

Investigation of aerosol indirect effects on monsoon clouds using ground-based measurements over a high-altitude site in Western Ghats

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Abstract: The effect of aerosols on cloud droplet number concentration and droplet effective radius is investigated from ground-based measurements over a high-altitude site where in clouds pass over the surface.

15 First aerosol indirect effect AIE estimates were made using i) relative changes in cloud droplet number concentration (AIE_n) and ii) relative changes in droplet effective radius (AIE_s) with relative changes in aerosol for different cloud liquid water contents (LWC). AIE estimates from two different methods reveal that there is systematic overestimation in AIE_n as compared to that of AIE_s . Aerosol indirect effects (AIE_n and AIE_s) and Dispersion effect (DE) at different LWC regimes ranging from 0.05 to 0.50 gm^{-3} were estimated. The analysis demonstrates that there is overestimation of AIE_n as compared to AIE_s which is mainly due to DE. Aerosol effects on spectral dispersion in droplet size distribution plays an important role in altering Twomey's cooling effect and thereby changes in climate. This study shows that the higher DE in the medium LWC regime which offsets the AIE by 30%.

25 1. Introduction

There exists a strong correlation between aerosol and cloud microphysics, more aerosols leads to many smaller cloud droplets for a fixed liquid water content, which increases the cloud optical thickness and cloud albedo, this is termed as AIE which in turn cool the Earth's surface. But anthropogenic aerosols having complex chemical composition will broaden cloud droplet spectral width and reduce the AIE (*Liu and Daum, 2002*). Several theoretical and observational studies of aerosol-cloud interaction and estimation of AIE on maritime as well as continental clouds have been conducted worldwide. However, magnitude of this AIE still remains uncertain because of the complexity in the estimation (*Menon et al., 2008*).

In the estimation of AIE_n in many numerical models, the effect of dispersion is not considered, but the field studies of the indirect aerosol effect show that polluted marine clouds and clouds of continental origin generally have both larger number concentration and a larger relative dispersion (ϵ) of the droplet size distribution relative to clean marine clouds (Liu and Daum, 2002). Hudson and Yum (1997) reported that the spectral width (σ) of cloud droplet spectra was greater with higher N_{ccn} from the Atlantic Stratocumulus Transition Experiment (ASTEX) conducted in 1992. They also observed that this small σ difference can be noticed if cloud parcels with droplets of same mean diameter were considered. Liu and Daum (2002) shows the relation between relative dispersion of cloud droplet size distribution and number concentration of cloud droplets from 13 different experiments, of which 11 cases show higher relative dispersion with higher N_{ccn} , and a slight decrease in the other two.

Pandithurai et al. (2012) reported aerosol-cloud relationships and estimated AIE for warm continental cumuli over Indian sub-continent using Cloud Aerosol Interactions and Precipitation Enhancement Experiment (CAIPEEX) aircraft campaign data sets and observed influence of higher Cloud Condensation Nuclei (CCN) concentration on cloud droplet dispersion as it is significantly offsetting AIE over continental clouds. They also indicated that the dispersion effect may be implicitly included in the estimation of AIEs derived using relative changes in cloud effective radius (R_{eff}) with relative changes in aerosol concentration (N_{acc}). Hudson and Noble (2013) reported that the increase in relative dispersion is largely due to decrease in droplet mean radius rather than increase in spectral width. However, Liu et al. (2013), Pandithurai et al. (2012) and the present study showed that the spectral width of droplet spectra shows increase if we consider a fixed mean droplet radius. Martins et al. (2009) showed that the increase in aerosol loading due to biomass burning aerosols decreases the spectral width and in turn enhances the Twomy cooling effect. These contrasting reports suggest that more work is needed on this topic.

Aerosol semi-direct effect also plays an important role as absorbing aerosols enhances the atmospheric heating due to absorption of solar radiation, which in turn changes the relative humidity and thereby inhibit cloud development and burn-off the clouds (Huang et al., 2014). Since the percentage/fraction of absorbing aerosol at this site during study period (monsoon season) is very less compared to scattering type aerosol, which is detailed in section 2, the semi-direct effect due to absorbing aerosol (eg., BC) is comparatively very less. So the aerosol semi-direct effect is not dealt in this study.

Thus in the present work aerosol effects on cloud microphysical properties from collocated simultaneous measurements over Mahabaleshwar, a Western Ghat region in the Indian state of Maharashtra have been

studied. Detailed analysis of AIE and its offset due to dispersion effect (DE) at different LWC regimes are presented.

2. Data Used

5 The recently set up High Altitude Cloud Physics Laboratory (HACPL) at Mahabaleshwar (17.56⁰ N, 73.4⁰ E, 1348 m AMSL) has various ground based instruments for observation of aerosol as well as clouds. The observatory is situated in the Western Ghats, wherein during monsoon season the temperature is varied between 17 and 23°C and relative humidity varied between 85- 100% with an average rainfall of 500 cm/yr. It is also observed that the site is covered by warm continental clouds most of the time during the summer monsoon
10 season. Interestingly, observation from laboratory have shown that the total aerosol concentration in the size range (5 nm to 30 µm) varies from 100 to 25000 cm⁻³ and occasionally it reached up to 40000 particles/cm³ and CCN concentration from 400 to 20000 cm⁻³. The aerosol and CCN concentration measurement shows that the region experiences higher aerosol concentrations during monsoon season in spite of washout/scavenging due to precipitation (Leena et al., 2015). The chemical composition of sub-micron aerosols over this site during
15 monsoon season is found to be dominated by organics (77%), sulphates (14%), chlorides (4%), and nitrates. Significant fraction of secondary organic aerosols could be formed from VOCs emitted from the wet vegetation. Also, the monsoon winds can bring significant amount coarse sea salt particles.

In this work, we have made an attempt to study the influence of aerosol on cloud microphysical properties and their relationship using data collected during 2013 monsoon season. We have related cloud microphysical
20 properties such as cloud droplet number concentration (CDNC) and cloud droplet effective radius (R_{eff}) to CCN concentration (N_{ccn}) measured simultaneously by using various ground based instruments and the details are described below. It is to be noted that for the present study data considered were explicitly for non rainy conditions, cloud probes were operated only during non-rainy cloudy conditions and possibility of even small drizzles were eliminated by comparing the data with rain rate obtained from impact disdrometer. After
25 scrutinizing the entire data set, about 20 hours of data were considered for analysis as detailed in Table 1.

2.1. CCN counter

The CCN counter, which is a continuous flow thermal gradient instrument (Roberts and Nenes, 2005) manufactured by Droplet Measurement Technologies (DMT Inc.) was used for CCN concentration measurement. It was configured to operate at a fixed super saturation (SS) of 0.6% and sampling rate of 1 Hz.
30 This CCN counter has a thermal gradient diffusion chamber in which a super saturated water vapour condition is

created. This supersaturated water vapour condenses on the CCN in the sample air to form droplets. An optical particle counter (OPC) using side scattering technology counts and sizes the activated drops.

2.2. Cloud Combination Probe

Cloud microphysical parameters were collected with a Cloud Combination Probe (CCP) from DMT Inc, which is a combination of Cloud Droplet probe (CDP) and a hotwire liquid water content probe (HW-LWC). The CDP measures the cloud droplet size distribution (DSD) and concentration of droplet sizes from 3 to 50 μm , categorized into 30 channels. The CDP uses a laser to illuminate particles and forward-scatter their light. The detected scattered light is then used to size the particles. The intensity of the scattered light depends upon the size, composition, and shape of the particle. LWC was measured using the hotwire probe and also estimated from CDP drop size distribution measurements. For calculating spectral dispersion in cloud droplet size distribution and to estimate AIE, Cloud droplet mean radius (r_m) spectral width of DSD (σ) and relative dispersion (ϵ) were calculated from the data obtained from CDP. The mathematical definition of r_m , σ and ϵ is as follows

$$r_m = \frac{\sum_{i=1}^n N_i r_i}{N}$$

$$\sigma = \left[\frac{\sum_{i=1}^n N_i (r_i - r_m)^2}{N} \right]^{1/2}$$

$$\epsilon = \sigma / r_m$$

where

N_i = droplet concentration within channel i

r_i = midpoint radius for size channel i

N = total droplet concentration

3. Analysis Procedure

As per Twomey's hypothesis, cloud droplet number concentration increases and cloud droplet size decreases with an increase in aerosol concentration for a fixed liquid water path (LWP). AIE can be estimated by utilizing both cloud droplet number (AIE_n) and droplet effective diameter (AIE_s) (Feingold *et al.*, 2003) through the following expressions:

$$AIE_n = \frac{1}{3} \frac{\Delta \log N_c}{\Delta \log \alpha} \dots (1) \quad AIE_s = -\frac{\Delta \log R_{eff}}{\Delta \log \alpha} \dots (2)$$

where,

5 ΔN_c = the relative change in cloud droplet number concentration,

R_{eff} = the cloud drop effective radius and

α = Aerosol or CCN concentration

R_{eff} in the above relation is the ratio between third and second moment of CDP measured cloud droplet size distribution (*Hansen and Travis., 1974*). The R_{eff} , is an important parameter in aerosol-cloud interaction studies,

10 and can be expressed as

$$R_{eff} = \frac{\sum_{i=1}^n p_i r_i^3}{\sum_{i=1}^n p_i r_i^2} \dots (3)$$

where n is the number of droplet sizing bins (30 bins for CDP), p_i is the particle count for bin i and r_i is the

15 mean radius in μm of bin i . For R_{eff} parameterization, climate models use a 1/3 power law which relates cloud liquid water content (LWC) and number concentration (N_c) (*Slingo, 1990; Bower et al., 1994; Liu et al., 2006*), and the same can be written as

$$R_{eff} = \beta \left(\frac{3LWC}{4\pi\rho_w N_c} \right)^{\frac{1}{3}} \dots (4)$$

20 where

ρ_w = the water density

β =effective radius ratio, which is a function of the spectral shape of the cloud droplet size distribution.

Previous studies show that considering the dispersion effect is important for estimating AIE (*Liu et al., 2008; Peng and Lohmann, 2013*).

Liu and Daum (2000) and Liu et al. (2002) put forward an expression corresponding to a Gamma distribution to describe the β dependence on spectral droplet size distribution, which is as follows

$$\beta = \frac{(1 + 2\varepsilon^2)^{2/3}}{(1 + \varepsilon^2)^{1/3}} \dots (5)$$

5 where ‘ ε ’ is the relative dispersion of the droplet size distribution and can be defined as the ratio of the standard deviation to the mean radius of the droplet size distribution. Liu et al., (2008) studied the relation between β and (L/N_c) (water per droplet or specific cloud water content) to avoid uncertainties in the existing $\beta(\varepsilon)$ - N_c relation to explain the dispersion effect. The relation between β and (L/N_c) can be expressed as

$$\beta = \alpha_\beta \left(\frac{L}{N_c} \right)^{-b_\beta} \dots (6)$$

10 where α_β is the intercept of the power law fit between β and L/N , b_β is the dispersion factor defined as the percentage of the offset in the Twomey cooling effect, due to the dispersion in the cloud droplet size distribution.

4. Results and Discussion

15 To understand the interaction of aerosol with cloud microphysical parameters, cloud droplet number concentration (CDNC) and its effective radius (ED) against CCN concentration were plotted and displayed in Figure 1. It can be noted from this figure that higher CCN causes more number of cloud droplets. At the same time higher droplet number regime is having a smaller effective droplet diameter. For estimating AIE, CCN concentration, cloud droplet number concentration and droplet effective diameter at different liquid water contents were examined. Figure 2 depicts the relation between the cloud microphysical parameters, CDNC, Droplet Effective Diameter (ED) and LWC. It clearly shows the reduction in the ED with increase in CDNC for all LWC values. Sharing the same available liquid water content by more cloud droplets leads to reduction in its size.

4.1. AIE estimation from droplet effective diameter and number concentration

25 It is to be noted that the correlation between CCN & CDNC and CCN & R_{eff} are statistically significant for all LWC bins considered in this study. The significant test for CCN versus CDNC and CCN versus R_{eff} for

different LWC bins can be seen that they are statistically significant with strong confidence level. Figure 3a shows the variation of CDNC with CCN concentration for three different narrow LWC bins of 0.20, 0.21 and 0.22 gm^{-3} . It shows an increase in CDNC with increasing in CCN concentration. The value exponent of power law fit between CDNC and CCN were 0.24, 0.23 and 0.22. This results into AIE_n estimates of 0.08 and 0.076 and 0.074 respectively.

Analysis of the variation of R_{eff} with CCN concentration (Figure 3b) shows that the reduction in R_{eff} with increase in CCN concentration. The linear fit to the log-log plot of R_{eff} and CCN shows a good correlation for each individual LWC bin where the value of exponent (AIEs) varies as -0.055, -0.051 and -0.050, respectively for LWC bins of 0.20, 0.21 and 0.22 gm^{-3} . From this, it is clear that increased CCN concentration led to more number of cloud droplets of smaller size, which is generally termed as aerosol indirect effect, represented herein by AIE_s . It can be noted that AIE_n values are higher about 30-40% as compared to AIE_s . This clearly suggests that the overestimate of AIE_n may be due to some other effects such as dispersion and entrainment. Some of the previous investigations of AIE_n using aircraft measurements generally relate sub-cloud aerosol measurements with in-cloud droplet number concentrations without grouping it into constant LWC bins and attributed the overestimation to entrainment mixing processes (Shao and Liu, 2009). The systematic discrepancy between AIE_n and AIE_s is caused primarily by the differential loss of cloud liquid water between clean and polluted clouds (Shao and Liu, 2006). Previous studies have also shown that droplet dispersion effect can partly offset the Twomey's cooling effect. However, there is no clear demonstration of observed differences between AIE_n and AIE_s . This warrants the study why AIE_n overestimate AIE_s . This study clearly provides an observational demonstration of overestimation of AIE_n as compared to AIEs and the droplet dispersion effect offset brings down the AIE_n close to AIE_s .

We have analyzed the LWC variation of non-precipitating clouds for the period considered in this analysis and presented in Figure 4 as frequency distribution of LWC. It can be seen that it is varied from 0-1 gm^{-3} with a maximum percentage occurrence is between 0.05 to 0.5 gm^{-3} . Thus considering this, aerosol indirect effect in terms of number concentration and droplet size has been calculated for each bin of liquid water content. From Figure 5, it is observed that beyond 0.35 gm^{-3} of LWC, the variability of the values seems to be high due to less availability of data and lower statistical significance which is separated by a dotted line in the figure. For present analysis of AIE, CCN and cloud microphysical properties (CDNC, D_{eff}) were first grouped by LWC for which a bin size of 0.01 gm^{-3} LWC was selected. In Figure 5, the aerosol indirect effect and dispersion factor for the

mentioned LWC ranges is represented, in which green line corresponds to AIE_s , black line corresponds to AIE_n and blue line shows dispersion factor.

The estimated AIE shows its maximum value at lower LWC regime, reduced to half in the moderate LWC regime and falls to minimum at higher LWC range. In this observation, the maximum AIE estimated from droplet concentration and size is 0.103 (AIE_n) and 0.088 (AIE_s), respectively. *Pandithurai et al. (2012)* using CAIPEEX aircraft data, reported AIE over Indian region from both methods are found to be 0.13 (AIE_n) and 0.07 (AIE_s). In the lower LWC regime, reduced collision coalescence may cause a large number of smaller droplets with almost same size, which increases the AIE and reduces the dispersion factor.

10 4.2. Dispersion Effect

It is well known from earlier studies that droplet relative dispersion (ϵ) is an important cloud microphysical parameter which is defined as the ratio of spectral width of cloud droplet size distribution (σ) to mean radius (r_m). *Liu and Daum (2002)* noted that higher CCN concentration will cause broader cloud droplet spectra, which will reduce cloud albedo thus tending to reduce the AIE. From Figure 6a, it can be seen that the relative dispersion is increasing with CCN concentration which is similar to the previous studies (*Liu and Daum, 2002; Pandithurai et al., 2012*).

Apart from this, Figure 6b shows the observed variation of σ with CCN concentration, for fixed mean radius for three narrow ranges 4 - 4.5 μm , 4.5 - 5 μm and 5 - 5.5 μm , in which σ is increasing with N_{ccn} . This observation is similar to that reported by previous investigators through aircraft measurements (*Hudson and Yum, 1997; Miles et al., 2000; Pandithurai et al., 2012*). *Hudson and Yum (1997)* demonstrated that standard deviation of cloud droplet spectra will be higher at higher CCN concentration from the ASTEX experiment of 1992. In general, clean maritime clouds have larger droplet size and larger σ , while polluted continental clouds have smaller drops with smaller σ . According to *Yum and Hudson (2005)*, the large dispersion observed in the continental polluted cloud was possibly due to smaller droplets rather than broader droplet spectrum. From this observation, we can see that the increase in relative dispersion in polluted continental clouds is not only because of the reduction in droplet radius at higher N_{ccn} but also due to increase in σ . According to *Liu and Daum (2002)*, anthropogenic aerosols have a complex chemical composition which leads to broader activated drop size distribution, and those small droplets in the cloud compete for water vapor and broaden the size spectrum.

Further, effective radius ratio, β , which is an increasing function of relative dispersion, ϵ of cloud droplet. The value of β varied from 1.06 to 1.28. Similarly the value of ϵ varied from 0.27 to 0.59. Higher ϵ corresponds to

higher CCN concentration. Figure 7 shows the relation between water per droplet (L/N) and effective radius ratio, β . It is found that β decreases with L/N and the linear fit to the data gives the following relation with a correlation coefficient of 0.73. The dispersion factor estimated from whole data set is 0.07. A value of $b_{\beta}=0.07$ indicates that the dispersion can offset AIE by 21% (3×0.07).

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4.3. AIE estimation considering dispersion effect.

Earlier studies (*Peng and Lohmann, 2003; Liu et al., 2008 and Liu and Daum, 2002*) have reported that the dispersion effect tends to offset the AIE in the range from 10 to 80%. On the contrary, *Martins et al. (2009)* reported that the dispersion effect enhances the Twomey cooling for biomass burning aerosols in the dry region.

10 *Shao and Liu (2006)* proposed that the real strength of the AIE should be sum of the Twomey's cooling effect and the compensating effects such as droplet dispersion effect. In most of the previous works, the AIE is calculated mainly by one of the methods either aerosol versus cloud droplet number or aerosol versus R_{eff} . Few studies reported accurate representation of AIE needs to consider i) dispersion effect, ii) entrainment effects. In this study, we present both the methods and systematic difference which can be minimized by considering the dispersion effect. From this study, the maximum dispersion factor b_{β} found to be 0.098 for LWC of 0.21 gm^{-3} , the corresponding AIE_n was 0.077, the dispersion effect is found to be -0.023, which can offset the AIE_n effect by 29.6%, so the resultant AIE_n is 0.054 ($0.077 - 0.023$). This resultant AIE_n is close to the estimated AIE_s (0.051) using R_{eff} for the same LWC bin. From this, we can clearly state that dispersion effect is implicitly included in the estimation of AIE_s . This analysis is presented in Figure 8 which shows the variation of AIE_s and resultant AIE_n , i.e., $\text{AIE}_n + \text{dispersion effect}$ for different LWC regimes along with dispersion offset in percentage. Dispersion effect is very less at lower LWC regime where AIE has its maximum value; and tends to increase with LWC and shows maximum at moderate LWC regime then found to be reducing at higher LWC regime. From the figure we can see that resultant AIE_n is closely agreeing with AIE_s for all LWC range except in the lower LWC where dispersion effect is too small, and the AIE has its maximum value.

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5. Summary and Conclusion

Aerosol indirect and dispersion effects on monsoon clouds over a high altitude site in Western Ghats using in-situ instruments were studied. Advantage of simultaneous measurement of aerosol and cloud microphysical properties have been utilized from this observatory and studied i) discrepancy between aerosol indirect effect estimates from different methods, ii) the variation in albedo susceptibility with LWC, iii) the dispersion effect

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variation with LWC and the resultant AIE. Over Indian subcontinent, the relative dispersion found to increase with aerosol concentration in continental clouds sampled through aircraft measurements over several regions during the CAIPEEX experiment (*Pandithurai et al., 2012; Prabha et al., 2012*). Higher concentration of anthropogenic aerosols with different chemical composition and sources can contribute to cloud droplets of different sizes. It was also observed that an increase in spectral width of the drop size distribution (σ) with N_{ccn} for a given mean cloud droplet radius, indicating a broader spectrum at high aerosol concentration. As *Liu* (2002) suggested, this increasing tendency of σ and hence ϵ might be because of several possible reasons, such as mixing of clouds at different developmental stages, or may be due to strong collision-coalescence.

The AIE found to be maximum in low liquid water content clouds but at the same time dispersion of cloud droplet size distribution is very less. While considering clouds with moderate liquid water content we can see an increase in dispersion effect which offsets the AIE. At higher LWC, the dispersion as well as AIE seems to be reducing. In this study, we investigated the effect of aerosol on cloud effective radius and droplet number concentration. The estimated aerosol indirect effect derived from effective radius changes (i.e., AIE_s), the maximum observed is 0.088, and effect on droplet concentration, AIE_n , the maximum found is 0.103, similarly the maximum dispersion offset obtained is 29.6%. From the resultant AIE_n , it is clear that in AIE estimate using droplet concentration, dispersion effect should be taken in to account for correct estimate of AIE. It is also confirmed that AIE estimation from effective droplet radius implicitly includes dispersion effect. As the droplet effective radius is a ratio of third and second moment of DSD, the dispersion in DSD is implicitly included and hence the AIE estimates obtained through relative changes in observed R_{eff} does not need to correct for the dispersion effect.

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Table 1. Data sampling dates/periods used in this study during which no precipitation was recorded.

Date	From	To	Total (minutes)
06/06/2013	16:07	16:40	33
11/06/2013	8:38	9:13	35
12/06/2013	9:10	12:37	207
23/06/2013	9:54	10:59	65
15/07/2013	22:35	22:51	16
25/07/2013	9:53	10:37	41
07/08/2013	19:51	22:40	169
16/08/2013	10:04	17:19	75
18/08/2013	18:02 21:00	20:39 23:02	279
19/08/2013	19:58	23:01	183
20/08/2013	22:04	23:31	87
TOTAL			1190

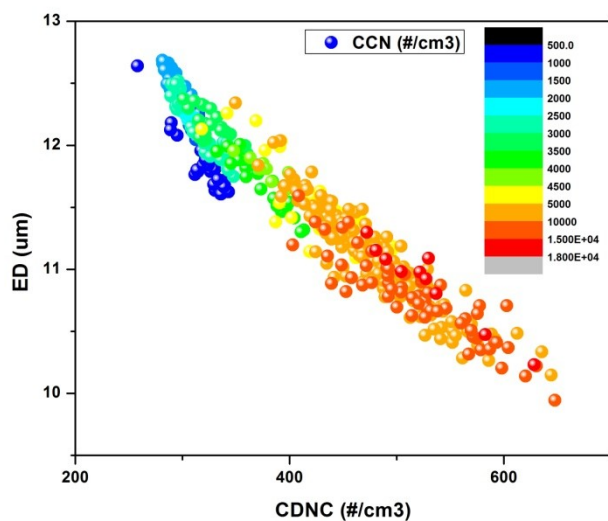


Figure 1 (a). Variation of cloud microphysical parameter (droplet number concentration and effective diameter) with CCN number concentration.

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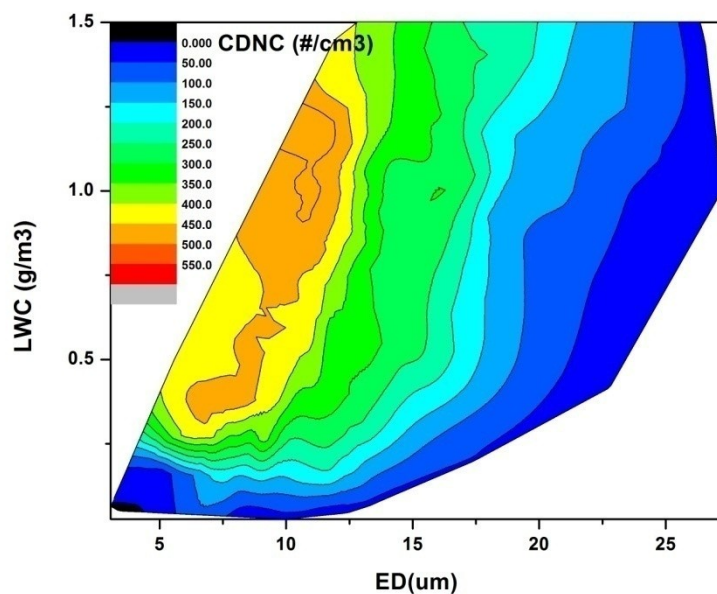
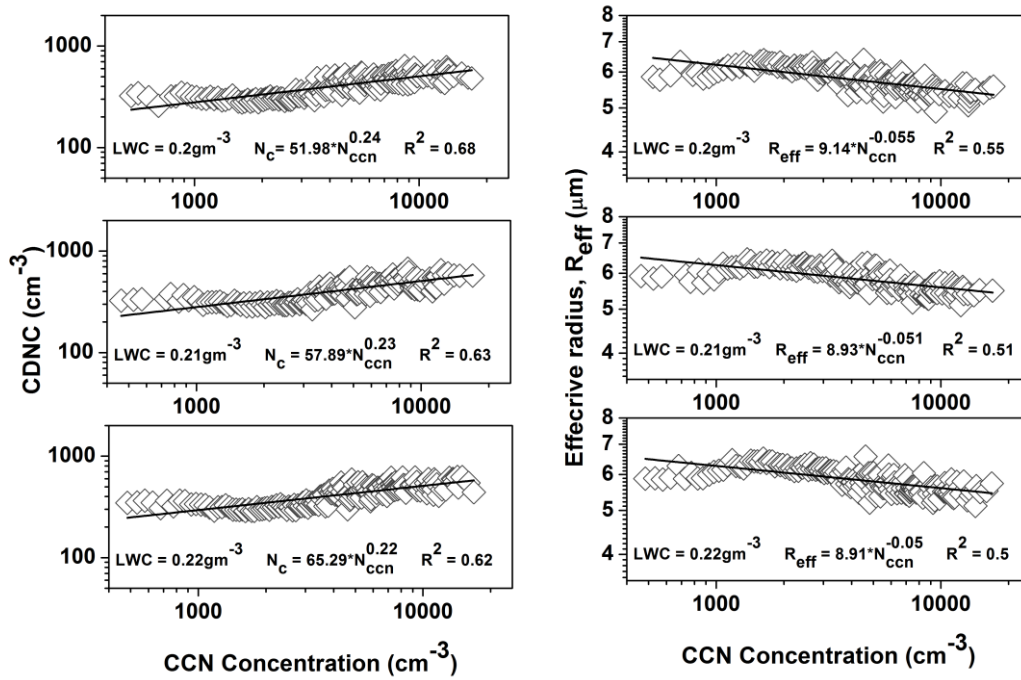


Figure 2. The contour plot shows relationship between cloud micro physical parameters, cloud droplet number concentration (CDNC), droplet effective diameter (ED) and LWC . ED decreases with increase in CDNC for all LWC.

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15 **Figure 3 a and b.** Scatterplot showing relation between cloud condensation nuclei (CCN) concentration measured at 0.6% supersaturation and cloud droplet number concentration (CDNC) and CCN concentration versus cloud effective radius (R_{eff}) for three LWC bins (0.20, 0.21 and 0.22 gm⁻³).

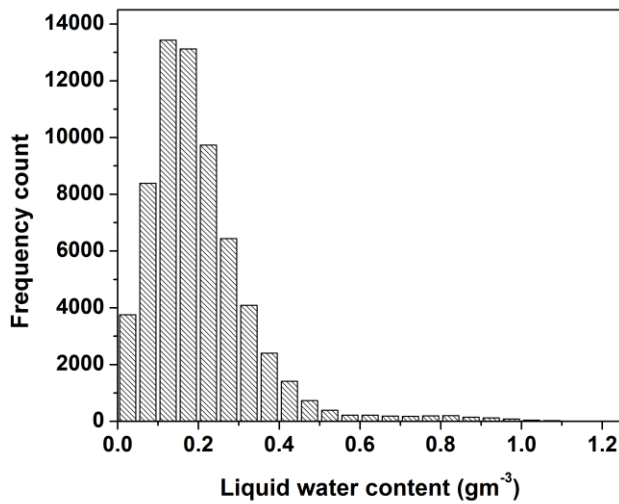


Figure 4. Frequency distribution plot of the measured LWC data.

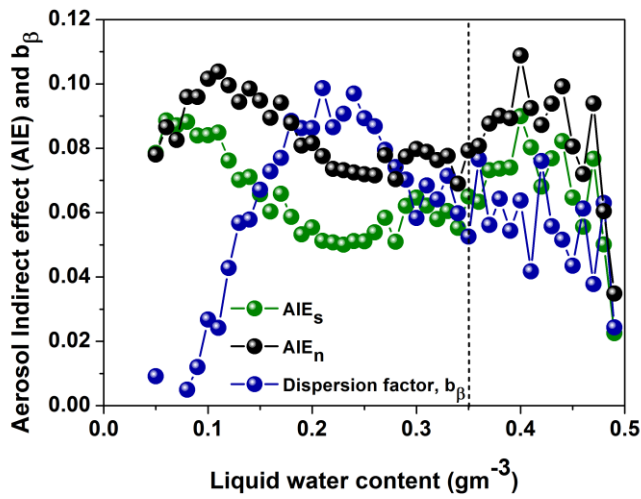


Figure 5. Aerosol indirect effect estimates using AIE_n , AIE_s and dispersion factor for the LWC ranges is represented. . Green, Blue and Green lines corresponds to AIE_s , AIE_n and dispersion factor respectively.

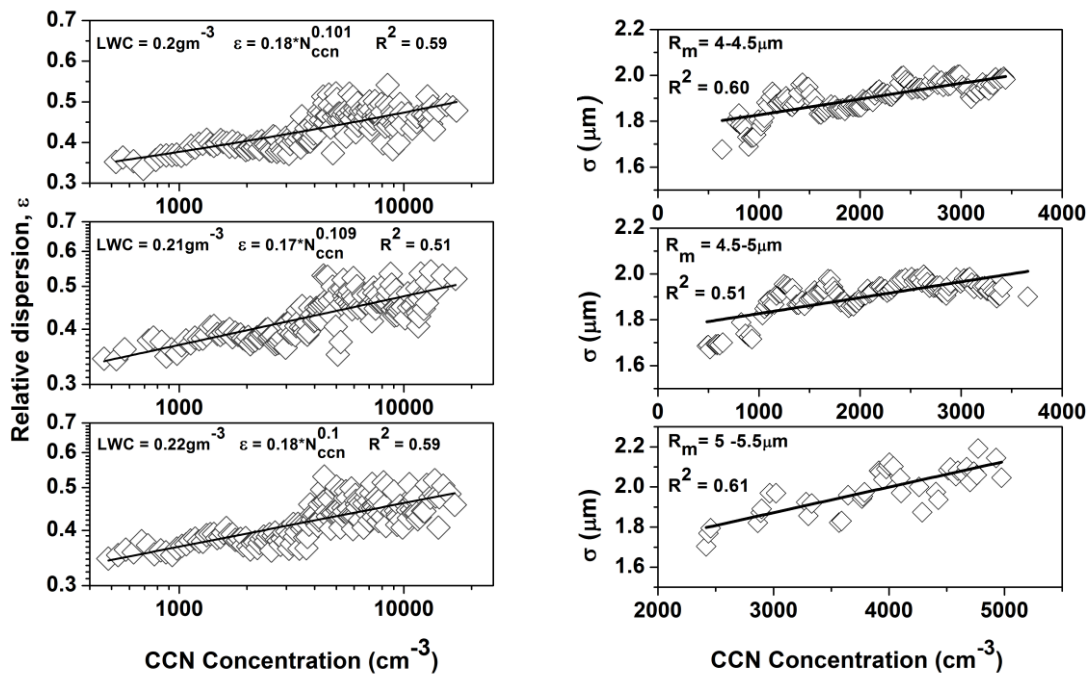


Figure 6a and 6b. Scatterplots showing the changes in relative dispersion (ϵ) and spectral width of cloud droplet (σ) with CCN number concentration.

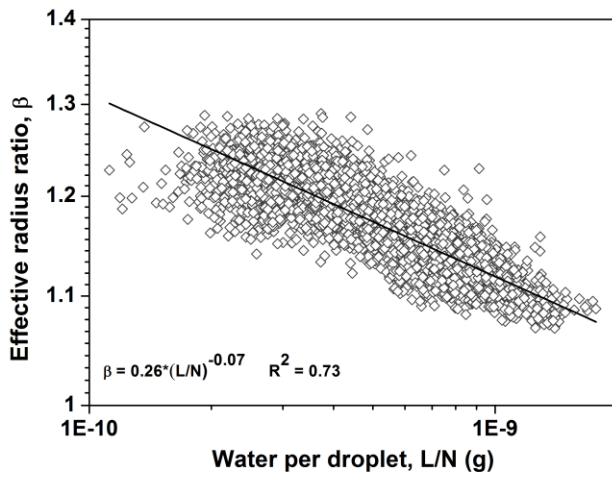


Figure 7. Scatterplot between water per droplet (L/N) and effective radius ratio (β).

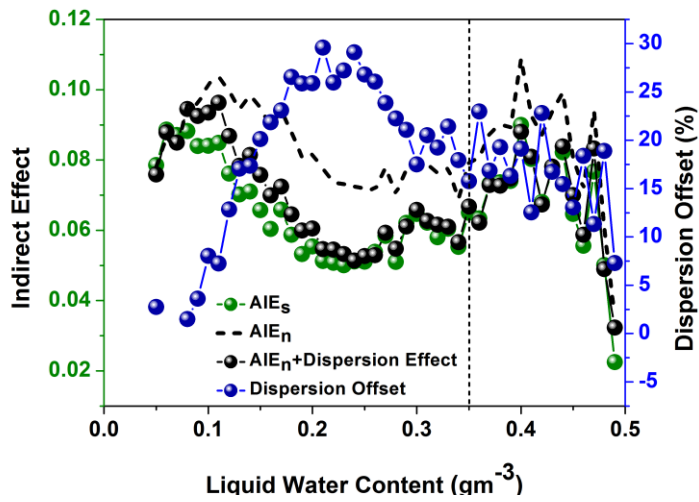


Figure 8. Aerosol indirect effect estimated after considering dispersion effect in AIE_n , Left axis represents AIE, right axis represents dispersion offset in percentage.