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CCN closure results from Indian Continental Tropical Convergence Zone (CTCZ) aircraft experiment

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ABSTRACT

In-situ aircraft measurements of cloud condensation nuclei (CCN) and aerosol size distribution were carried out over the region spanning from 24.78 °N to 29.5 °N and 78.1 °E to 85.0 °E from June 29 to July 3, 2009 during the Indian Continental Tropical Convergence Zone (CTCZ) campaign, consisting of total 9 flight sorties. CCN measurements were conducted at a constant supersaturation (SS) of 0.84%. It was found that at higher altitudes (4.7–6.7 km), 30 nm sized particles were dominating while at lower altitudes (0.6–3.7 km), 50 nm particles. Overall, CCN closure ratio ($CCN_{predicted}/CCN_{measured}$) at different altitudes using Köhler theory and assuming pure ammonium sulfate was 1.375 ($R^2 = 0.80$). For each sortie, the closure ratio varied with height and depended greatly on measured CN concentrations. A case study of Khajuraho sortie showed that the closure ratio increased from 1.07 to 1.40 as the CCN concentration increased from 1000 cm^{-3} to 4000 cm^{-3}. Results of CCN closure improved significantly (overprediction improved by 37.5% and 34.6% for Pantnagar and Gaya, respectively) with the assumption of internally mixed aerosols composed of ammonium sulfate and insoluble organics. Hygroscopicity parameter calculated for these two sorties ($\kappa = 0.51$ and 0.5) indicates the presence of moderately hygroscopic organic species along with some inorganic content.

1. Introduction

Aerosols are defined as fine solid or liquid particles suspended in gas which originated from natural sources viz. windborne dust, sea spray and anthropogenic activities such as combustion of fossil fuels, with size ranging from 0.001 μm to 100 μm. The particles which have sufficient soluble mass to grow or activate into cloud droplets at atmospheric supersaturation are termed as the cloud condensation nuclei (CCN). Higher concentration of CCN leads to formation of droplets of smaller mean diameter, which increases the cloud reflectivity and hence cooling effect (Twomey, 1977). The second indirect effect is related to the inhibition of precipitation in clouds with small mean droplet diameters, which affects the extent and lifetime of clouds (Albrecht, 1989). The uncertainty in estimating the indirect radiative forcing is due to incomplete description of spatial and vertical distributions of anthropogenic aerosols (Asa-Awuku et al., 2011).

Physico-chemical processes involved in the activation of particles into CCN are governed by the droplet curvature and solute effects on the water vapor pressure. The Köhler theory, composed of both these competing effects, is the theoretical basis to predict CCN activity (Cantrell et al., 2001). It determines the "critical" supersaturation (expressed in percentage, %), S_c, required by a particle to behave as a CCN and finally activate into cloud droplets. For a given aerosol particle, S_c depends on the dry diameter and chemical composition. Earlier simplified forms of Köhler theory include completely insoluble and non-hygroscopic species (i.e. organics), and completely soluble fraction, usually ammonium sulfate ($(NH_4)_2SO_4$) or sodium chloride (NaCl). Problem arises when water soluble organic carbon (WSOC) is present in the atmosphere originating from either biomass burning or SOA formation (Petters and Kreidenweis, 2007).

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Extended Köhler theory closed this gap by introducing the effect of WSOCs as a solute and their role in surface tension of cloud droplets (Wang et al., 2010). Until to this date, Köhler theory is used to predict CCN concentration on the basis of aerosol size distribution, chemical composition and dynamical forcing. Comparison of modeled CCN with the observed CCN concentration is the ultimate test of Köhler theory (Asa-Awuku et al., 2011).

In the past, several ground based and aircraft based CCN closure studies have been performed with varying degrees of success. CCN measurements (SS = 0.4%) using static diffusion cloud chamber were performed at Chebogue Point during North Atlantic Regional Experiment (NARE) (Liu et al., 1996). It was found that aerosol composition from filter samples and size distributions from passive cavity aerosol spectrometer probe (PCASP-100X) results in closure for the 75% of time. Most of the closure studies carried out by neglecting the effect of WSOC constituents on CCN activity overestimated the CCN concentration on an average by 30–50% which ultimately results in 20–40% uncertainty in aerosol indirect climate forcing (Broekhuizen et al., 2006; Dusek et al., 2003; Stroud et al., 2007). Several other ground based closure studies using filter based chemical composition and aerosol mass spectrometer (AMS) data derived that size averaged chemical data overpredicted CCN concentration to an extent of 21 ± 18% and 35.8 ± 28.5%, respectively, for SS = 0.2%–1.1% (Medina et al., 2007; Patidar et al., 2012). Few recent studies have come up with a finding that assumption of internal mixing state of aerosols and size averaged chemical composition leads to 2–36% CCN prediction bias (Cubison et al., 2008).

Assuming pure ammonium sulfate, i.e. mainly of marine origin, VanReken et al. (2003)achieved CCN closure ratio of 1.047 and 1.241 at 0.2% and 0.85% SS, respectively using two continuous-flow stream wise temperature gradient chambers, during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus (CRYSTAL-FACE) aircraft experiment. Rissman et al. (2006) making similar assumption also achieved closure ratio ranging from 1.18 to 3.68 during Atmospheric Radiation Measurement (ARM) program in May 2003 in Lamont, Oklahoma. They concluded that aerosols possibly were less hygroscopic than assumed. Table 1 summarizes the results of several aircraft-based closure studies. The effective hygroscopicity parameter (κ) provides a link between dry particle diameter and CCN activity which explains the effect of chemical composition of aerosols (Petters and Kreidenweis, 2007). For a multi-component system, the effective hygroscopicity parameter can be calculated by volume weighted average of κ of different components, referred as a simple mixing rule. Typical values of hygroscopicity parameter for ambient aerosols range from 0.1 to 0.5 (Asa-Awuku et al., 2011). Researchers have shown that considering the effect of soluble organic fraction and hygroscopicity improves the closure with deviation < 20% (Rose et al., 2010; Gunthe et al., 2009).

This study aims to understand the vertical and spatial variation of key aerosol parameters. Size distribution, chemical composition of the aerosols, and their activation properties are the major factors affecting CCN activity, depending on the type of the air mass being sampled which varied with the altitude. In-situ aircraft observations were carried out during the early monsoon season (June 29–July 3) of year 2009 across the Indian Continental Tropical Convergence Zone region (CTCZ). CCN closure analysis was performed using measured CCN by the CCN counter and the predicted CCN utilizing size distribution measured by SMPS and chemical composition from PM_1 samples collected during the flight sorties, assuming submicron aerosols as pure ammonium sulfate.

2. Methods

2.1. Aircraft experiment

Airborne measurements were carried out across the Indian CTCZ region, characterized by intense convergence over South Asia producing large scale precipitation in the monsoon season. Multi-level measurements from surface to 5600 m altitude were carried out for nine different locations. CCN measurements obtained for only six sorties viz., Kanpur, Nainital, Pantnagar, Khajuraho and Gorakhpur, are discussed here. The details of the aircraft used, sorties, and instruments on board are summarized in Jai Devi et al. (2011). A key challenge of in-situ aircraft aerosol measurements is to collect aerosol samples that represent ambient concentration as closely as possible. To achieve it, aerosols must be efficiently sampled without losses in the sampling probe, which requires isokinetic flow. The aircraft was connected to a shrouded community inlet to minimize non-isokinetic losses mainly for particles greater than 1 μm. The shrouded probe used in the experiment decelerates the air before it enters the probe resulting in better airflow characteristics near the probe inlet, which ensures transmission efficiency of 95% for particles within diameter range of 0.01–5 μm (Baron and Willeke, 2001). In the aircraft experiment, the droplet measurement technology (DMT) CCN counter was used to measure the CCN concentrations at a constant SS = 0.84%. The scanning mobility particle sizer (TSI3936) in combination with butanol condensation particle counter (CPC, TSI 3775) was used for measuring the aerosol size

Table 1
Summary of previous closure studies based on aircraft measurements.

Reference	Aerosol chemical composition	Name of campaign	CCN activity of organics	$CCN_{predicted}/CCN_{measured}$
Chuang et al. (2000)	Ammonium sulfate	ACE-2	Inactive	1.12–11.11
VanReken et al. (2003)	Ammonium sulfate	CRYSTAL FACE	Inactive	1.047 at 0.2% SS 1.201at 0.85% SS
Snider et al. (2003)	NaCl, sulfate, nitrate ammonium, and organic acid anions	ACE-2	Active	30%
Conant et al. (2004)	Ammonium sulfate and organic carbon	CRYSTAL FACE	Active	20%
Rissman et al. (2006)	Ammonium sulfate	ARM Aerosol	Active	1.18–3.68
Wang et al. (2008)	Sulfate, nitrate and organic carbon	MASE	Active	0.93
Lance et al. (2009)	Ammonium sulfate and organic carbon	GoMACCS	Active	3% to 36%

distribution from 0.016 to 0.57 μm, with a sample flow rate of 0.3 L min^{-1} (Jai Devi et al., 2011).

2.2. Instrumentation

2.2.1. The DMT CCN counter

The DMT CCN counter consists of a cylindrical continuous-flow thermal gradient diffusion chamber. A constant streamwise temperature gradient was applied such that the difference between water vapor mass diffusivity and air thermal diffusivity leads to a quasi-uniform centerline supersaturation. The working principle of the CCN counter is described in detail elsewhere (Roberts and Nenes, 2005; Lance et al., 2006) and experimental setup is provided in Patidar et al. (2012). CCNc was sent for calibration to Droplet Measurement Technologies (DMT), USA and afterwards it was also calibrated at IIT Kanpur laboratory with $(NH_4)_2SO_4$ and NaCl aerosols and was found to be in good agreement with manufacturer's calibration (Patidar et al., 2012). Based on the Köhler theory, the critical dry particle diameter D_{50}, for monodispersed particles at a given SS = 0.84% was computed (Rose et al., 2008).

CCN measurements are subject to large uncertainties due to differences in the counting efficiency of CPC and CCN counter as a result of wall losses and supersaturation depletion mainly for higher aerosol concentration (Rose et al., 2010; Lathem and Nenes, 2011). Details of error budget in CCN measurements are given in Patidar et al. (2012) and also, the aircraft measurements are inherently more difficult than the surface measurements. Correction for depletion in supersaturation is not required for our sampling data as CCN concentrations were less than 5000 cm^{-3} (Lathem and Nenes, 2011).

2.2.2. Constant pressure inlet

DMT constant pressure inlet (CPI) was installed at the inlet of the CCN counter to maintain a constant pressure of 600 mbar corresponding to the highest altitude planned during the experiment. Since, we did not have external recording of the ambient static pressure, following relation is used to convert the aircraft altitude (ft) into the ambient static pressure (mbar) (Jacobson, 1999, 27 pp.).

$$P_d = P_{a,s} * \exp\left\{\frac{g}{\Gamma_s * R} * \ln\left(\left(1-\frac{Z * \Gamma_s}{T_{a,s}}\right)\right)\right\} \quad (1)$$

where Z = altitude in standard atmosphere (m), $T_{a,s}$ = sea level temperature (298 K), $_s$ = environmental lapse rate (6.5 K/km), $P_{a,s}$ = sea level pressure (1013.25 mb), P_d = desired pressure (mbar) at the flying altitude Z, g = gravitational constant, R = universal gas constant (287.04 m^2/s^2 K).

Also, the CCN concentration decreases due to expansion of sample volume owing to reduced pressure. So, the true CCN concentration is obtained by applying the flow correction (Lance et al., 2009).

2.2.3. Scanning mobility particle sizer (SMPS)

Aerosol number-size distribution was measured using SMPS (TSI 3936) in combination with butanol CPC (TSI 3775) in the size range from 0.016 μm to 0.57 μm, with a sample inlet flow of 0.3 L min^{-1}. Additionally, an auxiliary pump, an orifice and a dead volume were used to maintain a constant inlet flow rate in pressurized mode at higher altitudes and during ascend and descend. The particle shape is assumed to be spherical while selecting size in terms of electrical mobility equivalent diameter (Rose et al., 2010). Aerosol number-size distributions were also corrected for multiple charging and diffusional losses of aerosol in the sampling line based on in-built aerosol instrument manager (AIM) software algorithm.

2.3. Data analysis methods

2.3.1. HYSPLIT back trajectory and satellite data

NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php) was used to examine history of air masses sampled during the aircraft measurements (Draxler and Hess, 1998). In addition, the attenuated back scatter signal (ABS) retrieved from Micro-Pulse Lidar (MPL) (http://mplnet.gsfc.nasa.gov/data.html) was also used to study the vertical variation of aerosols and clouds and their influence on CCN profile. The MPL, which is operational at Indian Institute of Technology, Kanpur since May, 2009 is a part of NASA MPLNET network (Spinhirne, 1993; Welton et al., 2001; Misra et al., 2012). Detailed information on MPL system can be found in Misra et al. (2012). In this study, we used level 1.5 retrieved attenuated backscatter signal profiles from 20-min cloud cleared Level 1 signal averages. The uncertainty in the retrieved attenuated backscatter values is less than 5% (Welton et al., 2001). The MPL data provides highly reliable and corroborating information about cloud presence, elevated aerosol layers and the planetary boundary layer (PBL).

2.3.2. PM_1 sampling and chemical analysis

The PM_1 impaction based single staged sampler developed at IIT Kanpur (Gupta et al., 2010, 2011) was installed on the aircraft, which sampled the aerosols during individual sorties. The mass of PM_1 collected on Teflon (Polytetrafluoroethylene (PTFE), Whatman) filters was determined via gravimetric analysis. The chemical composition of PM_1 aerosol mass was determined by analyzing water-soluble ions after extraction with ultrapure Milli-Q water using an ultra-sonicator. The solution obtained was analyzed by ion chromatograph (Metrohm compact, IC-761) for anion (SO_4^{2-}) and cation (NH_4^+) (Chakraborty and Gupta, 2010).

2.3.3. CCN closure

The data used in the CCN closure analysis includes the aerosol size distribution measured by SMPS and aerosol chemical composition obtained from the PM_1 filter sampling. The CCN concentrations measured ($CCN_{measured}$) with a frequency of 1 s were averaged corresponding to each size distribution measured by SMPS (i.e. 90 s). The predicted CCN concentrations ($CCN_{predicted}$) were obtained from the size distributions and chemical composition of aerosols. The equation based on Köhler theory was used to determine the CCN activation diameter at a given SS($\equiv S_c$) which is given as,

$$S_c = \left[\frac{256}{27}\left(\frac{M_w\sigma}{RT\rho_w}\right)^3\left(\frac{M_s}{\rho_s}\right)\left(\frac{\rho_w}{M_w}\right)\frac{D_p^{-3}}{\varepsilon_s v_s}\right]^{1/2} \quad (2)$$

where M_w (0.01801 kg mol^{-1}) and M_S (0.1321 kg mol^{-1})

are the molar masses of solvent (water) and solute (ammonium sulfate), respectively, ρ_w (1000 kg m^{-3}) and ρ_s (1760 kg m^{-3}) are their densities, respectively. R is a universal gas constant and T is the ambient temperature (K). The effective van't Hoff factor v_s (which includes the effect of osmotic coefficient) is taken as 2.5 and σ, the surface tension of water, taken as 0.07197 J m^{-2} (Patidar et al., 2012). ε_s is the solute volume fraction, taken as unity since the aerosols were assumed to be composed of a pure soluble compound such as $(NH_4)_2SO_4$. The assumed particle size may not exactly represent the real physical scenario since the ambient particle is not necessarily a sphere.

$CCN_{predicted}$ was then calculated by integrating the size distributions from activation diameter, D_p, obtained from Eq. (2), up to the largest diameter measured through SMPS. The chemical composition and mixing state of the aerosols play a crucial role in their activation, which further influence the CCN spectrum. The presence of organic carbon as a completely insoluble fraction has a considerable effect on CCN spectrum (Zhang et al., 2011). We have studied the sensitivity of closure to the presence of estimated organic carbon. We assume insoluble organic carbon (with no effect on surface tension) and ammonium sulfate to be internally-mixed in the aerosols. The solute volume fraction in this case is given by,

$$\varepsilon_s = \frac{\frac{m_s}{\rho_s}}{\frac{m_s}{\rho_s} + \frac{m_i}{\rho_i}} \quad (3)$$

where m_s and m_i are the masses of ammonium sulfate and insoluble organic carbon, respectively, and ρ_i (1500 kg m^{-3}), ρ_s (1760 kg m^{-3}) are the densities of insoluble organic carbon and ammonium sulfate (Medina et al., 2007; Bougiatioti et al., 2009; Patidar et al., 2012). Similar studies have been done by Conant et al. (2004) who found that assumption of internal mixing of organic carbon (OC) with $(NH_4)_2SO_4$ has less impact on CCN spectrum.

2.3.4. Effective hygroscopicity parameter (κ) calculation

The effective hygroscopicity parameter (κ) is a measure of the overall hygroscopicity of all the aerosol components. It can be calculated according to the simple mixing rule (Petters and Kreidenweis, 2007).

$$\kappa = \sum_i \varepsilon_i \kappa_i \quad (4)$$

where, ε_i and κ_i are the volume fraction and hygroscopicity parameter of individual components. This parameterization is used to describe the measured CCN activity of aerosols in a multi-component system (Petters and Kreidenweis, 2007).

3. Results and discussion

3.1. CCN airborne measurements

Table 2 summarizes the cloud condensation nuclei (CCN) and condensation nuclei (CN) concentrations and their standard deviation measured during aircraft experiment. Fig. 1(a) shows the variation of CCN and CN concentrations as a function of aircraft altitude for the multilevel profiling carried out over Kanpur on 29 June 2009 during the afternoon. The average CCN and CN concentrations for this sortie were 1487 ± 388 cm^{-3} and 15,034 ± 3846 cm^{-3}, respectively. For this sortie, the MPLNET retrieved attenuated backscatter signal (ABS) is shown in Fig. 1(b), which clearly showed that the aerosol layer was present at ~6 km. It also shows that PBL extended over an altitude of ~1 km, which suggests the upward mixing of the particles that probably increases the aerosol loading within the PBL. Secondary aerosols and vehicular emissions are the major factors which contribute to the elevated particle concentration at lower altitudes in Kanpur (Chakraborty and Gupta, 2010; Gupta and Mandariya, 2013). Fig. 1(c and d) shows the average measured aerosol size distribution at multiple sampling heights for the same sortie. The statistical parameters (geometric mean diameter, D_g, geometric standard deviation, σ, and number concentration, N_g) for these aerosol size distributions are given in Table 3. The change in aerosol number-size distribution can be due to different sources such as biomass burning, combustion of fossil fuels from the industrial processes or airborne soil particles and long-range transportation of mineral dust aerosols (Eck et al., 2010; Ram et al., 2010; Mishra and Tripathi, 2008; Chinnam et al., 2006; Tripathi et al., 2007). These aerosol size distributions can also vary with altitude suggesting the different sources of origin and their aging (Rissman et al., 2006). The presence of different aerosol types at varying altitudes has also been previously observed during an aircraft experiment over Kanpur region in 2008 (Jai Devi et al., 2011). At higher altitude (5.7–6.7 km), the size distributions were observed to be unimodal with lower median diameter (~30 nm) while at lower altitudes distributions were affected by larger median diameter (~50 nm) suggesting the presence of freshly emitted aerosols from combustion of fossil fuels from industrial processes (Fig. 1(c and d)). At a height of 4.7 km, bimodal size distribution was observed with first peak at ~22 nm and second peak at ~40 nm indicating the presence of mixture of locally emitted fresh particles and those formed by homogeneous

Table 2
Summary of sorties and measurements of CCN and CN as a part of pilot CTCZ experiment.

Date	Sortie	Sampling Site	Time of flight (h) IST	Height above msl (m)	CCN (cm^{-3})		CN (cm^{-3})	
					Avg.	Std.	Avg.	Std.
29June2009	Kanpur–Pantnagar–Nainital–Kanpur	Nainital	09:15–13:00	518.16–5486.4	1363	327	18,292	3770
		Pantnagar			2601	364	9885	1970
	Kanpur	Kanpur	14:30–17:30	609.6–5486.4	1487	388	15,034	3846
30June2009	Kanpur–Lucknow–Gaya–Kanpur	Gaya	10:15–14:30	914.4–3657.6	2698	655	11,361	5570
1July2009	Kanpur–Lucknow–Gorakhpur–Kanpur	Gorakhpur	09:35–13:35	914.4–3657.6	2288	801	6810	2603
	Kanpur–Lucknow–Khajuraho	Khajuraho	15:00–18:15	914.4–5486.6	2391	850	8910	2107
2July2009	Kanpur	Kanpur	10:35–12:25	1219.2–5486.6	5293	978	9689	1526
	Kanpur	Kanpur	14:15–15:15	4572–5486.6	4431	1552	8587	3744

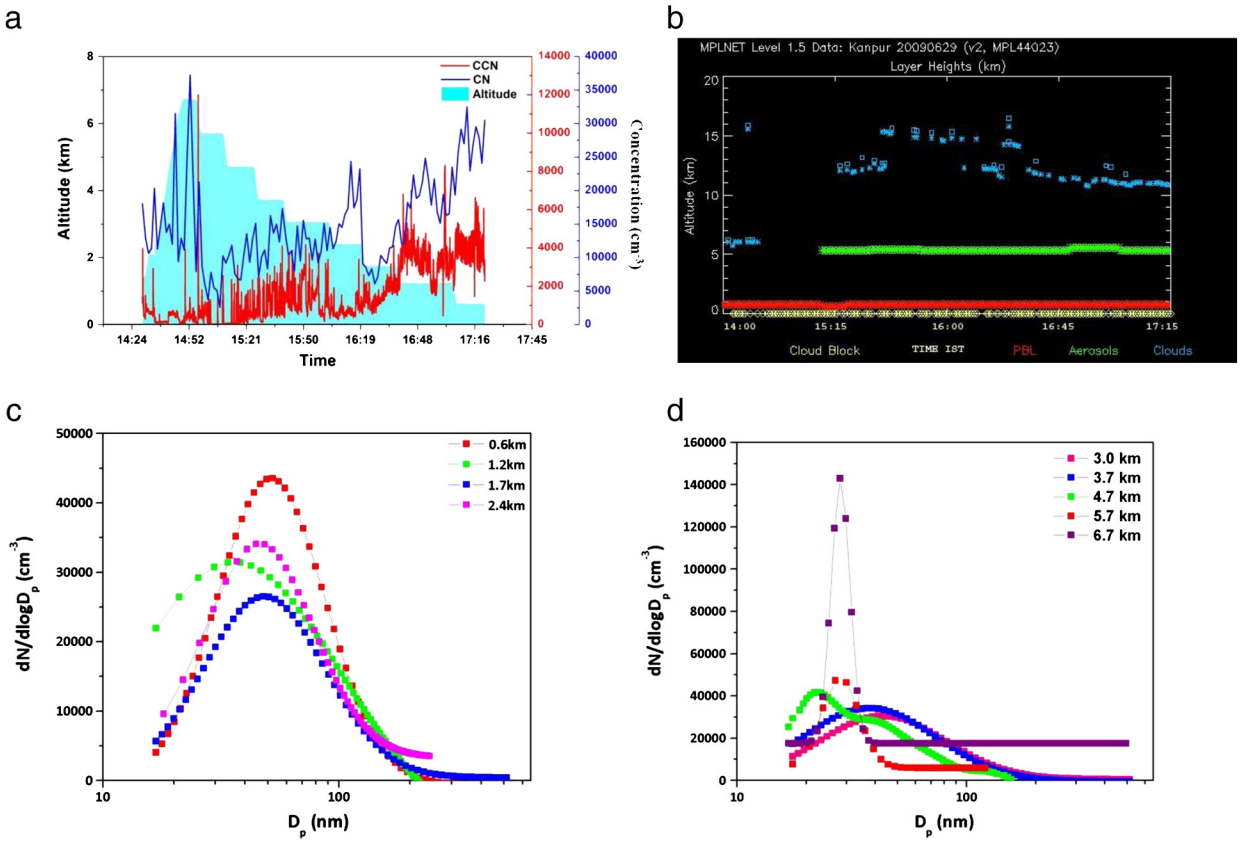

Fig. 1. (a) Time variation of CCN, CN and aircraft altitudes during the aircraft sampling over Kanpur (29 June 2009) (shaded area represents the altitude). (b) Shows the MPLNET level 1.5 aerosol/cloud occurrence for the same sortie. (c) and (d) show the measured aerosol size distribution for the same sortie.

nucleation events (O'Dowd et al., 2000). These fine mode particles may arise from the gas to particle conversion process in the atmosphere or they may be originating from vehicular emissions (Chakraborty and Gupta, 2010). Apart from this, the MPLNET image (Fig. 1b) shows the presence of clouds at around 14:00 IST at 6 km altitude, just before the sampling time. Thus, cloud evaporation may lead to new particle formation causing existing particle to shrivel up to lower size range (Aitken mode) explaining higher particle concentration at 6.7 km.

3.2. CCN closure

In this analysis, we use CCN and size distribution data from six out of nine sorties including Pantnagar, Gaya, Kanpur (forenoon), Kanpur (afternoon), Gorakhpur and Khajuraho with the assumption that the aerosols are mainly composed of pure ammonium sulfate. Table 4 summarizes the details of the closure analysis.

A good correlation between measured CCN and predicted CCN was observed for all sorties (Fig. 2). Overall, the closure ratio for the present closure analysis varies from 1.16 to 1.94 with respect to the different flights. The deviation of the closure data points from the 1:1 line could be possibly due to a number of reasons. The uncertainties in $CCN_{predicted}$ could be related to the uncertainties in the instrument supersaturation and the aerosol size distribution measurements (Lance et al., 2009). In addition, the size distribution obtained from SMPS has the scan time of 90 s and therefore it may not be

Table 3
Statistical parameters (*Dg*, σ, and *Ng*) for averaged aerosol size distribution for Kanpur sortie, 29 June.

Height (km)	*Dg* (nm)	σ (unitless)	N_g (cm^{-3})
6.7	28.49	0.09	20,661 ± 9334
5.7	29.19	0.18	7859 ± 4265
4.7	22.24	1.07	9841 ± 2401
3.7	59.75	0.63	12,356 ± 3500
3.0	64.03	0.66	11,976 ± 2321
2.4	60.69	0.63	16,341 ± 3669
1.7	58.36	0.58	9994 ± 2950
1.2	50.2	1.04	18,662 ± 3485
0.6	68.05	0.52	27,620 ± 2990

Table 4
CCN closure results for considered six sorties.

Multileveling over	Data points	$CCN_{predicted}/CCN_{measured}$
Pantnagar	17	1.56 ± 0.26
Gaya	50	1.94 ± 0.24
Gorakhpur	20	1.18 ± 0.13
Khajuraho	75	1.24 ± 0.18
Kanpur (FN)	31	1.16 ± 0.11
Kanpur (AN)	08	1.17 ± 0.15
Average $CCN_{predicted}/CCN_{measured}$ = 1.375 ± 0.17		

able to capture the rapidly changing aerosol size distribution, while CCN counter measures the CCN concentration for every 1 s and is thus able to characterize the rapidly changing aerosol size distribution (Rissman et al., 2006; VanReken et al., 2003). The overprediction bias (closure ratio > 1) depends mainly on particle concentration. For particle concentration ~ 8910 cm^{-3} (Khajuraho sortie), the average overprediction bias is low which suggests cleaner air mass. The overprediction bias increases to 1.94 as the particle concentration rises to ~11,000 cm^{-3} for Gaya sortie. This suggests presence of higher percentage of insoluble components which inhibits CCN activity. Also, the coincidence error in CPC and SS depletion in CCN are the major factors affecting the closure ratio.

The closure ratio varies considerably with respect to changing altitude of individual flight. For example, the closure ratio of the Kanpur sortie varies from 0.75to 1.51, which likely suggests that the sampling of air masses of different characteristics at different altitudes. At lower altitudes, aerosol loading resulted due to pollution emitted from anthropogenic activities such as fossil fuel combustion. According to Roberts et al. (2003), smoke aerosols originated from biomass burning and carried through long-range transport, may also serve as CCN. In addition, coal based thermal power plants, and vehicular emissions may serve as the large source of aerosol incursion (Mehta et al., 2009; Gupta and Mandariya, 2013).

Earlier, airborne measurements across the British Isles and the North Sea showed better closure results with the assumption of pure ammonium sulfate, for marine air mass rather than continental (Martin et al., 1994). Following the same assumption, 5% overprediction for SS = 0.2% and 20% for SS = 0.85% were observed during CRYSTAL-FACE study (VanReken et al., 2003). However, closure studies conducted over the regions influenced by industrial and urban sources found that incorporation of hygroscopicity of water soluble organic mass fraction improves the CCN prediction significantly (Asa-Awuku et al., 2011). Also, they found that CCN activity increases with increase in O/C. Closure ratio obtained between 1.18 and 3.68 during Atmospheric Measurement Program's (ARM) Aerosol Intensive Observational Period (IOP) suggested presence of less hygroscopic species (Rissman et al., 2006). Chuang et al. (2000) used ground based chemical data for airborne closure study, which overpredicted CCN concentration by more than tenfold. It was proposed to have airborne chemical composition data for better closure studies. In spite of the spatial and temporal variability, several studies achieved good closure results because of the presence of less organic carbon content in the aerosols and were less affected by anthropogenic sources (Broekhuizen et al., 2006). Medina et al. (2007) suggested that information on size resolved chemical composition and aerosol mixing state is required during changing wind directions to improve closure. High (CCN > 10,000 #/cm^3) and (CN > 25,000 #/cm^3) concentration was obtained during two flights within and downwind of Houston regional plume and over Houston ship channel in the Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS) campaign 2006. An overprediction of 3–36% is obtained by using chemical composition from C-ToF-AMS. Assumption of internally mixed aerosols with pure ammonium sulfate composition attained a closure ratio of 1.36. However, incorporation of internally mixed water soluble fraction improved the closure ratio to 1.03. It was also suggested to include size resolved chemical composition, their mixing state and aerosol surfactant properties to further improve the CCN closure (Lance et al., 2009). Unlike Rissman et al. (2006) which employed characteristics of externally mixed aerosols, introduction of water insoluble organic fraction and internal mixing state (ammonium sulfate and organics) improved our closure results by 34–37.5% for Gaya and Pantnagar, respectively (see discussion in Section 3.2.2). The information on size resolved chemical composition and its WSOC content could further improve the closure ratio. However, VanReken et al. (2003) discovered assumption of pure $(NH4)_2SO_4$ over regions affected by marine aerosols as a good estimate to achieve good closure results. A very recent study of Secondary Organic Aerosol (SOA) formation by the oxidation of organic species emitted by the Deepwater Horizon oil spill found that presence of organic rich aerosol particles increased the critical supersaturation from

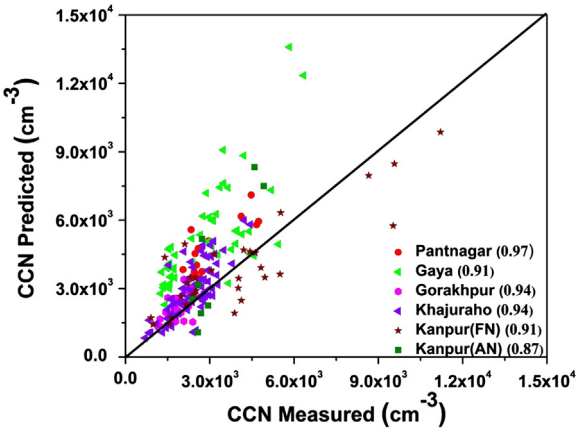

Fig. 2. CCN closure for six sorties, assuming that the aerosols are composed of pure ammonium sulfate.

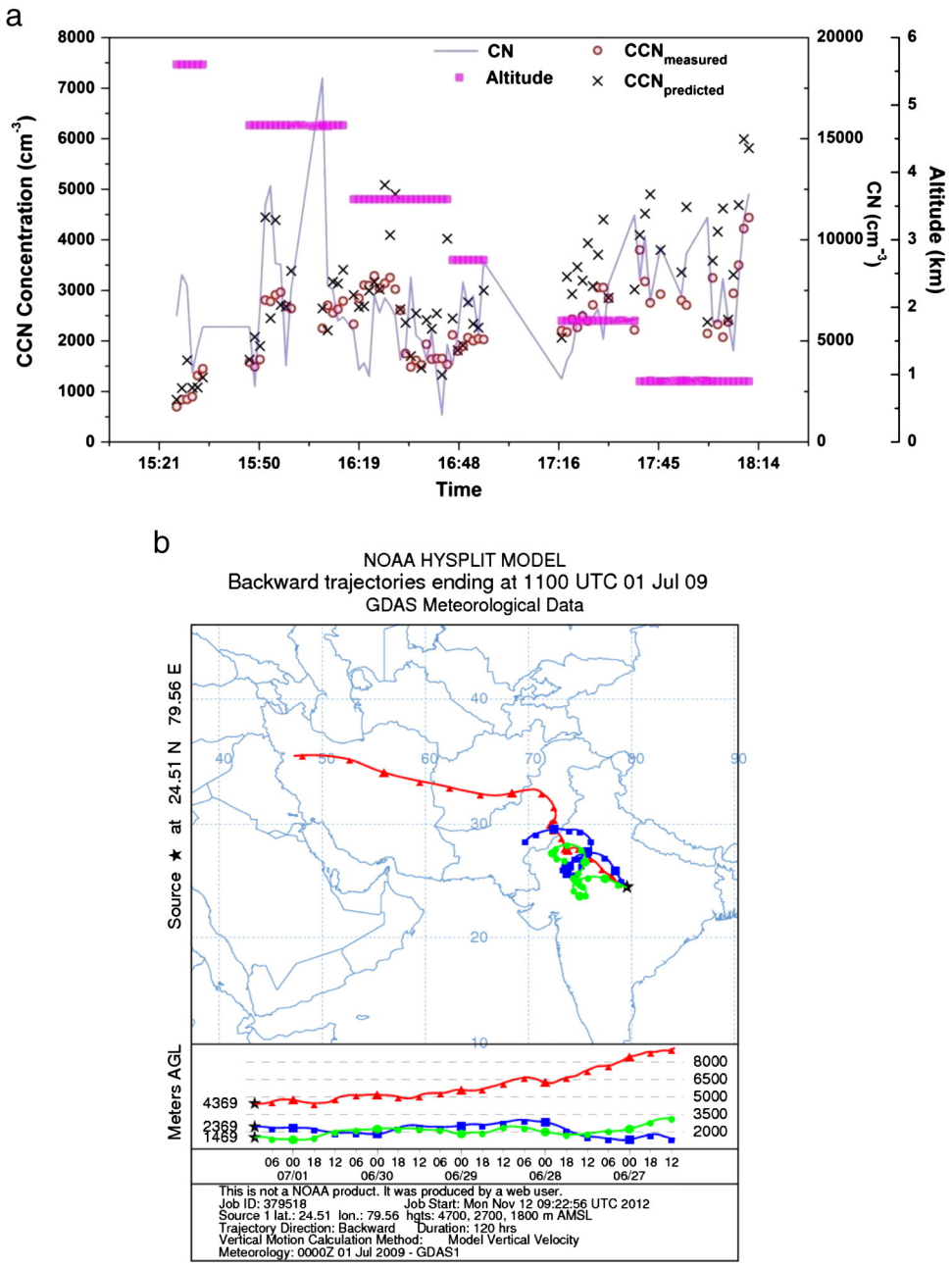

Fig. 3. (a) Time variation of $CCN_{predicted}$, $CCN_{measured}$, CN and altitude for Khajuraho sortie, (b) NOAA HYSPLIT 3 days backward trajectory for Khajuraho sortie (Kanpur–Lucknow–Khajurao) at heights 4700, 2700, and 1800 m.

$0.3 \pm -.05\%$ to $0.4 \pm 0.1\%$. Also, the κ value (0.05–0.1) derived from the chemical composition and humidified particle light extinction from C-ToF-AMS and cavity ring down spectrometer, respectively, showed the presence of externally mixed aerosols and their size dependent chemistry (Moore et al., 2012).

3.2.1. A case study of Khajuraho: 1 July 2009

The aircraft measurements were performed over Khajuraho at six different altitudes ranging from 900 m to 5600 m. Fig. 3(a) shows the time series of the $CCN_{predicted}$, $CCN_{measured}$, CN and aircraft altitudes. The concentration of the $CCN_{measured}$ varies significantly with the height, ranging from 1000–4000 cm^{-3}. The highest $CCN_{measured}$ concentrations were sampled at 900 m which probably has the maximum overprediction bias of 1.40. The air mass with higher CN concentration indicates the presence of elevated aerosol sources such as vehicular emissions, having relatively high level of insoluble species. This often leads to the higher closure ratio (Rissman et al., 2006). The moderate CCN concentration was sampled at 1800 m, 2700 m and 3600 m with closure ratio of 1.17–1.29. Both CCN concentration and closure ratio decrease as the height increases to 4700 and 5600 m. Sometimes the CN peak is not followed by the CCN peak

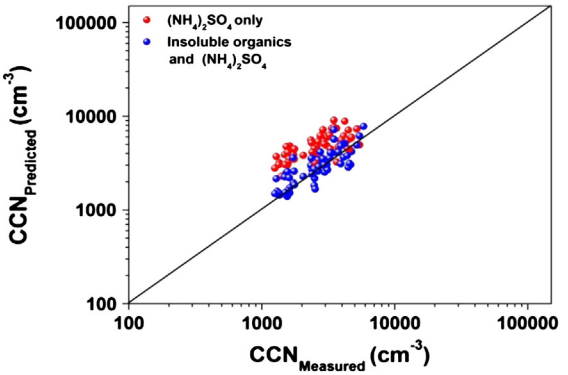

Fig. 4. Illustrates the change in the closure ratio with the assumption of internally mixed insoluble organic fraction for Gaya and Pantnagar sorties.

suggesting the presence of fresh combustion aerosol or some nucleation event causing a sudden increase in the particle concentration (Medina et al., 2007). The particle concentration was found to be maximum at a height of 4700 m possibly due to air mass coming from parts of Iraq, Pakistan and Afghanistan (shown in Fig. 3(b)). Also, the CCN concentration is observed to be minimum because of the depression in CCN activity due to the presence of mineral dust aerosols. The back trajectories computed over all the six heights (0.91, 1.75, 2.7, 3.7, 4.7, and 5.6 km) suggest that the air mass was of continental origin and was affected by anthropogenic pollution. In addition, the air mass back trajectories at 1800–4700 m, in Fig. 3(b), indicate that the air mass was purely continental in nature.

On the other hand, the closure study performed over the Kanpur region (2 July 2009, forenoon) shows an underprediction of 0.89 and 0.75 at 1400 m and 4000 m, respectively, with an average closure ratio for all heights equal to 1.16 (Table 4). At the height of 3400 m, contrary to the Khajuraho sortie, in spite of low particle concentration, the overprediction bias is high (1.43). This could be due to coagulation and cloud processing which reduced the number of particles.

3.2.2. CCN closure with insoluble organic carbon

In order to examine the effect of insoluble species and their mixing state on CCN activation, we assume that aerosol is composed of two components as an internal mixture of soluble $(NH_4)_2SO_4$ and hydrophobic organic carbon (Medina et al., 2007), unlike we presented earlier in the closure analysis, with only soluble $(NH_4)_2SO_4$. This has been achieved with the aid of the modified Köhler equation. Due to availability of chemical data for Pantnagar and Gaya only, we have performed this sensitivity analysis for these two sorties.

The organic carbon mass was determined by subtracting the mass of ammonium sulfate from the PM_1 mass (Medina et al., 2007). In the present study, it was assumed that the organic carbon fraction was height-independent. The PM_1 mass for the Pantnagar and Gaya sorties was 92.3 μg m^{-3} and 135.3 μg m^{-3}, resulting in the organic carbon mass as 74.0 μg m^{-3} and 103.5 μg m^{-3}, respectively. This gives the solute volume fraction, ε_s value for Pantnagar and Gaya sorties as 0.18 and 0.20, respectively. Fig. 4 shows the comparison of closure results for $(NH_4)_2SO_4$ alone and for internally mixed insoluble organic carbon and $(NH_4)_2SO_4$. The inclusion of insoluble organic fraction in the aerosol composition shows an overall improvement from 1.75 to 1.12 in the closure ratio. The underprediction in few cases may be reconciled by considering that either the organics in the air mass sampled are partially soluble or they lower the droplet surface tension (thus facilitating the activation of particles and increasing the CCN concentration) (Lance et al., 2009). The overprediction for Pantnagar improves by 37.5% while for Gaya sortie it was 34.6%. Assuming internally mixed insoluble organic fraction to the aerosol composition, improves the closure ratio but this hypothesis could not be completely true, as our measurements lack the single particle information of chemical composition and the aerosol mixing state (Lance et al., 2009).

Following the mixing rule, the effective hygroscopicity parameter was also calculated for these sorties, using κ for organic carbon = 0.1 and κ for $(NH_4)_2SO_4$ = 0.6 (Wang et al., 2010; Pringle et al., 2010; Gunthe et al., 2009). Atmospheric particles can have hygroscopicity ranging from 0.1 to 0.9 (Petters and Kreidenweis, 2007). κ values calculated for Pantnagar and Gaya are 0.51 and 0.5, respectively, which suggests that aerosol population was dominated either by inorganic hygroscopic species or moderately hygroscopic organic content, which have capacity to behave as CCN (Petters and Kreidenweis, 2007). Other airborne measurements also suggested that inclusion of WSOC reduces the hygroscopicity although higher κ values ~0.88 are observed which corresponds to the species which are equally soluble as $(NH_4)_2SO_4$ (Asa-Awuku et al., 2011). A strong positive relationship between organic O/C ratio and κ has been revealed through various laboratory and field studies (Jimenez et al., 2009 and references therein). Vertical distribution of κ is not shown here because of the assumption involved that organic carbon fraction is height-independent.

4. Conclusion

In-situ aircraft measurements of CCN concentration at SS of 0.84% and aerosol size distribution have been carried out during the CTCZ aircraft campaign. Out of total 9 sorties, we present closure analysis from six flights. The aerosol CCN activation depends not only on the solute volume fraction but also on the number concentration and size distribution of aerosols obtained from SMPS, and their mixing state. The closure analysis assuming aerosols is composed of pure ammonium sulfate showed a very good agreement between the $CCN_{measured}$ and $CCN_{predicted}$ concentrations with an average closure ratio of 1.375, n = 229 and R^2 = 0.80. For these sorties, the CCN closure ratio varied greatly with altitude suggesting that the sampled air mass has different characteristics at different altitudes. The sampled airmass might be influenced by the vehicular emissions and biomass burning aerosols from local source or long-range transport. The closure ratio also varies with CN concentrations i.e. for high CCN concentrations the closure ratio was high. The internally mixed fraction of insoluble organic carbon (considering no surface tension effect) significantly improved the closure ratio by ~36%. The effective hygroscopicity parameter κ, calculated by simple mixing rule is also consistent with the assumption and suggests that $CCN_{predicted}$ concentration is

highly sensitive for aerosols with high insoluble organic fraction (~80% for selected two sorties).

CCN closure analysis on airborne measurements over the Indian region is a step towards understanding the dependence of CCN efficiency on the size distribution and chemical composition of the aerosol, which is one of the key linkages between aerosols, clouds and ultimately climate. It also improves our understanding of hygroscopic properties of aerosols of Indian origin.

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