

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

Anonymous Referee #1

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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 PM_{2.5} 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.

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Main comments:

- The analysis is overly superficial and limited to only a few histograms and scatter-plots 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.

- I am not convinced that the use of PM_{2.5} is a good proxy for cloud condensation nuclei. The authors justify its use by invoking a positive correlation between PM_{2.5} 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 PM_{2.5} and AOT are well correlated, the use of PM_{2.5} is debatable because 1) surface observations are not representative of cloud base observations (unless the boundary layer is well mixed) and 2) only large particles (accumulation mode) are likely to become CCN. PM_{2.5} contains particles with size smaller than 2.5 μm , which includes Aitken and accumulation mode. Figure 4 clearly shows that small particles dominate the aerosol distribution, demonstrating that PM_{2.5} is an inadequate proxy for CCN.

- The dataset section is incomplete: is effective radius a function of liquid water path?. How did the authors calculate N_d ? 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 clear-sky 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).

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- 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.
- Figure 3 is highly scattered. In fact, the correlation and the slopes are controlled by a few outliers with $\text{PM}_{2.5} > 30$.
- 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 $\text{PM}_{2.5}$ for concentrations larger than $10 \mu\text{g}/\text{m}^3$.

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