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# Long-term observation of aerosol–cloud relationships in the Mid-Atlantic of the United States

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## Abstract

Long-term ground-based observations (2006 to 2010) of aerosol and cloud properties derived from passive radiometric sensors deployed at an atmospheric measurement field station in the Baltimore–Washington corridor operated by Howard University were used to examine aerosol indirect effect on cloud optical depth (COD), liquid water path (LWP), cloud droplets effective radius ( $R_e$ ) and cloud droplets number concentration ( $N_d$ ). A higher frequency of clouds with large COD ( $> 20$ ) and small  $R_e$  ( $< 7 \mu\text{m}$ ) was found during summer of 2006 and 2007 along with higher frequency of abundant aerosol loading. The five-year data are screened for summer months only and are separated into clean and polluted cases based on aerosol particulate matter with aerodynamic diameter  $\leq 2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ) value. Evidence of aerosol indirect effect is found where for polluted cases the mean and median values of COD and  $N_d$  distributions were elevated while the mean and median values of  $R_e$  were decreased. Further reinforcing this conclusion is the result that the mean and median values of LWP distributions did not show prominent difference between clean and polluted cases, this implies that differences between the two cases of influential factors on cloud properties were relatively controlled. Moreover aerosol indirect effects were found insignificant when LWP was small but significant when LWP was large through the analysis of sensitivity of  $N_d$  to LWP under different aerosol loading and the measurements of aerosol size distribution.

## 1 Introduction

The importance of aerosol-cloud interaction has been recognized for a long time, as the aerosol indirect effect (AIE) on cloud radiative properties is crucial to atmospheric energy balance (IPCC, 2007; Twomey, 1974). A growing list of studies have employed space- and ground-based observations to provide convincing evidence of AIE: that high aerosol concentration tend to increase concentration of cloud droplets, reduce

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cloud droplet effective radius ( $R_e$ ) and then increase cloud albedo (Twomey, 1977 the aerosol first indirect effect). High aerosol concentration tends to increase concentration of cloud droplets, reduce cloud droplet effective radius ( $R_e$ ) and then increase cloud albedo (Twomey, 1977 the aerosol first indirect effect). By using satellite remote sensing, Han et al. (1998) showed that cloud-droplet concentrations correlate to the CCN at different regions. Feingold et al. (2001) defined the ratio of logarithmic  $R_e$  and aerosol optical depth (AOD) to stand for the AIE and Feingold et al. (2003, 2006) reported the observed AIE by using ground-based observations of cloud and aerosol properties provided by the Atmospheric Radiation Measurement (ARM) at the Southern Great Plains (SGP) site in Oklahoma. Also with ARM SGP observations Kim et al. (2003, 2008, 2012) demonstrated the positive relationship between cloud optical depth (COD) and LWP, an inverse relationship between  $R_e$  and aerosol scattering coefficient and the role of adiabaticity in AIE. Nzeffe et al. (2008) showed that  $R_e$  reduced under polluted air-masses for given LWP based on ground-based observations from Howard University Beltsville Campus (HUBC) facility.

The complexity of the climate system and the inadequacy of measurements and methodologies have made it very challenging to obtain a more detailed understanding of AIE (McComiskey and Feingold, 2012; Stevens and Feingold, 2009). For example the question of whether increased aerosol loading may enhance or suppress cloud LWP remains unresolved in the literature despite the evidence of aerosol impact on  $R_e$  (Albrecht, 1989; Ackerman et al., 2004; Storelvmo et al., 2006; Lee et al., 2009; Lebsock et al., 2008; Zheng et al., 2010; Coakley and Walsh, 2002; Brioude et al., 2009). Part of the challenge is reducing the uncertainty in estimates of AIE, which as pointed out by McComiskey and Feingold (2012) requires “process scale” studies. Though field experiments that produce “process scale” observations of aerosol-cloud interaction continue to occur, historically they have been insufficient in number, regional diversity and duration.

In this study, ground-based observations over a period of five years (from 2006 to 2010) are used from an atmospheric measurement field station (39.054° N and

76.877° W) at HUBC. Analysis of derived AOD from a nearby Aerosol Robotic Network (AERONET) (Holben et al., 1998) and a collocated air quality monitoring station operated by the Maryland Department of the Environment (MDE) showed that this region experiences frequent episodes of elevated anthropogenic aerosol loading associated with urban pollution conditions during summer months. Nzeffe et al. (2008) reported that the air flow across the region under polluted condition is predominantly from west-southwest based on back trajectory analysis. They found that polluted aerosols reduced cloud effective droplet size from analysis of six months of observations of aerosol and cloud properties in 2005. This study extends the work of Nzeffe et al. (2008) to systematically investigate aerosol impacts on COD, LWP,  $R_e$  and cloud droplets number concentration ( $N_d$ ) from five years of ground-based observations during summer.

## 2 Measurements

The Howard University Beltsville Campus facility in Beltsville, MD is situated in a rural-suburban transition region between Washington, DC and Baltimore, MD urban centers. It has a wide range of sensors deployed to observe atmospheric radiation, surface fluxes, aerosol, cloud properties and other climate and weather processes (Nzeffe et al., 2008). The MultiFilter Rotating Shadowband Radiometer (MFRSR) is a sensor with a rotating shadow band, measuring global downwelling irradiance, diffuse irradiance and direct beam irradiance calculated from global and diffuse irradiance. More detail on the MFRSR can be found in Harrison et al. (1994). It is calibrated using data acquired on clear sky days via the Langley regression which is based on linear regressions of the log of direct beam irradiance vs. airmass (Harrison et al., 1994; Harrison and Michalsky, 1994). AOD can be derived through Langley regression and the calibration constant  $I_0$  is used to compute transmittances during cloudy conditions and then infer COD (Min and Harrison, 1996). LWP is retrieved from brightness temperature observations from the dual frequency (23.8 and 31.4 GHz) Microwave radiometer (MWR) (Westwater et al., 2001). Based on Mie theory  $R_e$  is retrieved with COD by a Nonlinear

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Least Squares Method with measurements of LWP (Min and Harrison, 1996; Min et al., 2004; Nzeffe et al., 2008); the  $N_d$  is obtained from a parameterization in terms of COD and  $R_e$  (Boers et al., 2006; Bennartz, 2007; Min et al., 2012). Since AOD cannot be retrieved from the MFRSR during cloud periods, the daily averaged AOD during clear-sky periods within the day is used. The AOD retrieved from the MFRSR at the HUBC site is consistent with that from AERONET observation at NASA Goddard Space Flight Center (GSFC), around 5 miles southeast of the HUBC facility with correlation coefficient of 0.94 during the study period (from 2006 January to 2010 December). Particulate matter with aerodynamic diameter  $\leq 2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ) data obtained from samplers operated by the MDE at the HUBC site are also used in this study to estimate aerosol loading under cloudy conditions and the measured  $\text{PM}_{2.5}$  values during clear-sky conditions have a good positive relationship with MFRSR retrieved AOD with correlation coefficient of 0.67.

This investigation is limited to summer months (June, July and August) because during this time the Mid-Atlantic region experiences the largest variation of aerosol loading due to episodic summertime pollution events. Strong convection that occurs during summer results in more boundary layer clouds, which are tightly coupled with surface aerosols. The inter-seasonal variation of aerosol loading is also large but so is the variation of dynamical and thermodynamic conditions that dominate cloud micro- and macro- physical properties. The latter effect on cloud properties could complicate the analysis of aerosol cloud interaction on this scale. For this reason the study is confined to the summer months. The other major constraint in the data analysis for this study is that cases with low LWP ( $< 40 \text{ g m}^{-2}$ ) are screened. Instrument uncertainty and retrieval errors associated with the climatological profiles and the microwave absorption model results in a large uncertainty of retrieved LWP between 20 and  $30 \text{ g m}^{-2}$  (Turner et al., 2007). For thin clouds the radiative flux is sensitive to the small changes of LWP (Min and Duan, 2005) and consequently the  $R_e$  retrieved from combination of MWR LWP and MFRSR COD has relatively larger uncertainties, mainly due to the uncertainty of MWR measurements (Min et al., 2003)

### 3 Results

Figure 1a displays the probability distribution for AOD for all years. It is apparent that more frequent episodes of high aerosol loading occur during the summers of 2006 and 2007 as compared to 2008, 2009 and 2010. Hereafter 2006 and 2007 are denoted as polluted years while 2008, 2009 and 2010 are denoted as clean years for convenience. The comparison of COD distributions is consistent with that of the AODs, where during the polluted years the fraction of COD larger than 20 is much larger than that during the clean years (Fig. 1b). In contrast, as shown in the distributions of LWP (Fig. 1c), more high LWP episodes occurred during the clean years than that during the polluted years. This is counter intuitive as it is expected that COD is proportional to LWP given understanding of atmospheric dynamic and convective processes. The ratio of LWP and COD can be approximated as proportional to  $\rho \cdot R_e$  ( $\rho$  is density of liquid water) with the assumption that liquid water content is constant or linear with height above the cloud base (Han et al., 1994; Wood and Hartmann, 2006). It suggests that larger COD in the polluted years is not due to the increase of LWP but rather due to microphysical changes in cloud effective radius. The result is further corroborated as the fraction of smaller  $R_e$  is found much larger during the polluted years (Fig. 1d). Therefore, the increased fraction of COD larger than 20 during the polluted years is likely related to enhanced aerosol loading episodes in the region.

Sun photometers, such as the MFRSR, cannot readily discern AOD from COD under cloudy conditions. Thus, in situ measurements from gravimetric samplers of  $PM_{2.5}$  are used instead to more carefully examine cloud properties under different levels of aerosol loading. The entire 5 years of summer observations of cloud properties (3 min average) have been segregated into a clean group ( $PM_{2.5} \leq 10 \mu\text{g m}^{-3}$ ) and a polluted group ( $PM_{2.5} \geq 30 \mu\text{g m}^{-3}$ ). To reduce the uncertainties in the retrieval of LWP and  $R_e$  and avoid contamination from precipitation events, only the data of cloud optical depth larger than 5, LWP between  $40 \text{ g m}^{-2}$  and  $180 \text{ g m}^{-2}$ , and  $R_e$  less than  $15 \mu\text{m}$  are used for the study; these are all for clouds lasting longer than thirty minutes (Min et al., 2003;

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Nzeffe et al., 2008). In addition to LWP and COD it is useful to also consider the impact on cloud droplet size distribution under different aerosol loading. The cloud droplet size distribution can be represented to the first order by  $R_e$  and droplet number concentration. Figure 2 shows that the distribution of number concentration and COD under the polluted conditions shifts to larger values than that under clean conditions. In the meantime, more clouds with small  $R_e$  occur under the polluted conditions. The average number concentration and COD are 60 %, and 17 % larger, respectively while the average  $R_e$  and LWP are 15 % and 4 % smaller, respectively under polluted condition as compared to clean condition. This result is consistent with the effect (Twomey, 1974) and supports the hypothesis of this study that more cloud droplets are formed under polluted conditions and the total LWP is not very sensitive to aerosol loading, resulting in the decrease of  $R_e$  and then the increase of COD.

Nzeffe et al. (2008) found that  $R_e$  is proportional to LWP, but this dependency is less in polluted airmasses as compared to clean airmass. Here the observed pairs are further segregated into three LWP ranges (90–100, 120–130 and 150–180 g m<sup>-2</sup>) to assess the dependency of AIE on LWP (Fig. 3a). A negative relationship between  $R_e$  and PM<sub>2.5</sub> is found by using linear regression under all three LWP ranges, with the slopes of -0.02, -0.04, and -0.12 and correlation coefficients of -0.11, -0.25 and -0.63, respectively. This is consistent with AIE. The slopes (absolute value) increase with increasing LWP. This result suggests that AIE is more prominent for larger LWP clouds, which are formed under dynamical conditions that produce an abundant supply of moisture. Note also that the spread of  $R_e$  values decreases with increasing PM<sub>2.5</sub>. There is less competition among droplets at lower cloud condensation nuclei concentration thus a wider spectrum of droplets are activated and grow into cloud droplets. This is in contrast to higher CCN concentration where the competition limits the spectral range of droplet activation. In addition to the effective radius it is also important to evaluate the impact of aerosol loading on integrated cloud droplet number concentration (ICDN) and how that changes with LWP. As expected, (Fig. 3b) ICDN is proportional to LWP but is less so when aerosol loading is small (PM<sub>2.5</sub> < 10 µg cm<sup>3</sup>) as compared to when

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aerosol loading is large ( $PM_{2.5} > 30 \mu\text{g cm}^{-3}$ ). Cloud droplet concentrations become noticeably elevated when LWP is above  $150 \text{g m}^{-2}$  for enhanced aerosol loading. Under clean conditions with limited CCN, most CCN are likely activated into cloud droplets with sufficient but relatively low supersaturation; in this case cloud droplet growth dominates activation in development of the cloud. On the other hand, the number of cloud droplets that are activated and grow under polluted conditions with abundant CCN are limited by the degree of supersaturation that exist. This is due to competition among droplets in the nucleation and growth process which is based on their size and hygroscopicity. Thus, as supersaturation increases more CCN will nucleate and grow leading to a larger number of cloud droplets (large LWP), but  $R_e$  for these cloud droplets will be limited or small as compared to less polluted conditions with an equivalent level of supersaturation given the greater competition among the droplets to nucleate and grow.

In situ observations of aerosol size distributions are used to illustrate aerosol size distributions under clean and polluted conditions which are measured by a Fast Mobility Particle Spectrometer (FMPS) at HUBC during the NASA DISCOVER AQ field campaign (CrumeYrolle et al., 2014) (Fig. 4). On a polluted day (20 July 2011), the total number concentration of observed aerosols is much higher than that on a clean day (14 July 2011) and it is almost entirely due to the higher number concentration of smaller particles. Unfortunately the field campaign only last one month at HUBC. The detailed aerosol size distribution observations suggest that the increase of  $PM_{2.5}$  in this area may be mainly due to the increase of smaller size particles. Aerosol optical properties are related to aerosol size distribution and aerosol angstrom coefficient decreases with increasing aerosol size. The long-term observations of aerosol optical properties and  $PM_{2.5}$  show that the observed daily angstrom coefficients have positive relationship with  $PM_{2.5}$  (Fig. 5). This correlation implies that the increase of  $PM_{2.5}$  is mainly due to increase of smaller size particles over the region which is consistent with the detailed size distributions shown in Fig. 4. More aerosol particles can supply more CCN, however activation of smaller nuclei is more difficult under low supersaturation



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under conditions of low aerosol loading most of the available CCN are quickly activated into droplets. Thus increasing LWP in these clouds favors droplet growth over droplet activation. Alternatively, competition among droplets to activate in air with limited supersaturation causes clouds that form under high aerosol loading but low LWP (below  $100 \text{ g}^{-2}$ ) to not have significantly higher ICDN or lower  $R_e$  as compared to those that form under lower aerosol loading. As LWP increases (associated with higher supersaturation) in these clouds, however, competition among droplets reduces and activation dominates growth, resulting in larger ICDN and lower  $R_e$  relative to low aerosol loading at similar levels of LWP. This assumes that LWP correlates with supersaturation which depends on the strength of updraft velocities. These results are further confirmed by in situ observation of aerosol size distribution. The observations show that increase of  $\text{PM}_{2.5}$  in this region is mainly due to increase of small size particles. Given that small particles require higher levels of supersaturation to activate relative to large particle, it follows from the in situ data that ICDNs under polluted conditions is similar to the ICDNs under clean conditions when LWP is smaller than  $100 \text{ g m}^{-1}$ . The analysis based on the long-term observations including diverse dynamical and aerosol regimes provides climate assessment of AIE in the mid-Atlantic corridor, where frequent severe pollution episodes occur.

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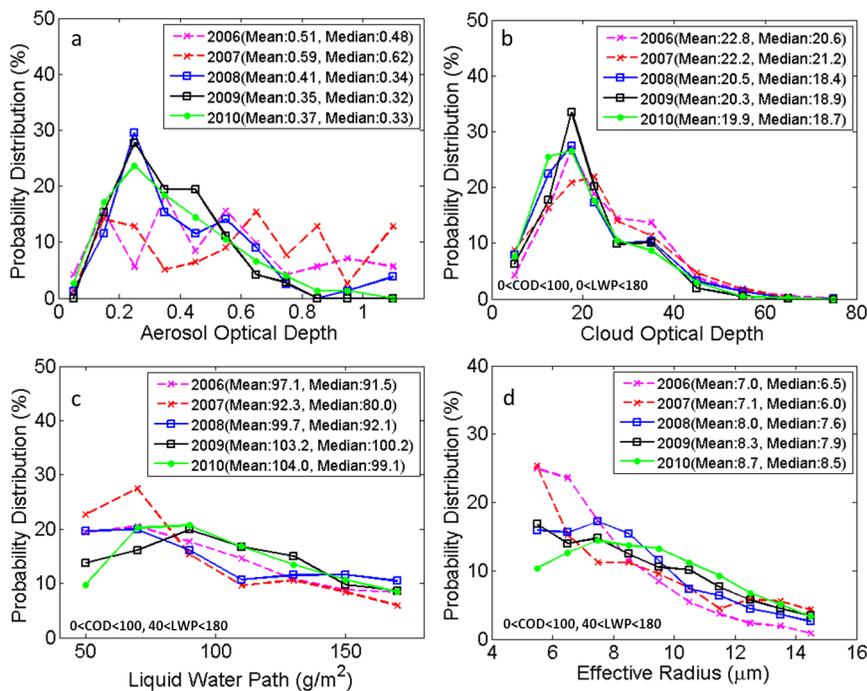
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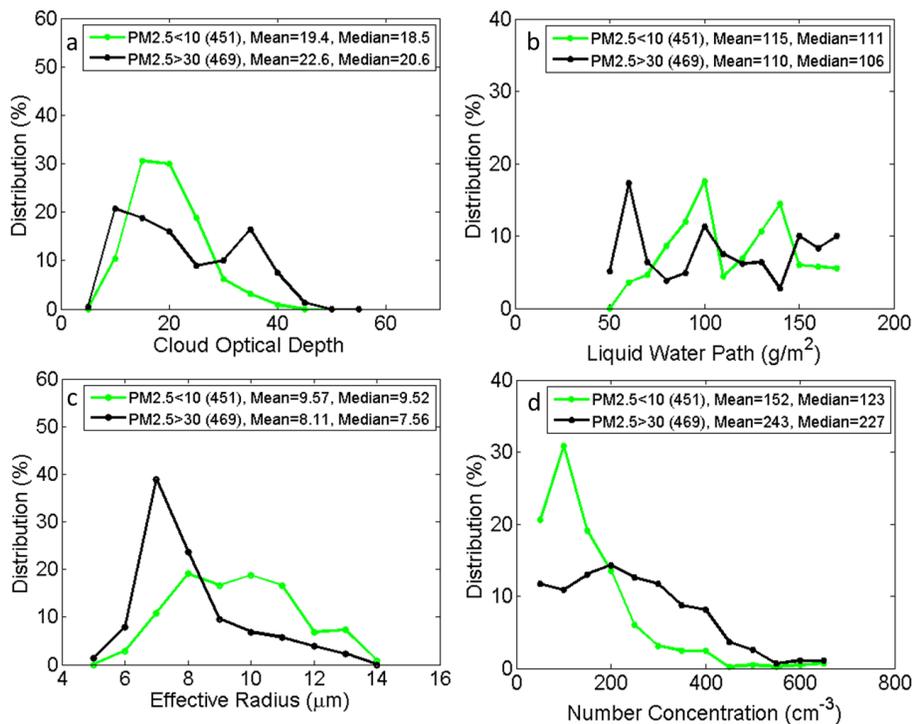


**Figure 1.** The distributions of aerosol and cloud properties. **(a)** Daily AOD measurements (bins: interval is 0.1 when AOD is from 0 to 1.0 and the last bin is  $\text{AOD} \geq 1.0$ ). **(b)** 3 min averaged COD (bins: the first bin  $\text{COD} < 10$ , interval is 5 when COD is from 10 to 70 and the last bin is  $\text{COD} > 70$ ). **(c)** 3 min averaged LWP measurements (bins: interval is  $20 \text{ g m}^{-2}$  when LWP is from  $40 \text{ g m}^{-2}$  to  $180 \text{ g m}^{-2}$ ) and **(d)** 3 min averaged  $R_e$  (bins: interval is  $1 \mu\text{m}$  when  $R_e$  is from 5 to  $15 \mu\text{m}$ ).

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**Figure 2.** The distributions of cloud properties under clean and polluted conditions. **(a)** Distributions of cloud optical depth. **(b)** Distributions of liquid water path. **(c)** Distributions of cloud effective radius and **(d)** cloud droplets number concentration.

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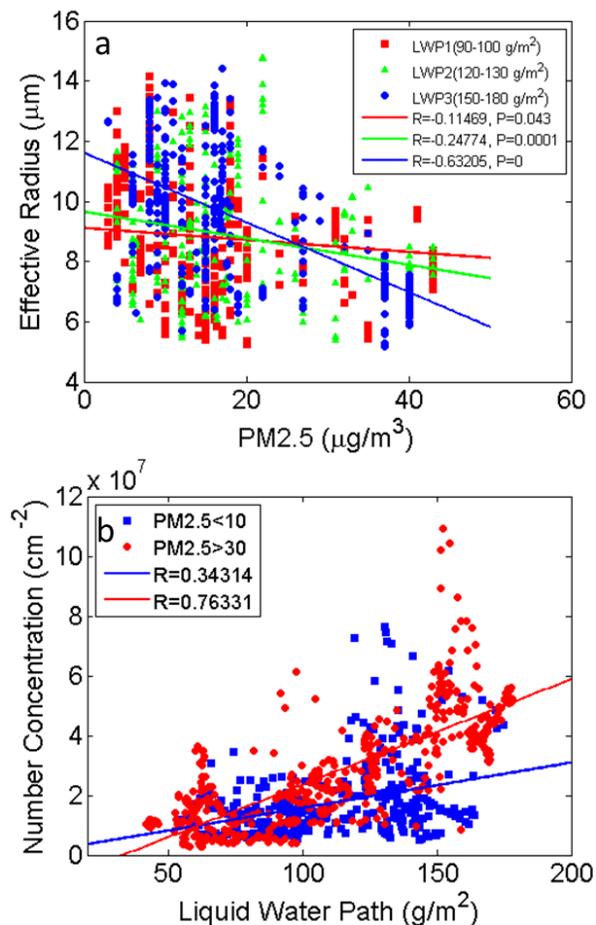
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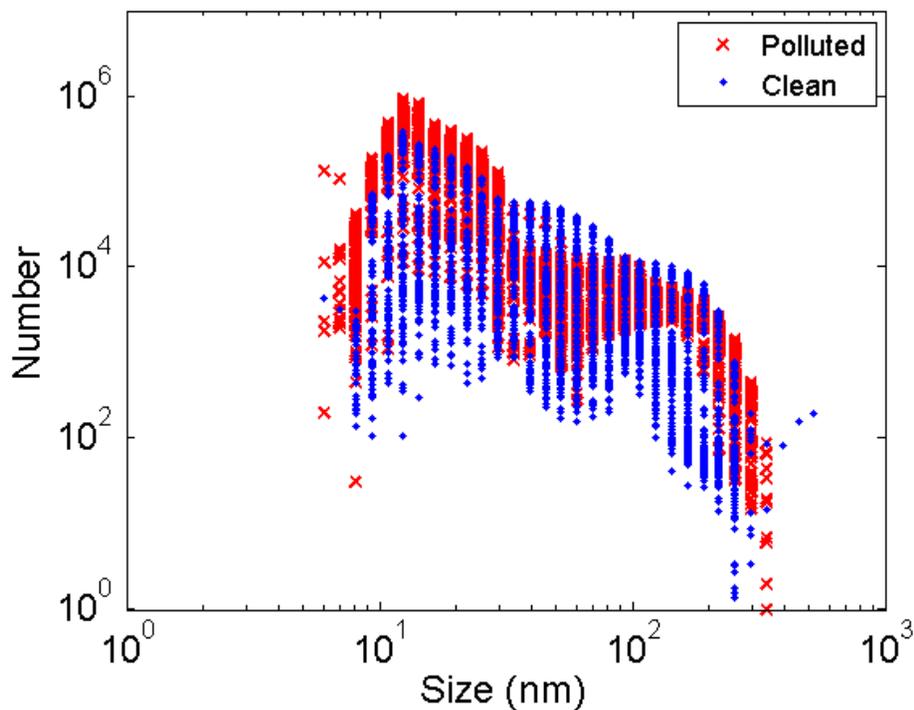
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**Figure 3.** (a) The relationship between  $R_e$  and  $\text{PM}_{2.5}$  under three different liquid water path ranges and (b) the relationship between cloud droplets number concentration and liquid water path under different  $\text{PM}_{2.5}$  value.

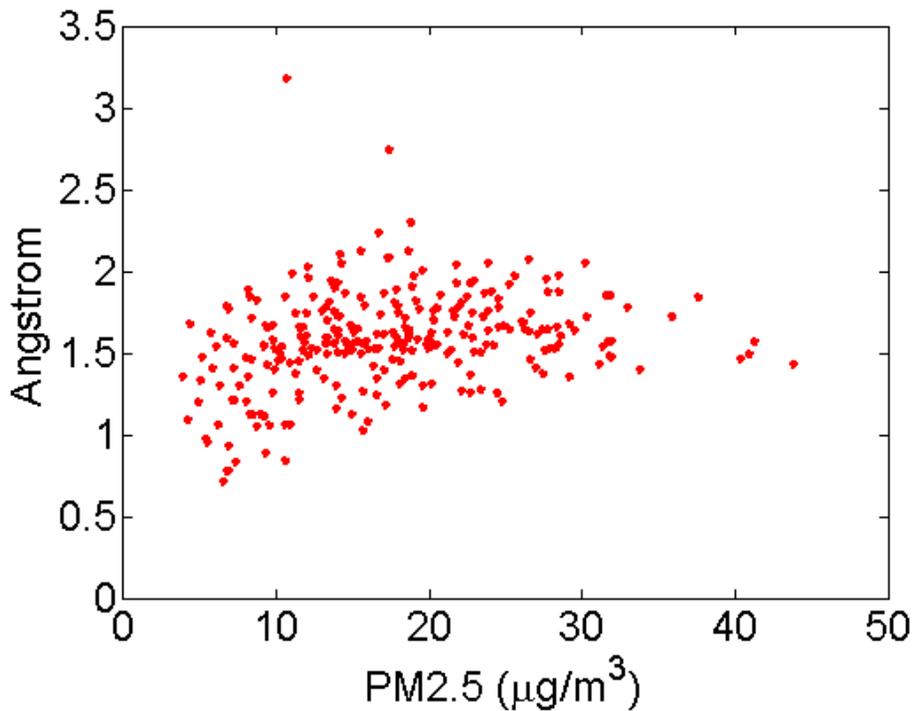
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**Figure 4.** FMPS measured aerosol size distribution on the polluted day (20 July 2011) and clean day (14 July 2011) during DISCOVER AQ field campaign.

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**Figure 5.** The relationship of Angstrom coefficient and  $PM_{2.5}$ .

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