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Interactive comment on "Long-term observation of aerosol–cloud relationships in the Mid-Atlantic of the United States" *by* S. Li et al.

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We thank Referee #1 for very thorough and constructive comments, which have helped to improve the quality of the paper. Below are our responses to those comments. The response follows each comment.

Anonymous Referee #1 Received and published: 15 August 2014 Review Li et al. 2014, ACPD:

This manuscript report general statistics and correlations between cloud properties, retrieved from a microwave radiometer and a MultiFilter Rotating Shadowband Radiometer (MFRSR), and PM2.5 aerosols at a site in Maryland USA. The manuscript suggests some aerosol modulation in cloud microphysics. The qualitative results are similar to many other studies that used observations over land, but unfortunately the qualitative analysis is insufficient and the conclusions are speculative, as they are not supported by the results. The authors need to include a more comprehensive analysis of the atmospheric modulation and a more adequate data screening that takes into account clear-sky contamination and cloud horizontal heterogeneities.

Main comments: - The analysis is overly superficial and limited to only a few histograms and scatterplots that do not demonstrate the occurrence of an indirect-effect: what type of clouds were sampled? Boundary layer clouds?, middle cloud? Convective clouds? The authors used liquid water to screen convective clouds, but nothing is said about the cloud base height, cloud top, boundary layer decoupling, atmospheric humidity, or stability. All these factors well could explain the relationships the authors interpret as aerosol indirect effects.

Response: Because of the limitation of instrument, the measurements of cloud vertical profile and cloud top information are not available at HUBC site. However in order to focus on clouds that are meteorologically similar, well developed, we used cloud lasting time (30 minutes continuous cloud), LWP (40-180 g/mˆ2), COD (less than 100) and Re (5-15 um) to constrain the cloud samples and we only chose cloud samples in summer time to remove seasonality. Based on the referee's comments, we tested our results by adding ceilometer derived cloud base height information. For most of the cases we chose, the cloud base height are below 2km. The analysis result based on cases with cloud base height lower than 2km are similar with current result (fig 1).

- I am not convinced that the use of PM2.5 is a good proxy for cloud condensation nuclei. The authors justify its use by invoking a positive correlation between PM2.5 and aerosol optical thickness AOT (r=0.67), which explain a variance of 45%. I suspect this correlation is the consequence of the dominant aerosol annual cycle, but if the data is deseasonalized, the correlation will be lower. Even if the PM2.5 and AOT are well correlated, the use of PM2.5 is debatable because 1) surface observations are not representative of cloud base observations (unless the boundary layer is well mixed)

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and 2) only large particles (accumulation mode) are likely to become CCN. PM2.5 contains particles with size smaller than 2.5um, which includes Aitken and accumulation mode. Figure 4 clearly shows that small particles dominate the aerosol distribution, demonstrating that PM2.5 is an inadequate proxy for CCN.

Response: In our investigation, PM2.5 is not treated as the proxy of CCN, but it is the proxy of aerosol. Whether the aerosol particles can be turned into CCN also depends on other conditions for example dynamical conditions which are associated with moisture supply. That is an important statement in this paper which was discussed in the last two paragraphs in result chapter. The relationship of PM2.5 and AOT at HUBC were tested in different seasons based on five years data and the correlation coefficient varied between 0.6 and 0.75 and in the summer it is 0.64. Although PM2.5 is only the index of aerosol with aerodynamic diameter <2.5m, it may indicate the change of whole population of aerosol to some extent, especially if the shape of size distribution is assumed fixed. One of the reasons we chose the samples in the summer time is that we assume stronger convection help to mix boundary layer air well. Also, based on referee's suggestion, we tested our results by screening out the cloud samples with cloud base higher than 2kms and we got the similar results(fig 1).

- The dataset section is incomplete: is effective radius a function of liquid water path?. How did the authors calculate Nd? What are the underlying assumptions? How did they remove the effect of clear-sky contamination? Or spatial heterogeneities? According to Nzeffe et al. 2008, a ceilometer was available in the station. Why a ceilometer is not used for screening the effect of broken clouds or clear-sky contamination? (a clearsky bias can offer an alternative explanation for the analysis in Li et al.). Ceilometer data can be useful to qualitatively determine the occurrence of precipitation (another big uncertainty in this study).

Response: The effective radius is retrieved based on liquid water path and cloud optical depth. Also, Nd can be retrieved based on LWP and COD. The retrieval of Nd based on the assumption that the clouds in question are adiabatic; Nd is constant, and cloud liquid water content varies with altitude adiabatically. The retrievals of effective radius and Nd were described detailed in a bunch of citations listed in the second chapter. The MFRSR can measure direct normal, diffuse horizontal and total horizontal solar irradiances. The measured direct solar irradiance is used to screen clear-sky contamination and in our paper we only investigate those cases for which clouds continuously last longer than thirty minutes. The advantages of ceilometer on clear sky detection are more at the night time. In the day time, the MFRSR has the similar ability on that. For screening the precipitation contamination, we use the data of lwp smaller than 180g/mˆ2 and cloud droplet effective radius smaller than 15um.

- Histograms in Figure 1 and 2 do not prove the occurrence of an aerosol indirect effect. The superficial comparison between several years is irrelevant, as the authors did not provide any additional atmospheric information that can offer an alternative explanation for the differences (in fact, from the histograms I do not see any meaningful difference). Figure 2 does not demonstrate any aerosol indirect effect either. It certainly motivates further analysis, but it does not prove the authors' hypothesis.

Response: Thanks for comments. In figure 1, we showed that the distributions of cloud properties are similar when the AOD distributions are similar and the distributions of cloud properties change when the AOD distributions change. That is possible associated to aerosol indirect effect or other factors. Both the large-scale processes and aerosol loading could impact cloud optical depth, Re and LWP. The assumption adopted is that LWP is sensitive to dynamical processes. Figure 1 showed that the change of LWP is not as significant as COD and Re, even in the conversed way to the change of COD. That is why we suggest that is due to the change of aerosol loading. Moreover, considering the referee's comments, we analyzed the distribution of surface temperature and relative humidity and didn't find obvious trend corresponding to COD and Re. Different from the data in figure 1 in which data is separated in different years, in figure 2 data are separated based on different PM2.5. Figure 2 showed that the distribution of cloud properties varied greatly corresponding to different PM2.5.

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- Figure 3 is highly scattered. In fact, the correlation and the slopes are controlled by a few outliers with PM2.5>30.

Response: Figure 3 is highly scattered when PM2.5 is small. The effective radius varies in a relative large range when PM2.5 is small. However when PM2.5 is large (PM2.5>20), the value of Re is constrained.

- One-day observations are irrelevant in the context of the statistical analysis of this paper. Moreover, contrary to the authors' interpretation, Fig. 5 does not really prove any link between angstrom exponent and PM2.5 for concentrations larger than 10 ug/m3.

Response: The one-day data was collected during the DISCOVER-AQ campaign which was a NASA led field campaign during July 2011 to observe local and regional scale meteorology and aerosol and gas phase chemistry from surface based and airborne platforms across the Mid-Atlantic region. Although limited to one month, it represents a rare data set over the region with detailed information on air quality and meteorological processes. It is leveraged here in that manner to provide an additional piece of evidence that could help to explain the long-term observations. Because the aerosol size is small in this area in the summer, the aerosol indirect effect is not obvious when moisture supply is not strong which is shown in figure 3. When PM2.5>20, the link between angstrom coefficient and PM2.5 seems weak. One of the reasons is the relationship of PM2.5 and angstrom coefficient is not linear. In the other hand we mainly want to show that when PM2.5>20, for most samples angstrom coefficient is larger than 1.5 which means for most polluted cases, the particle size is small.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 18943, 2014.

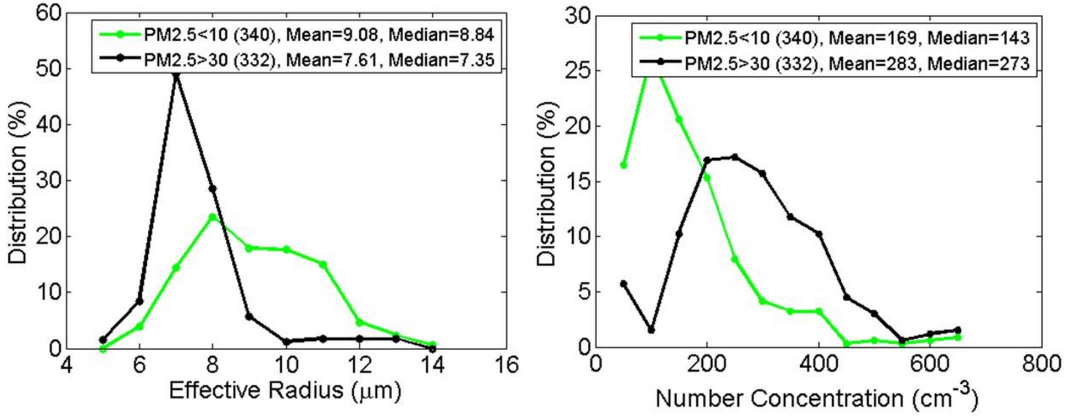

Fig. 1.

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