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• **Original Paper** •

Cloud Condensation Nuclei over the Bay of Bengal during the Indian Summer Monsoon

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ABSTRACT

The first measurements of cloud condensation nuclei (CCN) at five supersaturations were carried out onboard the research vessel "Sagar Kanya" (cruise SK-296) from the south to the head-bay of the Bay of Bengal as part of the Continental Tropical Convergence Zone (CTCZ) Project during the Indian summer monsoon of 2012. In this paper, we assess the diurnal variation in CCN distributions at supersaturations from 0.2% to 1% (in steps of 0.2%) and the power-law fit at supersaturation of 1%. The diurnal pattern shows peaks in CCN concentration (N_{CCN}) at supersaturations from 0.2% to 1% between 0600 and 0700 LST (local standard time, UTC+0530), with relatively low concentrations between 1200 and 1400 LST, followed by a peak at around 1800 LST. The power-law fit for the CCN distribution at different supersaturation levels relates the empirical exponent (k) of supersaturation (%) and the N_{CCN} at a supersaturation of 1%. The N_{CCN} at a supersaturation of 0.4% is observed to vary from 702 cm^{-3} to 1289 cm^{-3}, with a mean of 961 ± 161 cm^{-3} (95% confidence interval), representing the CCN activity of marine air masses. Whereas, the mean N_{CCN} of 1628 ± 193 cm^{-3} at a supersaturation of 1% is higher than anticipated for the marine background. When the number of CCN spectra is 1293, the value of k is 0.57 ± 0.03 (99% confidence interval) and its probability distribution shows cumulative counts significant at $k \approx 0.55 \pm 0.25$. The results are found to be better at representing the features of the marine environment (10^3 cm^{-3} and $k \approx 0.5$) and useful for validating CCN closure studies for Indian sea regions.

Key words: CTCZ, Bay of Bengal, monsoon, CCN, supersaturation, power-law relationship

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1. Introduction

Cloud condensation nuclei (CCN) are fractions of atmospheric aerosols that grow to the size of cloud droplets at a specified supersaturation level in marine, continental and in-cloud environments. Low and high CCN concentrations (N_{CCN}) are generally observed in the marine background and polluted regions, respectively (Petters and Kreidenweis, 2007; Andreae, 2009; Bougiatioti et al., 2011; Leena et al., 2016). Field observation campaigns (e.g. Andreae and Rosenfeld, 2008) have improved our understanding of CCN activity (Ervens et al., 2010). CCN formation is an important phenomenon in cloud physics (Fitzgerald, 1973, 1991) and the activation of hygroscopic aerosols to the size of CCN largely depends on the size, source, chemical composition and mixing state of particles that nucleate as CCN and grow into droplets (Su et al., 2010; Asmi et al., 2012). The aerosol hygroscopicity in CCN formation depends on the air mass that prevails over the regions of observation (Pruppacher and Klett, 2010). Though the chemical composition of nuclei that activate into CCN distributions is difficult to measure in real time (Murugavel and Chate, 2011; Krüger et al., 2014), a power-law fit for CCN distributions, as a function of supersaturation with the exponent (k) of supersaturation and C (the N_{CCN} in cm^{-3} at a supersaturation of 1%), represents the hygroscopicity of particles that nucleate to CCN size (McFiggans et al., 2006; Hegg et al., 2009). For inland stations in India, the CCN distributions along with power-law fits are reported as a function of specified supersaturation over short time scales (order of seconds) using commercially available CCN-100 counters (Leena et al., 2016; Varghese et al., 2015). Similar measurements of CCN distributions at various supersaturation values over Indian sea regions during the south-

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west summer monsoon can result in a better power-law formulation with C and k. Atmospheric measurements of CCN distributions at a wide range of supersaturation levels from the south to the head-bay region of the Bay of Bengal (BoB) have largely been neglected (CTCZ-Scientific Steering Committee, 2011), except shipborne observations reported by Ramana and Devi (2016) for the southernmost tail-bay region of the BoB.

The present work focuses on an analysis of the in-situ measurements of CCN distributions at a wide range of supersaturation over the barely explored region of the BoB from the south to the head-bay region using CCN-100 counters deployed onboard the research vessel "Sagar Kanya" (cruise SK-296) during the Indian summer monsoon of 2012. During this season, the prevailing air mass maintained a symmetrical supply of moisture over the BoB. Therefore, the measured CCN distributions at a wide range of supersaturation in the study region motivated us to quantify the power-law parameters for southwesterly clean air masses, and to compare them with other marine environments. Also, this work aims to showcase the first observations of CCN distributions at various levels of supersaturation carried out in the marine sector spread over an area from the south to the head-bay region of the BoB during the monsoon season, and to investigate the power-law fit for the empirical parameters C and k for the monsoonal air mass. Moreover, the results from the CCN distribution measurements at a wide range of supersaturation levels are expected to improve our understanding of the CCN activity for monsoonal clouds over Indian sea regions.

2. Instrumentation

Atmospheric particles that transform into CCN were measured with a CCN counter (model: CCN-100). This instrument is a continuous flow thermal gradient CCN counter proposed and designed by Roberts and Nenes (2005) and manufactured by Droplet Measurement Technologies. The working principle of this CCN counter is to expose the aerosol to a fixed supersaturation at a certain time and to measure the number of activated particles with an inbuilt optical particle counter. Aerosols continuously flow through the center part of a cylinder with a wetted wall. Between the aerosol flow and the wall there is a particle-free sheath flow. By controlling the temperature of the wall as well as keeping it wet (ensuring that the relative humidity is 100% just outside the cylinder wall), the movement of heat and water vapor towards the middle of the cylinder and the supersaturation value can be maintained. The supersaturation values are altered in a cycle for measurement of the activated N_{CCN}. The working principle (thermophoresis) of the instrument depends on the water molecules that diffuse towards the center faster than the heat added across the wall (water molecules diffuse mainly via heavier nitrogen and oxygen molecules, hence giving saturation ratios above 100%). Details on the measurement uncertainties and operational error are discussed elsewhere (Rose et al., 2008; Krüger et al., 2014). Full details on the operation, maintenance and calibration procedure of the CCN-100 counter can be found at http://www.dropletmeasurement.com. Also, Ramana and Devi (2016) described the deployment of CCN-100 onboard the research vessel "Sagar Nidhi" during a cruise over the BoB.

3. Study region

Figures 1a and b show the ORV Sagarkanya-296 track positions along with wind-rose diagrams, based on observed winds over the BoB during the cruise period from 10 July to 8 August 2012. As seen in Fig. 1a, since the departure of SK-296 from Chennai port on 10 July 2012, it sailed till 13 July 2012 almost parallel to the entire coastline of the Indian peninsula and reached the head-bay region of the BoB on 19 July 2012. The research vessel "Sagar Kanya" remained stationary in the head-bay region from 19 July to 2 August 2012 (Fig. 1a) and thereafter started its return expedition on 2 August 2012 towards the south of the bay, parallel but relatively far away from the Indian coastline, and arrived at Chennai port on 8 August 2012. The Indian summer monsoon season generally extends from June to September when the ITCZ shifts its position over India, maintaining monsoonal cloud cover and moisture supply over the entire country. The dominant circulation pattern is southwesterly clean air masses from June to September, with strong near-surface winds over the Ocean. CCN distributions were continuously monitored at a wide range of supersaturation levels over a period that included the expedition of the SK-296 cruise from the south to the head-bay region of the BoB (10 July to 8 August 2012). A very high total N_{CCN} ($\sim$ 7500 cm^{-3}) was recorded on 17 July 2012 when SK-296 was at Paradeep port in the BoB, and also on 5 August 2012 (due to rain). The prevailing southwesterly air mass maintains the symmetric moisture supply over the BoB, as evident from the wind-rose diagram (Fig. 1b). Several rain showers were encountered during the campaign period, while typical monsoonal clouds passed over the SK-296 track positions across the BoB. The prevailing weather conditions in the marine environment of the BoB during July to August 2012 are described in Ramana and Devi (2016). Over the south to the head-bay region of the BoB, the sampling period (10 July to 7 August 2012) of the SK-296 cruise was long enough to represent the monsoonal pattern of CCN distributions at various supersaturation levels.

4. CCN distribution data

The CCN distributions at supersaturations of 0.2%, 0.4%, 0.6%, 0.8% and 1% (covering typical range of supersaturation of the marine to the in-cloud environment) were monitored over the region from the south to the head-bay region of the BoB, round the clock, during 10 July to 7 August 2012. The activated N_{CCN} is given with a temporal resolution of one second and, since it takes a few minutes for the system

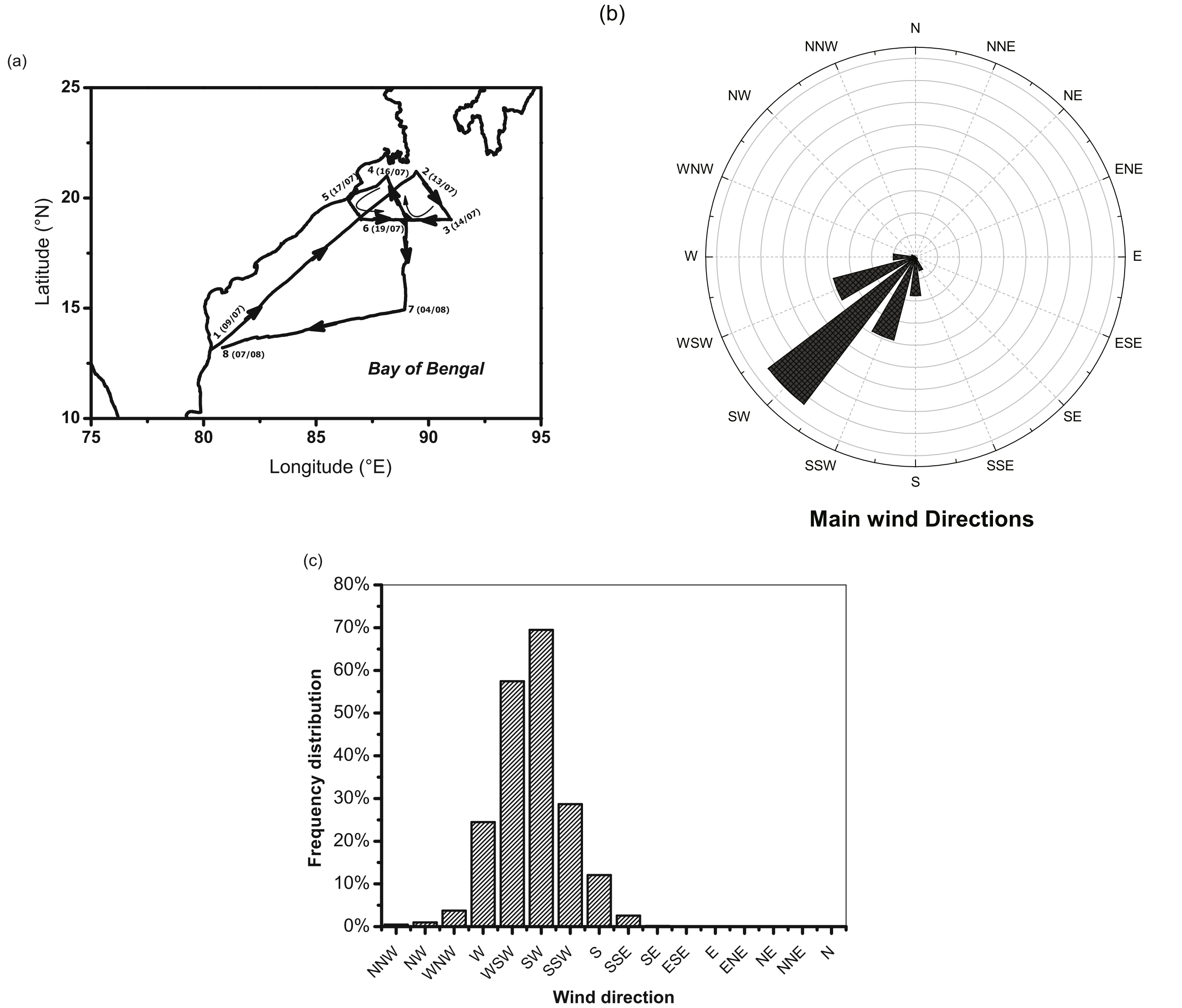

Fig. 1. (a) SK-296 track positions. (b) Wind-rose diagram (black shadings mark the southwest and south-southwest winds). (c) Frequency distribution of winds.

to come to equilibrium state with the supersaturation, a measurement cycle of 30 minutes is considered for the aforementioned levels of supersaturation (Krüger et al., 2014). The data obtained for CCN distributions as a function of supersaturation have been averaged on an hourly basis for the entire period of the SK-296 cruise campaign to showcase the results on the diurnal scale for the Indian summer monsoon of 2012. Figure 2 shows the diurnal pattern of the CCN distribution for the measurement period from 10 July to 7 August 2012 at each level of supersaturation from 0.2% to 1% (in steps of 0.2%). The diurnal cycle includes the peaks in CCN distributions between 0600 and 0700 LST (local standard time, UTC+0530) of about 634, 1122, 1425, 1619 and 1857 cm^{-3}, followed by lower concentrations of about 543, 736, 874, 1100 and 1428 cm^{-3} between 1200 and 1400 LST and subsequent peaks at 1800 LST of 784, 1262, 1587, 1754 and 2027 cm^{-3}, for supersaturations of 0.2%, 0.4%, 0.6%, 0.8% and 1%, respectively (Fig. 2). The diurnal cycle for CCN distributions at different values of supersaturation are believed to follow the monsoonal pattern of ventilation coefficients (product of mixing height and wind speed), which diurnally modulates the nucleating particle concentrations over Indian sea regions (Murugavel and Chate, 2011), including the southernmost tail-bay region of the BoB (Ramana and Devi, 2016). Information on the composition of the aerosol population fraction is embedded in the empirical parameter k, which can be extracted from the power-law fit of CCN distributions at various supersaturation levels (SK-296 cruise) over the south to the head-bay region of the BoB.

The power-law $N_{\mathrm{CCN},S} = CS^k$ of CCN distributions at different values of supersaturation describes the CCN activation, where $N_{\mathrm{CCN},S}$ is the concentration of CCN at a specified supersaturation S, C is the CCN concentration at a supersaturation of 1%, and k is the slope of the power-law fit curve. The diurnal variation of k is plotted in Fig. 2b, and shows a first peak at around 0600 LST, a low at 1200 LST, and another

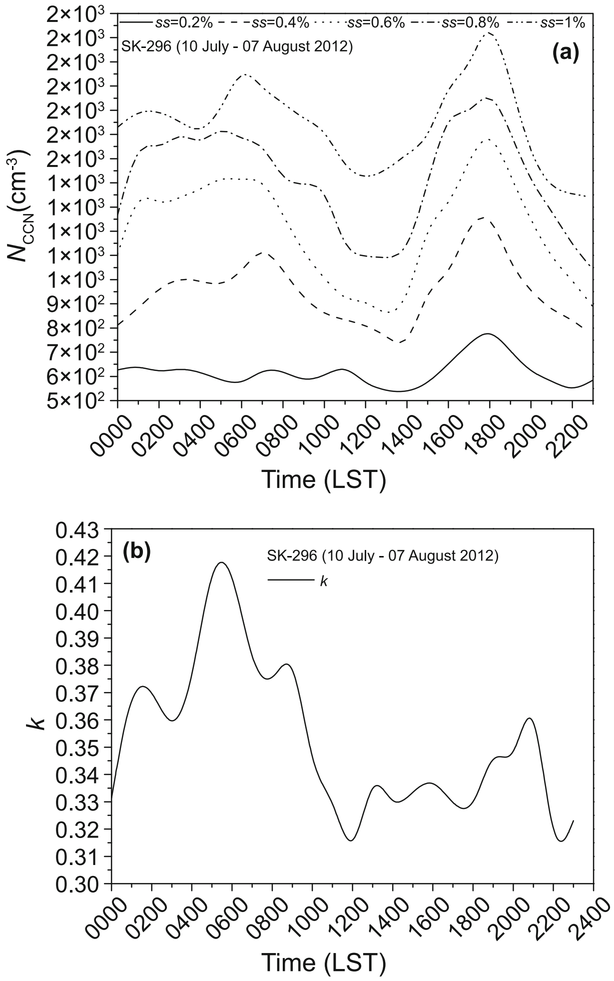

Fig. 2. Diurnal patterns of (a) the CCN distributions at each supersaturation level from 0.2% to 1% (in steps of 0.2%) and (b) k, for the measurement period from 10 July to 7 August 2012.

peak at 2000 LST. Thus, the diurnal pattern of k is purely due to ventilation conditions.

The variations in the CCN distribution along with the standard deviation (Fig. 3) show an increase in N_{CCN} with the level of supersaturation from 0.2% to 1%. The variations in the slope (k) in Fig. 3 seem to be synchronous with C, where k contains information about the source and mixing state of particles analogous to that of the hygroscopicity of nucleated particles. The results suggest that, during the monsoon season, there is a dominance of hygroscopic particles over the BoB. Furthermore, the average values of C and k in Fig. 3 are $C = 1659 \pm 29$ cm^{-3} (at a supersaturation of 1%) and $k = 0.57 \pm 0.03$ ($R^2 = 0.99$) for the entire dataset (number of CCN distribution spectra ≈ 1300). For N_{CCN} measured along and off the central Californian coast during August 2007, Hegg et al. (2009) reported the power-law fit parameters as $C \approx 328 \pm 10$ cm^{-3} and $k \approx 0.72 \pm 0.06$ ($R^2 = 0.99$). For the present dataset for the SK-296 observation period between 10 July and 7 August 2012, the mean N_{CCN} of 1628 ± 193 cm^{-3} (at a supersaturation of 1%) appears to be higher than the anticipated value for the marine background, for which C values are more typically of the order of a few hundred CCN cm^{-3} and $k \approx 0.5$, as suggested by Hegg and Hobbs (1992) and Hegg et al. (2008). Many observational studies (Dinger et al., 1970; Gras, 1990; Pruppacher and Klett, 2010) have reported a value of $k \approx 0.5$ for the maritime environment. Hegg et al. (1991) suggested a value of $k > 0.5$ for N_{CCN} during monsoon. For the marine region, off the central Californian coast, Hudson et al. (2000) measured background C and k values of about 450 cm^{-3} and 0.65, respectively; while in June and late July, Hudson (2007) reported a value of $C \approx 10^3$ cm^{-3}. Thus, our LSTCCN distribution results from the SK-296 expedition corroborate reasonably well with the C and k values reported for marine environments.

Ramana and Devi (2016) reported N_{CCN} at a supersaturation of 0.4% of about 1245–2225 cm^{-3} (mean $N_{CCN} \approx 1801 \pm 486$ cm^{-3}), 191–938 cm^{-3} (mean $N_{CCN} \approx 418 \pm 161$ cm^{-3}) and 64–1420 cm^{-3} (mean $N_{CCN} \approx 291 \pm 209$ cm^{-3}) for coastal (21 July 2012), clean marine (23 July to 11 August 2012) and shipping lane ranges (13–16 August 2012), respectively, in the southernmost tail-bay region of the BoB. Furthermore, for the sampling period from 21 July to 16 August 2012, they reported the N_{CCN} as 837 ± 285 cm^{-3} at a supersaturation of 0.4% which is a mean of 1801, 418 and 291 cm^{-3}. Similarly, for the entire dataset of the SK-296 expedition (10 July to 7 August 2012), N_{CCN} at a supersaturation of 0.4% varied from 702 to 1289 cm^{-3}, with a mean of 961 ± 151 cm^{-3}. The mean N_{CCN} (837 ± 285 cm^{-3}) at a supersaturation of 0.4% from the southernmost tail-bay region of the BoB reported by Ramana and Devi (2016) was lower, by about 15%, than the mean N_{CCN} of the present study (961 ± 151 cm^{-3} at a supersaturation of 0.4%) for the south to the head-bay region of the BoB over the sampling period from 10 July to 7 August 2012 during the SK-296 cruise.

Figures 4a and b illustrate the probability distribution of k, with its cumulative counts in percent, for the period of observations from 10 July to 7 August 2012. Figure 4a shows the cumulative counts increase with k for the entire dataset

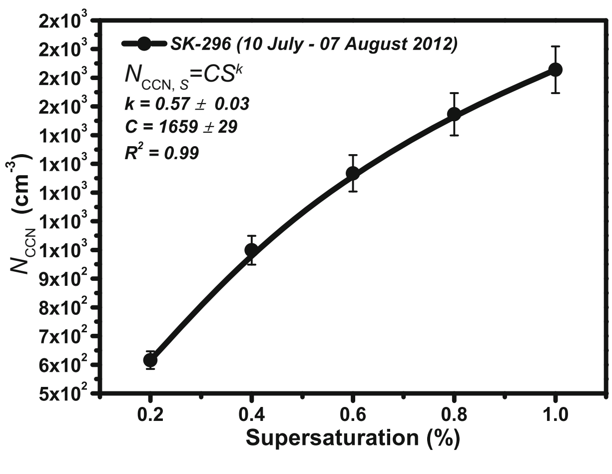

Fig. 3. Variations in CCN distribution along with standard deviations as a function of supersaturation (%) for the power-law fit on the entire dataset for the period 10 July to 7 August 2012.

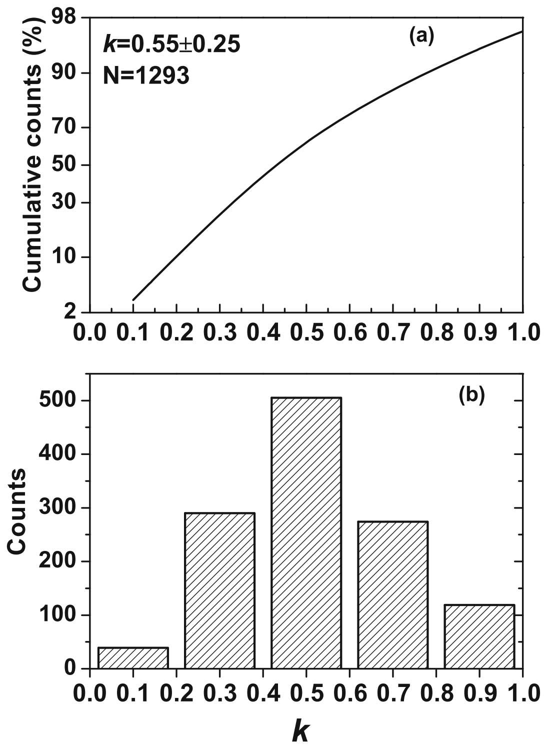

Fig. 4. Probability distributions of k (a) cumulative counts and (b) counts.

(number of CCN distribution spectra = 1293) obtained during the SK-296 cruise campaign. It is evident from Fig. 4a that the probability counts are significant at $k \approx 0.55 \pm 0.25$. A clear influence of marine-type air masses on the CCN distributions and power-law fit parameters C and k can be seen from the aforementioned analyses of the entire dataset obtained during the SK-296 cruise campaign. This is likely linked to increased natural sources of CCN in the south to the head-bay region of the BoB over the sampling period from 10 July to 7 August 2012 (SK-296) due to enhanced marine-derived aerosols in southwesterly air masses. The parameters C and k with a power-law fit on the entire dataset of the SK-296 cruise, and also from the probability distributions, show the best estimates for a typical marine environment in the tropics, and hence may be applicable to most cloud microphysical studies, including CCN closure studies.

5. Summary and conclusions

As part of the CTCZ programme, CCN distributions at supersaturations from 0.2% to 1% (in steps of 0.2%) were continuously monitored onboard the research vessel "Sagar Kanya" (SK-296 expedition) during the Indian summer monsoon of 2012. The results of the hourly mean CCN distributions at supersaturations of 0.2% to 1% for the entire dataset on the diurnal scale, and the power-law fit with empirical constants C and k, are discussed in a comparative analysis. The peaks in N_{CCN} appear during morning and evening hours, with lower N_{CCN} during noon hours, at supersaturations of 0.2%, 0.4%, 0.6%, 0.8% and 1%. For the entire dataset from the SK-296 cruise campaign (number of CCN distribution spectra = 1293), the mean CCN concentrations are 1628 ± 193 cm^{-3} and 961 ± 151 cm^{-3} at supersaturations of 1% and 0.4%, respectively; while from the power-law fit, $k = 0.57 \pm 0.03$ ($R^2 = 0.99$), and probability distributions, cumulative and probability counts show significance at $k = 0.55 \pm 0.25$. Though the mean N_{CCN} at a supersaturation of 1% is higher than expected for the marine background, the mean N_{CCN} at a supersaturation of 0.4%, as well as the C and k, broadly corroborate the results of marine environments. Knowledge of the parameter k is routinely considered to be sufficient for many cloud microphysical applications, while for the SK-296 dataset, $k \approx 0.57 \pm 0.03$ represents the CCN distributions in the marine environments of Indian sea regions. The values of C and k in the present study suggest that the track positions of SK-296 may be impacted by sources other than the sea surface in the case of a few events during the campaign. The quantitative evaluation of the contributing sources to the CCN distributions for Indian sea regions is beyond the scope of this study and can be addressed separately. Also, no significant trend in the monthly (July and August) arithmetic mean N_{CCN} was found for the sampling period, and the conclusion is that more shipborne CCN distribution data are expected to enable a more robust analysis of possible trends. The availability of shipborne data should facilitate an increase in our understanding of the processes linking N_{CCN}, aerosol concentrations and cloud droplet number concentrations (and cloud albedo) for the BoB region.

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