter, it started moving rapidly away from the location with high wind speeds due to the characteristic of TEJ. 521 Multiple layers of clouds have been nicely captured by in-situ measurements from 11 km to ~ 16 km. However, prominent 522 — lidar returns were not noticeable in the simultaneous observations of Mie and MPL. This is because of a strong lower - tropospheric cloud layer present at around 5 km limiting the detection of upper tropospheric cloud layers by both ground-524 based lidars. However, all these layers were prominently captured in CALIPSO observations as it is top-down laser probing. down laser probing. In summary, the data from both pilot campaigns illustrate the limitations of the ground-based and/or spaceborne lidars in detecting the complete cloud vertical structure. At the same time, in-situ data emphasize reasonable agreement of the balloon-borne

measurement with the ground-based as well as space-borne measurements and add to the remote sensing techniques while detecting the missing portion of the cloud vertical structure.

525 In summary, the data from both the pilot campaigns illustrate the limitations of the ground based and/or space borne lidars in

526 detecting the complete cloud vertical structure. At the same time, in situ data emphasize the consistency of the balloon borne

527 measurement with the ground based as well as space borne measurements and complements to the remote sensing technique

528 while detecting the missing portion of the cloud vertical structure.

A typical example of high-resolution vertical wind measurements obtained from MST Radar duringon 8 July 2017 is shown in Figure 8(f) and profiles of all the three-dimensional winds averaged between 02:30 LT to 03:30 LT are shown in Figures 8(a)-(c) to compare the wind measurements. We also superimpose the zonal and meridional winds in the respective panels obtained from radiosonde for comparison. Consistency in the measured winds in these two independent techniques panels obtained from radiosonde for comparison. Consistency in the measured winds in these two independent techniques panels obtained from radiosonde for comparison. Consistency in the measured winds in these two independent techniques panels obtained from radiosonde for comparison. Consistency in the measured winds in these two independent techniques panels obtained from fant and panels in the second panels of the measured winds in these two independent techniques panels of m/s, secretly wind velocities exceeding 50 m/s, and similar features panels of m/s, and panels of the panels of the measured winds and panels of synoptic-scale systems (Fig. 8a). In addition, satisfaction panels of the panels of the panels of the second panels of the pa

presence of clouds. Doppler width (Fig. 8e) shows higher values below the boundary layer and UTLS region suggesting

542 active turbulence.

occur in the

541

active turbulence.

3.2. Interpretation of aerosol and cloud features in a balloon profile

- In order to fulfill To fulfil the primary objectives of the campaign, it is a priority to distinguish aerosol and cloud in a balloon
- 545—borne in-situ profile. In connection with this, combined measurements of CPS and COBALD from a balloon sounding held
- 546— on 27 June 2019 at 23:30 LT is are interpreted as shown in Figure 9. This particular sounding is selected because it showcases
- 547—all the features that can be detectable by a CPS sonde in a profile such as liquid cloud, super cooled supercooled liquid cloud, ice cloud,
- and non-spherical particle layers. INSAT3D brightness temperature shown in Figure S1 indicates the evolution of a localized

cloud system north of the observational site initiated a few hours before the launch and eventually spreading over the site.

549 cloud system north of the observational site initiated few hours before the launch and eventually spreading over the site.

- To characterize the background conditions of the atmosphere, meteorological parameters such as relative humidity
- 551—(RH), and temperature (T) obtained from RS-11G radiosonde are plotted in Fig. 9a (wine red and blue colorcolour lines). In the Fig.
- 552—9a, SRH is also shown (in yellow colorcolour). The SRH and RH can be read from the same top-X scale in wine red colorcolour as shown in
 - 553 _Fig. 9a.

- -The CPS sonde usually features clouds that can be better identified with the information based on DOP. — and corresponding profiles of T, RH, and SRH. From Fig. 9d, DOP values close to 1 (from 0.6 to 1) are noticeable at different 556—altitude ranges in the profile viz., 3.5 to 5.5 km, 8.6 to 9 km and DOP values spread (-1 to 1) between 9 and 11 km. In the — altitude range from 3.5 to 5.5 km, CPS detected multiple liquid cloud layers, corresponding to the multiple layers of 100% 558—RH. However, the corresponding COBALD blue and red backscatter data points are limited (Fig. 9b). This is because 559 COBALD backscattered signals showed missing values due to saturation of photo-diodesphotodiodes in the presence of thick liquid 560 cloud layers and that had to be removed during post-processing of data and are not discussed further. The layer extending between 3.5 and 3.8 km (300 m thick) is observed with RH and T in the range 99-100% and 7--8.7°C, respectively, indicating saturation of water vapor vapour with respect to liquid (RH~=SRH) which is conducive forto the - formation of a (liquid) cloud. Further, the majority of droplet number concentration concentrations in this liquid cloud layer range between 0.1 564—to 1 #/cm³. A rough estimate onof particle size information (water droplet or ice crystal) can be inferred from CPS voltage data 565 (155). According to Fujiwara et al. (2016), 155 mostly lying below 1V suggests these droplets are sized ~2-13 µm. Another 566— liquid cloud layer extending from 4 to 4.4 km (400 m thick) is observed with vapor vapour saturation over liquid (100% RH) and 567 temperatures from 3-6°C. CPS shows that droplet number concentration peaks in the range 0.1-10 #/cm³ with the highest in 0.1-568—1 #/cm³. The intensity (I55) values (<1 V) indicate the majority of droplet sizes are ~2-13 μm. The third liquid layer in the range 569 of 3.5 to 5.5 km is observed between 5.1-5.5 km (400 m thick) with the highest droplet number concentrations in the range of 0.1-10 570 #/cm³, sized around 2-13 µm (I55<1 V). However, RH observations show 100%RH or RH>SRH ie. water vapor vapor super-571—saturated over ice at temperatures slightly below 0° C (0 to -3°C), suggesting that the cloud layer may be composed of super
- 573—saturation of vapor vapour over ice at 100%RH or RH>SRH and -21.5 to -23.5°C temperatures. The observed features of droplet

-cooled supercooled liquid droplets. Another clear super cooled supercooled cloud layer was detected between 8.6 and 9 km (400 m

_number concentration and particle size are similar to those <u>forof</u> the <u>super cooled</u> cloud found in the lower atmosphere. The

thick) with super-

only difference that could be noticeable is in the distribution of DOP values as shown in Figure S2, which indicate the more indicates the tendency of droplets toward non-sphericity in the mid-tropospheric supercooled liquid cloud. COBALD signals were found limited for all liquid/supercooled layers discussed above.

576 tendency of droplets towards non sphericity in the mid tropospheric super cooled liquid cloud. COBALD signals found
577 limited for all liquid/super cooled layer discussed above.

The top mostopmost layer in the upper troposphere spreading from 9.5-11 km is an ice cloud layer as per its DOP values.

The temperatures within the cloud are found in the range of -22 to -40°C. RH values are >SRH, suggesting the super-saturation of vaporvapour (over ice) within the ice cloud. Histogram The histogram of data for all the parameters obtained from COBALD and CPS for this

ice cloud layer (9-11 km) is shown in Figure 10. The number concentration of ice cloud particles (Fig. 10a) lies between 0.01 to 10 #/cm³ with a peak in the range of 0.1-1 #/cm³. Non-sphericity of particles is elearly seen by the wide distribution of DOP values in the range -0.4 to 1 with the majority of them lying close to 0 (Fig. 10b). In particular, DOP values close to 0 indicate section. 2) that both plane and cross-polarization intensities of scattered light (155 and 1125) are comparable. This happens when both detectors get saturated due to a large number of small size particles and/or a few large-sized ice particles or both. In support of this, the 155 values (Fig. 10c) are found to peak in the 7-8 V range (~7.5 V) for such cases. Further, if saturation services are due to large size then they may correspond to ~80-140 µm or greater ice particles (corresponding to 155 of services are due to large size then they may correspond to ~80-140 µm or greater ice particles (corresponding to 155 of services is clouds. Apart from this, the second peak in 155 noticed below 1V corresponds to ice particles roughly sized between and 14 µm.

The COBALD BSR corresponding to this ice cloud arejs symmetrically distributed from 1-10 and 10-100 for blue

592—_(Fig. 10d), red (Fig. 10e) wavelengths, respectively. However, there are some observations which are beyond 10(100) at blue

593—_(red) wavelengths. Similarly, the CI for this cloud (Fig. 10f) is found mostly between 10 and 20 but for a few instances, it is

594—_observed from 20 to 40. From the definition (see section 2), the CI is independent of the number concentration hence it can

595—_be used as an indicator of the mode radius of particles. With the assumption of athe single—mode log-normal size distribution of

596—_spherical aerosol/cloud particles, Mie calculations show CI is 4-10 for small particles of mode radius up to 1-2 µm and 14-20

597—_for large particles of 2-20 µm. CI converges to around 20 as a geometric limit for very large particles of mode radius > ~50

598—_µm. However, CI can have values >20 at mode radius 2-20 µm as CI is a non-monotonous function of mode radius and

599—_exhibits Mie oscillations (due to variations of scattering efficiencies with size parameter). The amplitude and frequencies of

600—Mie oscillations depend on the width of the log normal size distribution assumed. At width higher than say 2 (represent poly

589 can be applied for these ice clouds. Apart from this, a second peak in 155 noticed below 1V corresponds to ice particles

- 601 disperse aerosol populations) these oscillations are mitigated and lead to monotonous dependency of CI and mode radius.
- 602 For stratospheric aerosols in the size range 0.02 0.4 μm the CI is found to be in the range 5.7 (Rosen and Kjome, 1991). This

Mie oscillations depend on the width of the log-normal size distribution assumed. At a width higher than say 2 (representing polydisperse aerosol populations), these oscillations are mitigated and lead to a monotonous dependency of CI and mode radius. For stratospheric aerosols in the size range of 0.02-0.4 μm, the CI is found to be in the range of 5-7 (Rosen and Kjome, 1991). This is because stratospheric aerosols exhibit size distributions with narrow standard deviations. Aerosol size distributions in the
 604—UTLS region may also be assumed as log-normal (similar to stratospheric aerosols) hence the criteria CI<7 might have
 605—suited for cloud filtering in the ATAL region (see Section 2). For the present case of the ice cloud layer (9-11km) discussed above,
 606—CPS indicates the presence of small (2-14 μm) and very large ice particles (>80 μm). So, the standard deviation of log-normal
 607—size distribution in the cloud layer of large particle mode must be wider. Therefore, Mie oscillations may be expected to be
 608—at a minimum. Probably because of this, the majority of CI values for the cloud layer are found between 15 and 20, which may
 609—correspond to a mode radius of > ~50 μm (geometric limit). It may also be concluded that the CI of 20-40 (with very few
 610—values >30) corresponds to small particles of mode radius > 2-20 μm (due to Mie oscillations). COBALD size interpretations
 611 (based on CI) are in support of CPS-based size interpretations. Since the majority of CI falls between 15 and 20, the 155 of ~7.5V in CPS
 612 (based on CI) are in support of CPS-based size interpretations. Since the majority of CI falls between 15 and 20, the 155 of ~7.5V in CPS

612 -7.5V in CPS would have been caused by large size particles.
613 In the lower troposphere up to 2 km where water vapor is well sub-saturated (50-70 %RH), CPS also shows particle
614 signals (Fig. 9c). The DOP values range from -0.4 to 1 but with lower number concentrations (0.001-0.01 #/cm³) and less
615 than 1 V of backscatter intensity (155), indicating these particles as non-spherical in shape similar to the ice-cloud particles.
616 Since it is not possible to have ice-cloud particles at these lower altitudes in dry conditions (RH<70%), it may be possible
617 that these particles are coarse mode non-spherical aerosol particles. COBALD observations indicate CI of 11-12. Thus, both
618 the COBALD and CPS observations indicate aerosol may be of size -2.5 µm. To investigate the possible origin of these
619 coarse mode aerosol particles, Hysplit 7 day back trajectories for 5 days before and after the date of launch are calculated
620 and shown in Figure S3 (in different color lines). These Hysplit back trajectories (Stein et al., 2015) indicate the air parcel
621 path ways ending at every 1 km altitude from 1 to 5 km over Gadanki at the time of balloon launch (18 UTC). It can be seen
622 (from Fig. S3) that, the air masses were originated from the Indian Ocean passing through the Arabian Sea before reaching
623 the Gadanki location for heights 1 to 3 km. Therefore, the air masses were of marine origin, and the particles were possibly
624 coarse mode water soluble particle (such as sea salt) which can grow hygroscopic due to the availability of moisture over the

Ocean surface (Mishra et al., 2010; Ratnam et al., 2018). The rain water chemical analysis reported by Jain et al. (2019) at Gadanki supports this conclusion as they found dominance of water soluble ions during the southwest monsoon (June to September). Above 3 km altitude, the air masses are coming from the Saharan desert region within 7 days which may bring

628—non spherical coarse mode dust particles to the launch location (Mishra et al., 2010). Thus, in case of lower tropospheric

In the lower troposphere up to 2 km where water vapour is well sub-saturated (50-70 %RH), CPS also shows particle signals (Fig. 9c). The DOP values range from -0.4 to 1 but with lower number concentrations (0.001-0.01 #/cm³) and less than 1 V of backscatter intensity (155), indicating these particles as non-spherical in shape similar to the ice cloud particles. Since it is not possible to have ice cloud particles at these lower altitudes in dry conditions (RH<70%), it may be possible that these particles are coarse mode non-spherical aerosol particles. COBALD observations indicate a CI of 11-12. Thus, both the COBALD and CPS observations indicate aerosol may be of size ~2-5 μm. To investigate the possible origin of these coarse mode aerosol particles, Hysplit 7-day back trajectories for 5 days before and after the date of launch are calculated and shown in Figure S3 (in different colour lines). These Hysplit back trajectories (Stein et al., 2015) indicate the air parcel pathways ending at every 1 km altitude from 1 to 5 km over Gadanki at the time of balloon launch (18 UTC). It can be seen (from Fig. S3) that, the air masses originated from the Indian Ocean passing through the Arabian Sea before reaching the Gadanki location for heights of 1 to 3 km. Therefore, the air masses were of marine origin, and the particles were possibly coarse-mode water-soluble particles (such as sea salt) which can grow hygroscopically due to the availability of moisture over the Ocean surface (Mishra et al., 2010; Ratnam et al., 2018). The rainwater chemical analysis reported by Jain et al. (2019) at Gadanki supports this conclusion as they found dominance of water-soluble ions during the southwest monsoon (June to September). Above 3 km altitude, the air masses are coming from the Saharan desert region (within 7 days) which may bring non-spherical coarse mode dust particles to the launch location (Mishra et al., 2010). Thus, in the case of lower tropospheric coarse mode aerosol (water-soluble aerosol particles), the CI can be >7

In the altitudes of 6-8.5 km (Fig. 9), CPS has detected no cloud. However, COBALD data shows, that CI values ranging 631—from 3-8 in the altitude range of 6-7 km and 3-12 in the altitude range of 7-8.5 km may indicate the presence of aerosol particles undetectable

by <u>a CPS</u> (i.e., of sizes $<2 \,\mu m$). RH values indicate sub-saturated conditions throughout this altitude region. However, between

633 ___7-km and 8.5 km, RH increases and becomes greater than the ice saturation RH values (saturation with ice). Corresponding

634__to this RH change, Cl, as well as red channel BSR, is also found to increase. This suggests the growth of small aerosol

_particles under high humidity conditions until the RH approaches ice saturation where super_cooled_supercooled_liquid droplets are

_observed (8.6-9 km) in CPS whose features have been discussed already. Since the COBALD CI values are mostly <10 in

this altitude range, the majority of particles detected might be sized up to 1-2 µm.

637 this altitude range, the majority of particles detected might be sized up to 1.2 μm.

3.3. Statistics on COBALD colorcolour index

In order toTo generalize the optical properties specific to aerosol and cloud, combined data from COBALD and CPS

(from multiple launches) has been investigated in detail. The liquid/super cooled cloud, ice cloud and non-spherical particle

(41—layer depth are carefully identified with the help of DOP data from CPS (discussed in Section 2). The corresponding data of

(42—temperature, relative humidity, BSR, CI, and peak particle number concentration have been picked up for estimating

(43—statistics. Further, threshold values of COBALD parameters were tried to identify for the said categories of aerosol and cloud

(44—cases. Among 15 balloon soundings, those soundings were considered where CPS detected cloud particles and both blue and

(45—red channel data are not missing from COBALD. With these conditions, 8 balloon soundings were identified for estimating

646 statistics.

Table 3 shows the mean (median) values of CI and other parameters corresponding to the ice cloud layers from 7

648—Launches. Fig. 11(a) shows the complete statistics of CI in the form of a box plot for the same ice eloudscloud layers. Fig. 11(b)

649—shows a histogram of CI from each campaign indicated by different eolorscolours. From Table 3, ice clouds are seen above 9 km with

650—temperatures colder than -20°C. For example, an ice cloud layer was found between 9.3 to 16 km on 30 April 2019 with

651—temperatures in the range of -22 to -79°C, RH close to SRH and mean (median) value of CI is 19.4 (19.3), BSR is 16.4 (8.6) at

652 455 nm, 302 (147) at 940 nm, peak droplet concentration is in the range 10⁻¹ to 1#/cc. Similarly, from Table 3, the range of

- mean (median) values of BSR is noticed to be from 1.6 (1.4) to 17.2 (17.5) and 12.2 (8.7) to 318 (313) at 455 and 940 nm. 654—respectively. Therefore, it is difficult to arrive at threshold values of BSR for ice clouds based on Table 3. This may be partly due to the fact that because BSR depends not only on the particle number concentration but also the size. However, it is interesting to 656—note (except for a few cases in Table 3) that BSR data of ice clouds (at both channels) tend to be greater for densely populated 657—clouds. On the other hand, the difference between mean and median values of CI is not large, thus not much variance in CI 658— within the ice cloud. It is also clear from Table 3 and Fig. 11(a) that about 90-95 percentile of CI values of ice clouds are 659 above 15 and below 25 with mean/median values in the range 18-20. The same is also seen in the histogram of CI shown — (Fig. 11b) in different colors colours for different sounding dates where a greater number of points in a sounding are lying close to 661 20. Therefore, it may be concluded that the mean value of CI of ice clouds would be between 18 and 20. The data from 8 soundings are also analysed for CI (and other parameters) of liquid clouds. However, it is noted 663—that liquid clouds were not observed as often as ice eloudclouds in the balloon data. In the second campaign (8 July 2017) a liquid 664—cloud layer was observed at altitudealtitudes from 4.7 to 4.86 km (160 m) with RH>SRH, temperatures in the range of -0.4 to -1.65°C. — The mean value of CI corresponding to this liquid cloud layer is very high around 50. Similarly, another liquid cloud layer — was observed in the fourth campaign (01 Nov. 2018) in the altitude range of 2-2.3 km (300 m). The corresponding CI values are high and above 100 (up to 200). Couple A couple of thin super-cooled liquid cloud layers were also identified on the same 668 — sounding between 6.1-6.17 km (7 m) and 6.6-6.8 km (200 m). The corresponding CI values are found with mean (median) 669 values of 19.5 (19.4) and 32.6 (32.8), respectively. Apart from this, a strong boundary layer (liquid) cloud layer was 670 — observed on 23 Mar. 2019 (fifth campaign) between 0.9 and 1.2 km (300 m). The corresponding CI of liquid cloud was — found to be high with mean, and median values of 60-80. From the above discussion (including the liquid cloud cases not 672 discussed above), it is noticed that the CI for liquid clouds is high. The difference in CI values of liquid clouds can be attributed to the thickness of the cloud, and the density and droplet size of liquid clouds.
 - 673 attributed to the thickness of cloud, density and droplet size of liquid clouds.
 - Non-spherical large dust aerosol particles were identified by DOP values from CPS in the lower troposphere where 675—RH is far less than 100%. Statistics on COBLAD CI (and other parameters) for these non-spherical particle cases are

676 __presented in Table 4 using the data from 8 soundings. For example, a non-spherical particle layer was found between 0.5 and 677 __2.5 km altitudes on 06 June 2017 with temperatures in the range of 15.5 to 27.6 C and relative humidity is dry from 63.5 to

678—81.3%. The mean (median) value of CI corresponding to this non-spherical particle layer is 12.3 (12.5), BSR is 1.45 (1.4) at 679—455 nm, 6.5(6) at 940 nm and peak particle concentration is between 10⁻³ and 10⁻¹ #/cm³. The peak particle concentration of 680—all non-spherical layers is found to be in the same range and hence not shown. From Table 4, it can be noticed that the non-681—spherical particle (aerosol) layer is found from the near—surface to the 5 km altitude depending on the month or season. During 682—the monsoon season (font in blue colorcolour in Table 4), non-spherical particle layers were observed mostly from the near—surface 683—(500m) to 2.5 km whereas during pre-monsoon (font in wine red colorcolour) it is found from 0.5 up to 5 km. The reason for 684—the difference in layer thickness among seasons may be attributed to the mixing within the lower troposphere, long-range transport 685—and local sources. Since these layers are confined mostly to the lower troposphere, the temperatures are in the range of 27 to 686—below 0°C. From the above statistics (pre-monsoon and monsoon cases) it may be stated that the mean/median value of CI for the non-spherical particle layer is distributed between 11 and 15, irrespective of environmental humidity and season.

688 BSR values for non-spherical layer are between 1.4 and 3.5 at 455 nm, whereas little spread in red channel.

3.4. Illustration of the aerosol-cloud relationship

In this section, an attempt is made to demonstrate the method to identify the relationship, if any, between aerosol and cloud properties observed using balloon observations of the BACIS campaigns. In the present analysis we have restricted ourselves to only liquid or low level clouds as aerosol interactions in these cloud categories are well established (Bruce A. Albrecht, 1989; Twomey, 1977).

In this section, an attempt is made to demonstrate the method to identify the relationship, if any, between aerosol and cloud properties observed using balloon observations of the BACIS campaigns. In the present analysis, we have restricted ourselves to only liquid or low-level clouds as aerosol interactions in these cloud categories are well established (Bruce A. Albrecht, 1989; Twomey, 1977).

The scheme (discussed in sec. 2) is applied to the 15 balloon soundings of the BACIS campaigns and 6 launches

have been observed with low-level cloud and aerosol layers. Further, a scatter plot between logarithm values of the median

cloud particle count of the cloud layer and logarithm of median values of aerosol (blue) back scatter backscatter below cloud base (for 300,

697—400 and 500m) is plotted in Fig. 12. A linear fit (line) of log-log values is also shown separately for all depths. It is noticed

- 698 __for depths 100 and 200m below the cloud base relationship between aerosol, and the cloud cannot be discussed due to a lack of data points
- 699—of aerosol backscatter ratio from individual campaigns. This could be the result of the elimination of the high value of COBALD
- 700 __particle back scatterbackscatter (>3.15) observed in this region (100 and 200m below cloud base). In the cloud boundaries of about 100
- 701 __and 200m below the cloud base, an intermediate region exists where aerosol transformation to cloud particle/growth takes place.
- 702 Hence it is tricky to have the aerosol observation in this region. On the other hand, with similar elimination criteria (Section 2),

aerosol backscatter could be obtained (from all 5 campaigns) for depths 300m onwards (up to 500m) from eloud base. Athe cloud base. A good positive relationship is found between aerosol backscatter and cloud particle count with a statistically significant Pearson correlation coefficient of about 0.9 and slope (ACI index) of 0.77 and 0.86 when the aerosol is considered from 300 and 400m below the cloud base, respectively. For a depth of 500m from the cloud base, the slope has decreased to 0.67 (correlation coefficient is also not significant with p-value >0.05) indicating aerosol influence weakens if the region below 400m from the cloud base is considered. Therefore, it may be better to consider aerosols up to a depth of 400m (below the cloud base) for understanding their influence on cloud properties. It is also emphasized that the slope (ACI index) value obtained in this analysis at all depths is well within the theoretical range of 0 to 1. However, with a greater number of balloon soundings, it might be possible to have statistically significant aerosol data after constraining similar background/meteorological conditions to delineate their possible effects. Data obtained on 04 February 2020 was not considered in the analysis due to the high values of COBALD. The individual uncertainties in BSRb and Nc were assumed to be 5% and the combined uncertainty in the ACI index is estimated as discussed in Sec. 2.4.7 (equation.7). It is found that the combined uncertainty in the estimated ACI index is found from 0.01 to 0.23 and 0.08 to 0.13, respectively for particle backscatter data from 300 and 400m below cloud base.

Pearson correlation coefficient of about 0.9 and slope (ACI index) of 0.77 and 0.86 when aerosol is considered from 300 and 400m below cloud base, respectively. For depth of 500m from cloud base, the slope has decreased to 0.67 (correlation coefficient is also not significant with p-value >0.05) indicates aerosol influence weakens if region below 400m from cloud base is considered. Therefore, it may be better to consider aerosol up to a depth of 400m (below the cloud base) for understanding their influence on cloud properties. It is also emphasized that the slope (ACI index) value obtained in this analysis at all depths are well within the theoretical range of 0 to 1. However, with a greater number of balloon soundings it might be possible to have statistically significant aerosol data after constraining the similar background/meteorological conditions to delineate their possible effects. Sounding from (with a low level cloud layer) 04 February 2020 is ignored due to high values of COBALD return signal possibly due to an optical interference. The individual uncertainties in BSRb and Ne were assumed to be 5% and the combined uncertainty in ACI index is estimated as discussed in Sec. 2.4.7 (equation.7). It is found that the combined uncertainty in estimated ACI index is found from 0.01 to 0.23 and 0.08 to 0.13, respectively for particle backscatter data from 300 and 400m below cloud base.

4. Summary

- The BACIS (Balloon-borne Aerosol Cloud Interaction Studies) field campaigns have been conceptualized and
- 719—successfully conducted using multiple-instruments from Gadanki (13.45°N; 79.2° E), a location in Southern peninsular
- 720 _India. Meteorological balloon payloadpayloads with a combination of lightweight and specialized sondes such as COBALD and CPS
- 721—have been launched for the first time prior tobefore a CALIPSO satellite overpass (close by Gadanki). Ground-based Lidars
- 722 (MPL/Ceilometer/Mie lidar), and Radars (MST Radar/LAWP) were also operated during the campaign period. So far 15 balloon soundings have been conducted as part of the BACIS campaigns.
- 724 ——During the first two (pilot) campaigns, all essential ground—based and space—borne instruments were made available.
- 725 Consistency in balloon_Balloon_borne in-situ measurements (CPS and COBALD) is are assessed using the data from ground/space borne
- 726—spaceborne remote sensing instruments (CALIPSO, MPL and a Mie lidar) from two pilot campaigns (early hours of 6 June and 8 July
- 727—2017). The comparison shows a goodreasonable agreement within in-situ measurements as well as between ground-based/space_borne

728—and in-situ measurements. It is observed that the in-situ balloon soundings using a combination of specialized (COBALD and 729 CPS) sondes adds to the cloud and aerosol information than can be obtained from an individual ground/space bornespaceborne instrument.

730 instrument.

- In order to ______ To discriminate aerosol from clouds in a profile, combined observations of COBALD and CPS from a
 campaign held on 27 June 2019 were inferred in detail. Using CPS data, liquid, super cooled, supercooled, and ice clouds were identified.
 COBALD data of BSR corresponding to the ice clouds was found to be 1-10 (at blue channel) and CI of 10 to 20. In addition
 to cloud features, CPS has also detected cloud particle layers at low altitudes (under dry conditions). These layers may be regarded as non-spherical (coarse mode) aerosol particle layers as ice clouds (with non-spherical cloud particles) cannot exist at lower eights. An attempt is also made to infer the size of cloud particles using the CPS data of intensity of scattered light (I55) and the COBALD
- heights. An attempt is also made to infer the size of cloud particles using the CPS data of intensity of scattered light (I55) and the COBALD colour index. Based on CPS scattered light, the liquid droplet size (for the above case) is estimated to be 2-14 µm, and for ice particles, it is a combination of particles with 80-140 µm and 2-14 µm. The estimates of ice particle sizes using CI data from COBALD supported the size interpretations of ice particles by CPS.
 - at lower heights. An attempt is also made to infer the size of cloud particle using the CPS data of intensity of scattered light 737—(I55) and the COBALD color index. Based on CPS scattered light, the liquid droplet size (for the above case) is estimated to 738—be 2-14 µm, and for ice particles it is combination of particles with 80-140 µm and 2-14 µm. The estimates of ice particle 739—sizes using CI data from COBALD supported the size interpretations of ice particles by CPS.
 - - 21.4 to 3.5, respectively. These non-spherical particle layers may correspond to coarse mode (dust) aerosols as discussed.

— (aerosol) layers (in the lower troposphere), the mean values of CI and BSR (at 455 nm) are found to be between 11 to 15 and

The relationship between aerosol and cloud in low-level (liquid) eloudclouds is illustrated using balloon data from BACIS

748—campaigns. CPS cloud particle count and COBALD particle backscatter at the blue channel were considered as cloud and

749—aerosol proxies, respectively. A scheme is developed to carefully identify the cloud layers from CPS data and particle

750—(aerosol) backscatter below the cloud from COBALD data (in a profile). However, the relationships were analyzed

751—separately using particle backscatter data from 100 to 500m below the base height for the first cloud layer. The results show;

752—a statistically significant correlation of 0.9 and a slope (Aerosol-Cloud Interaction index, ACI) of 0.7 (0.86) obtained between

particle backscatter from 300m (500m) below the cloud base and the corresponding cloud particle count. The ACI index The ACI index value obtained is well within the theoretical limits of 0 to 1 indicative of the aerosol activation process of the cloud. The uncertainty in the estimated value of the ACI index is 0.01 to 0.23 and 0.08 to 0.13, respectively for backscatter data from 300 and 400m below the cloud base.

value obtained are well within the theoretical limits of 0 to 1 indicative of aerosol activation process of cloud. The uncertainty in the estimated value of ACI index is 0.01 to 0.23 and 0.08 to 0.13, respectively for back scatter data from 300 and 400m below the cloud base. -Statistical estimates/threshold value of CI, BSR for cloud (liquid/super-cooled/ice) and non-spherical particles 758— attempted here will greatly help to separate a COBALD profile with respect to for aerosol and cloud. However, immediate 759 efforts are needed to understand the portion of the COBALD profile with no cloud detection from CPS. This portion of 760—the COBALD profile may correspond to either aerosol with fine mode particles and/or a thin cloud not detectable by a CPS. On 761— the other hand, estimates of size discussed here (from CPS, COBALD) are purely based on Mie theory and laboratory data. - However, with assumptions of the log-normal distribution of particles and measurements from COBALD (BSR, CI), the theoretical 763 — estimate of the particle size distribution of aerosol/cloud is possible. It makes sense to cross-check rough estimates of size from a 764 CPS with COBALD size distributions rather than using CI variations. It is also planned to add a size distribution 765— measurement to the balloon payload for cross-verification and validation. Apart from this, in some of the cases, we have 766 noticed COBALD return signal saturated for liquid/super-cooled cloud in the presence of a thick liquid cloud. Hence the 767 information from a greater number of future launches will help to conclude the statistical figures/threshold values for liquid - clouds as well as other cases of clouds, to discriminate the aerosol/cloud in a profile and to better quantify the aerosol-cloud 769 relationship. Further to this, attempts will be made to quantify aerosol-cloud interactions (with the multi-instrument data), particularly the role of vertical wind and turbulence on the aerosol-cloud interactions, and ice cloud interactions, among others. In a nutshell, the results presented in the study indeed demonstrate the potential of the observational approach/method, to understand the aerosol-cloud process.

770 particularly the role of vertical wind and turbulence on the aerosol cloud interactions, ice cloud interactions, among others.

- 771 In a nutshell, the results presented in the study indeed demonstrate the potential of observational approach/method, paves the
- 772 way for future campaigns to understand aerosol cloud process.
- Code/Data Availability
- Data analyzed in the study is made available on Zenodo (10.5281/zenodo.5749293). Data
- will also be shared with the interested users under the collaboration.
 - 775 with the interested users upon request and under collaboration.
 - Author Contribution

- RKV is responsible for Conceptualization, Conducting <u>experimentexperiments</u>, Formal analysis, Visualization, Investigation,
- 778—Writing-original draft preparation; VRM is responsible for Supervision, helpedhelping in Visualization, Writing-review and editing.
- 779—FM, HR and FGW are responsible for Writing-review and editing; MBL, RRM, and RN helped in Visualization, Writing-review and editing; RN, ARST, HKA, and RBS helped in conducting the experiment, Writing-review and editing;

781 Competing interest

The authors declare that they have no conflict of interest. 783

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Table 1. List of instruments deployed (in BACIS) and the corresponding physical parameters obtained.

Sl.	Instrument	Purpose	Physical quantity (Unit)
No.	CALIPSO	Aerosol and cloud profiling	Total attenuated backscatter(km ⁻¹ sr ⁻¹)
2	MPL	Aerosol and cloud profiling	Backscatter coefficient(m ⁻¹ sr ⁻¹)
3	Mie Lidar	Aerosol and cloud profiling	Backscatter coefficient(km ⁻¹ sr ⁻¹)
ļ	COBALD	In-situ measurement of aerosol and cloud particles	Backscatter ratio
5	CPS	In situ measurement of cloud particles	Cloud particle number concentration(#/cc), degree of _polarization(DOP)
5	MST Radar	3-D Wind components, turbulence	Horizontal and vertical wind _components(m/s)
7	LAWP	3-D Wind components, turbulence	Horizontal and vertical wind components (m/s)
3	MWR	Meteorological parameters and cloud	Temperature(⁰ C), RH(%) and cloud liquid water content(g/m ³)
)	ICON	Ambient aerosol	BC concentration (µg/m³), Scattering coefficient and absorption coefficient (m¹)
10	Ceilometer	Boundary layer, cloud and aerosol	Back scatterBackscatter coefficient(km ⁻¹ sr ⁻¹)
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Table. 2. Date and time of <u>the BACIS</u> campaigns and the instruments operated during the corresponding <u>eampaigns</u>.

S. No.	Date & Time (LT)	MPL	Mie	Ceil	CPS	COB	MST	MWR	Aeth	CALI	LAWP
1	06-06-2017;	Y	Y	N	Y	Y	Y	N	Y	Y	Y
	01:57										
2	08-07-2017;	Y	Y	N	Y	Y	N	N	Y	Y	Y
	01:36										
3	29-09-2018;	Y	Y	N	Y	Y	N	N	Y	N	Y
	01:46										
4	01-11-2018;	N	Y	N	Y	Y	N	N	Y	N	Y
	22:13										
5	23-03-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	02:36										
6	30-04-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	23:16										

7	30-05-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	23:46										
8	27-06-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	23:45										
9	28-08-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	23:42										
10	09-10-2019;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	23:36										
11	20-12-2019;	N	Y	Y	Y	Y	Y	Y	Y	N	Y
	21:20										
12	04-02-2020;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	00:27										
13	10-03-2020;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	00:26										
14	19-06-2020;	N	Y	Y	Y	Y	Y	Y	Y	N	Y
	23:26										
15	19-08-2020;	N	Y	Y	Y	Y	Y	N	Y	N	Y
	22:39										

MPL – Micro Pulse Lidar; Mie – Mie Lidar; Ceil – Ceilometer; CPS – Cloud Particle Sensor (CPS);

COB – Compact Optical Backscatter AerosoL Detector (COBALD); MST – Indian MST Radar; LAWP – Lower Atmospheric Wind Profiler (LAWP); Aeth – Aethalometer; CALI – Calipso; MWR – Micro Wave Radiometer.

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our
Inde x (CI) and other phys ical para mete rs of the ice clou ds. Back scatt er<u>Th</u> e back scatt er ratio (BS R) in norm al (Itali c) font is for <u>a</u>450 nm (940 nm) chan

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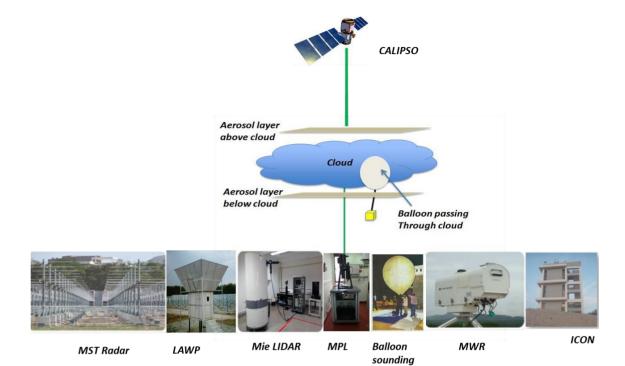
							_conc. (#/cc)
06-Jun-2017	1	13- 15.5	-53 to -74	~ SRH	19.2 _(19.2)	5.6(4.8) 90.4(73)	10 ⁻² to 10 ⁻¹
08-Jul-2017	2	10.5-16	-34 to -78	> SRH	18.7(18.6)	3(2.9) 37.5(35.2)	10 ⁻² to 10 ⁻¹
01-Nov-2018	4	12-12.6	-47 to -53	> SRH	19.5	17.2(17.5) 318(313.5)	10 ⁻¹ to 1
30-Apr-2019	6	9.3-16	-22 to -79	~SRH	19.4(19.3)	16.4(8.6) 302(147)	10 ⁻¹ to 1
30-May-2019	7	16.2- 17.4	-78 to -84.5	<srh< td=""><td>18</td><td>1.6(1.4) 12.2(8.7)</td><td>10⁻³ to 10⁻²</td></srh<>	18	1.6(1.4) 12.2(8.7)	10 ⁻³ to 10 ⁻²
27-Jun-2019	8	9.4-10.7	-23.7 to -35.2	>SRH	19.3(17.9)	5.1(3.1) 74.8(43.2)	10 ⁻¹ to 1
19-Jun-2020	14	14.2- 15.4	-62 to -75	<srh< td=""><td>21</td><td>7.9(7.9) 147.4(143.2)</td><td>10⁻¹ to 1</td></srh<>	21	7.9(7.9) 147.4(143.2)	10 ⁻¹ to 1

Date	Campaign no	Ice cloud altitude	Temperatur eTemperatu re range (°C)	RH condition	Mean (median) CI	Mean (median) BSR	Range— of _peak—
		(km)					ice
							particle
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e. The BSR in the normal (italic) font is 450 nm (940 nm). Blue (red) colour values are observed in the monsoon (pre-monsoon) months.

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Campaign Date	Non-	Temperature range (⁰ C)	RH	Mean	Mean
Date	spheri cal	range (°C)	range (%)	(median) CI	(median) BSR
	layer		(70)		
	altitude				
	(km)				
06-Jun-2017	0.5-2.5	27.6 to 15.5	63.5-81.3	12.3(12.5)	1.45(1.4)
					6.5(6)
08-Jul-2017	0.5-2.5	25.3 to 14.7	64.2-96.4	14.6(14.8)	2
					15.8
29-Sep-2018	0.5-1	22.6 to 20	92-94	12.3	3.3(3.2)
					30(29)
27-Jun-2019	0.5-1.5	27.6 to 19.8	57.3-70.3	11.4	1.6
					7.6
19-Jun-2020	0.5-2.5	28.8 to 14.2	57.2-94.4	12.6(12.8)	1.6
					8(8.1)
23-Mar-2019	1.5-3.5	23 to 6.5	32.7-70.3	12.6(12.8)	2
					13
30-Apr-2019	0.5-4	28 to 4.5	60.2-97.3	12.2(12.6)	3.3(2.6)
•					28(21.5)
30-May-2019	0.5-5	28.8 to -0.1	60-98	11.7(11.6)	3.2(2.9)
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d Interaction Studies (BACIS) campaign.



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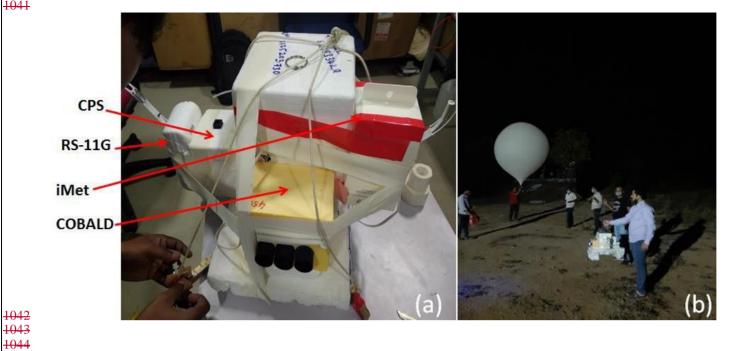
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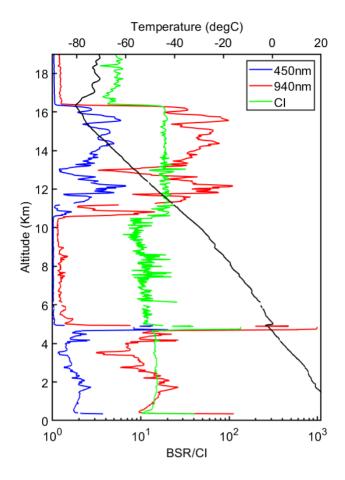
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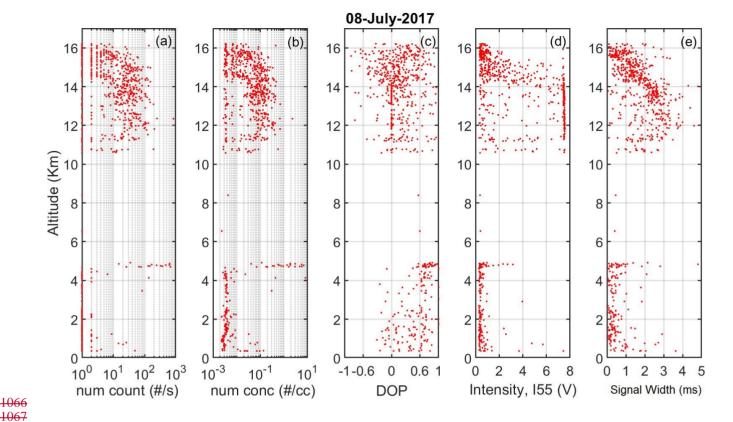
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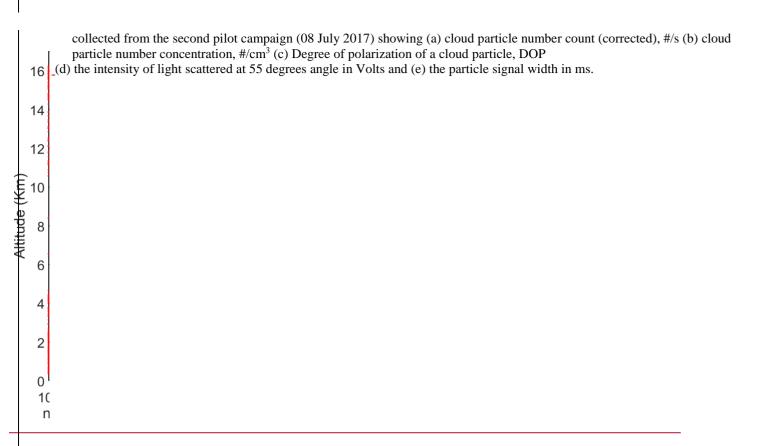
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nm) channels were obtained using a COBALD sonde launched during the second pilot campaign (08 July 2017). Color Colour Index (CI) estimated from BSR at both channels is also shown (in green colorcolour). 18 16 14 6 4 2 0 1 Figure 3. Back scatt er ratio (BS

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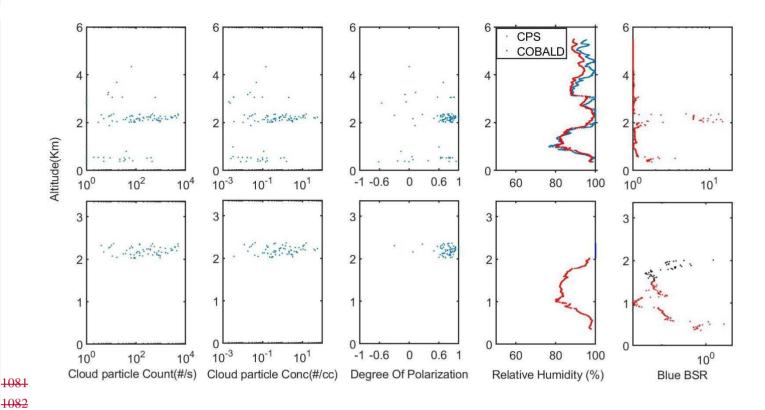




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	6	sounding held on 01 November 2018 up to the altitude of 6 km (as $\underline{\text{the}}$ focus is on $\underline{\text{the}}$ liquid cloud region). $\underline{\text{Bottom}}$ panel
		shows the same parameters but for the portion of the same profile where liquid cloud (blue dots) and aerosol (from cloud base to 500
	4	m below) were identified by the scheme.
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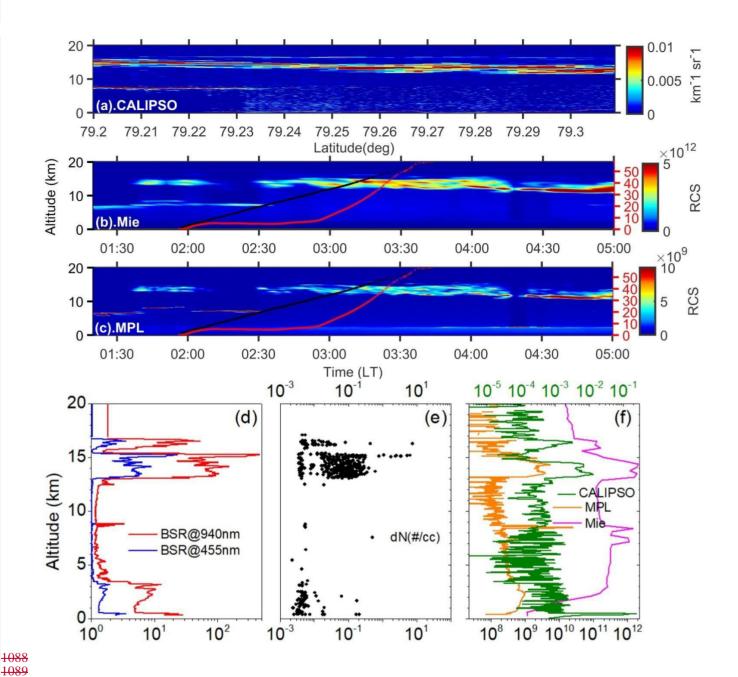
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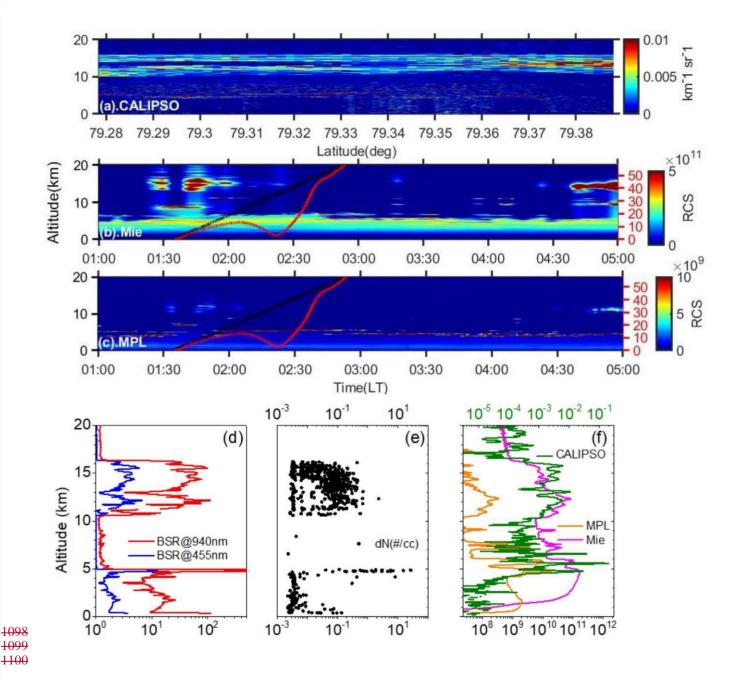
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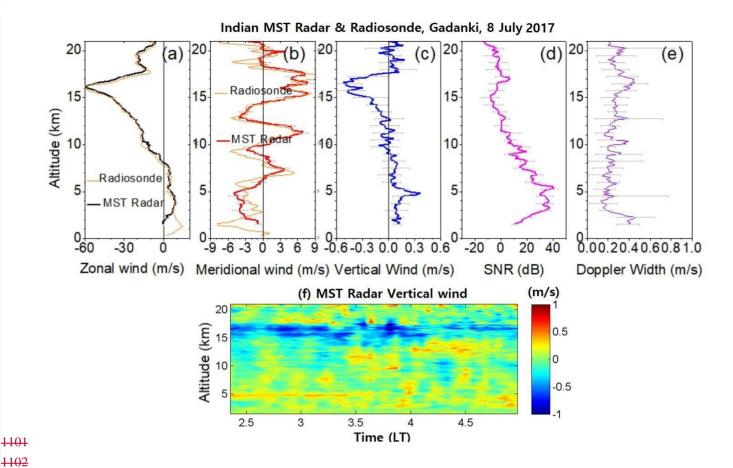


balloon sounding held on the early hours of 06 June 2017. The total attenuated back scatter backscatter from (a) CALIPSO and temporal variation in range corrected signal from (b) Mie lidar and (c) MPL. The red (black) lines over plotted on contour maps (b) and (c) represent balloon drift (altitude) in km with time. Drift as a function of time can be read with the right y-axis (red font) and altitude as a function of time can be read with the left y-axis. The profiles of BSR at two channels from COBALD (blue and red-coloured lines), particle number concentration from CPS (black coloured dots), RCS from MPL (orange), Mie lidar (magenta) and total attenuated back scatter backscatter from CALIPSO (olive green) lines shown in (d), (e) and (f) respectively. 20 10 0 20 10 0 20 Altitude (km) Figure 6. Mult iinstr ume nt

Altitude (km)

data from <u>a</u>





Noise Ratio (SNR) and (e) Doppler Width <u>were</u> obtained from Indian MST radar <u>duringon</u> 8 July 2017 averaged during 02:30 LT to 03:30 LT. Horizontal bars show standard deviation. Radiosonde observed zonal and meridional winds are also superimposed in the respective panels. (f) Time-altitude section of vertical wind obtained from Indian MST radar during the radiosonde launch time.

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Figure 8.

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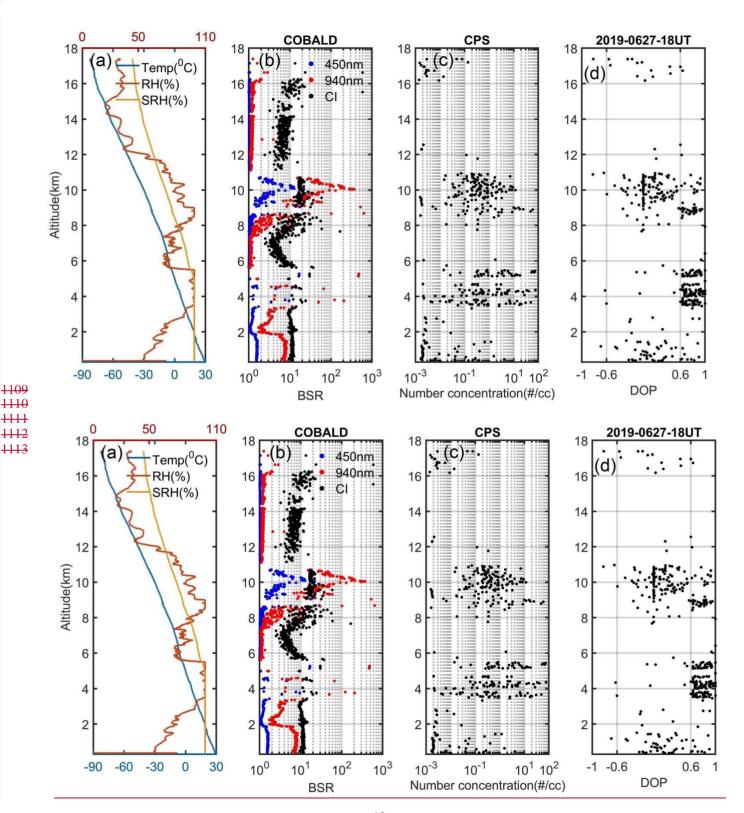


Figure 9. Com bine d obse rvati ons of COB ALD and **CPS** from ballo on soun ding were held on 27 June 2019 at 2330 LT. (a) Tem perat ure (T), Relat ive humi dity (RH) and Satur ation Relat ive Hum idity (SR H) (b) Back scatt

> er ratio

at 455 nm (blue), 940 nm (red) and ColorColour Index (Black). (c) cloudCloud particle number concentration and (d) Degree of polarization (DOP).



1119

1120

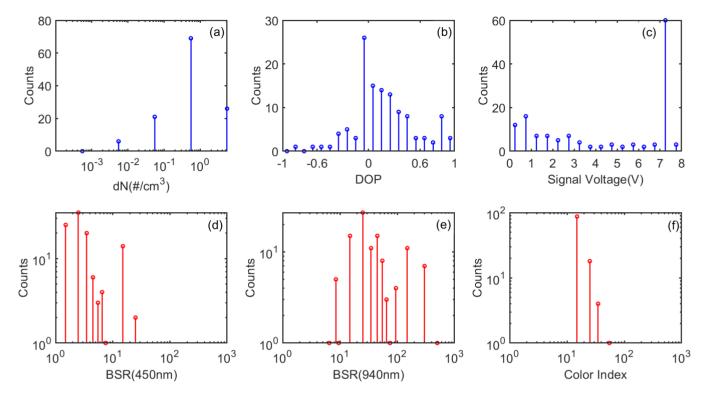
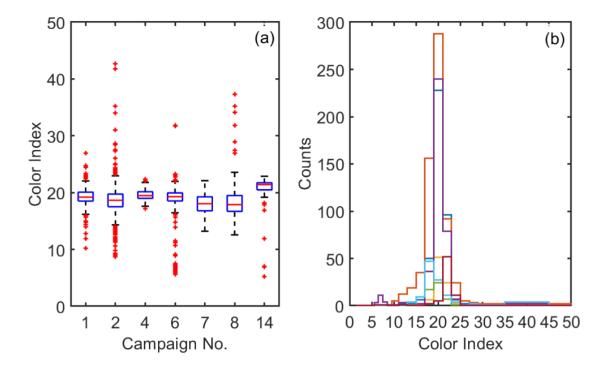


Figure 10. Histogram of (a) Droplet number concentration (dN) in #/cc (b) Degree of polarization (DOP) (c) Backscattered signal (Volts) (d) Backscatter ratio at 455 nm, (e) Backscatter ratio at 940 nm and (f) Color Index. The top panel shows the data from CPS and the bottom panel from COBALD for the ice cloud layer between 9 and 11 km from the sounding held on 27 June 2019.





	P) (c) Backscattered signal (Volts) (d) Backscatter ratio at 455 nm, (e) Backscatter ratio at 940 nm and (f) Colour Index.	The top
	panel shows the data from CPS and the bottom panel from COBALD for the ice cloud layer between 9 and 11 km from the	sounding
80	neid on 27 June 2017.	
60		
Counts 0		
20		
0		
ഇ 10 ¹		
Counts		
10 ⁰		
10		
Figure 10.		
<u>Hist</u>		
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(dN) in #/cc (b) Degr ee of polar izati		
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<u>on</u> (DO		

Index (CI) <u>was</u> observed for the ice clouds found in different campaigns. <u>The horizontal line in the centre of the box represents</u> the median. The upper and lower edges of the box represent the third quartile (Q3), and first quartile (Q1) respectively. <u>Similarly, the upper and lower whiskers represent Q3+1.5*(Q3-Q1) and Q1-1.5*(Q3-Q1).</u> The data points beyond the whiskers (outliers) are <u>shown with red star symbols.</u> (b) The histogram of the CI values from each campaign. <u>Different colours indicate the data from different campaigns.</u>

1	112 9
2	1130
3	1131
4	1132
5	1133
6	1134
7	1135
8	

-3 -2 -1 0 log of aerosol particle BSR(450nm)

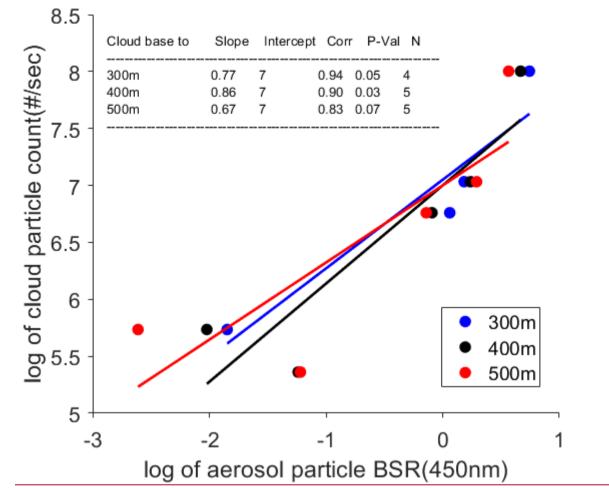


Figure 12. Scatter between logarithm values of COBALD median aerosol blue back scatter backscatter (x-axis) from 100, 200, 300, 400 and 500 meters below the cloud base and the corresponding CPS median cloud particle count (y-axis) obtained from five balloon soundings, with a linear fit (different colored lines). Table The table inside shows the detailed statistics.
