Review of Li etal. 2014. Long-term observation of aerosol–cloud relationships in the Mid-Atlantic of the United States

General Assessment.

In this work the authors use ground-based measurements in an attempt to investigate the effects of aerosol emissions on cloud microphysical properties in the mid-Atlantic region of the United States. By analyzing the distributions of cloud optical depth, liquid water path and aerosol optical depth, the authors conclude that aerosol emissions have affected the formation of clouds in the area during the summer months. The topic of the work is clearly relevant to the scientific community since evidence of the aerosol indirect effect is still scant. However there are several methodological and conceptual flaws in the work that compromise the author's conclusions. Therefore I find that the work is not suitable for publication in ACP and recommend its rejection.

Briefly, the authors omit important considerations regarding the effect of different meteorological regimes that may be in place and bias their conclusions. Most of the analysis methodology used has been developed specifically for stratocumulus clouds. The authors do not discriminate between convective and stratus regimes. It is not clear how the cloud vertical extent is playing a role in the authors' analysis and what the meaning of the derived effective size and droplet number is (i.e., does it represent cloud top, cloud base, both?). Furthermore, since only COD and LWP are being retrieved either Re or Nd, but not both, can be inferred. Also I find that extending directly conclusions based on the modeling of single clouds (for example high LWP leads to high COD) to distributions of cloud properties is conceptually wrong and must be avoided since many other factors (meteorology, aerosol-cloud feedbacks, onset of convection) may play a role. The authors should instead collocate aerosol measurement with cloud measurements instead of comparing annual distributions. The data set is however valuable and I encourage the authors to repeat their analysis and resubmit their work.

Specific Comments:

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Abstract: Aerosol indirect effect is used vaguely without really knowing what the authors mean by it. Please refer to specific cloud properties instead.

Line 6 and other places. It must be Cloud droplet effective radius and cloud droplet number concentration.

Lines 8-9. The authors must be more specific on what is meant by "a higher frequency of abundant aerosol loading".

Lines 12-13. It is not clear what "elevated" and "decreased" means here. What is the baseline for comparison? Were there any clean even that can be used as control?

Lines 15-18. It is not clear what the authors mean by "differences between the two cases of influential factors on cloud properties were relatively controlled."

Lines 22-24. This is a badly formulated sentence.

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Line 1. It was said before that an increasing number of studies show this, but that authors only cite one from 1977.

Lines 1-5. This is not the definition of aerosol indirect effect. What the authors describe is the radiative forcing from aerosol cloud interactions (RFaci). Please refer to the most current IPCC report.

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Lines 1-5. Only COD and LWP are retrieved independently. Nd and Re are derived using two different algorithms. Re is obtained from COD and LWP. Then Nd is obtained from COD and Re. Since obtaining COD from Re implies an assumption on the droplet size distribution, the same measurement cannot be used to obtain a unambiguous estimate of Nd. In other words, using only COD and LWP and some assumptions the authors can obtain either Re or Nd but they cannot consistently obtain both. There seems to be a methodological flaw here.

Lines 1-5. Along the same lines it seems that the authors forget the fact that clouds also have a vertical extent and that while COD and LWP and integrated quantities along the vertical Nd and Re are not. The physical meaning of the "retrieved" Re and Nd is not clear, should they have any meaning at all.

To give at least some consistency to their analysis the authors should focus on clouds that are meteorologically similar, well developed, and that have small vertical extent. It is not clear that after applying those filters enough data will be left to do a proper analysis.

The authors also omit the fact that the parameterization employed to obtain Nd was developed for extended stratocumulus, typical of the coast of California and Peru, but rarely seen in the Baltimore area.

Line 23. The authors screen for low LWP but it seems that no screening is made for high LWP. Does this mean that the analysis is applied to convective clouds as well? This would be a conceptual flaw as the simplified method used by the authors does not apply to convective clouds.

Line 25. Can the uncertainty in Re be quantified?

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Lines 9-11. Where COD is proportional to LWP (assuming that size is constant) for a given cloud the authors are incorrect at extending such conclusion to the distributions of COD and LWP. A higher frequency of LWP may correspond to a higher incidence of convective events which may lead to a higher scavenging rate of the aerosol therefore resulting in cleaner years, but for a different reason. Many other factors may affect the distributions and such comparisons cannot be taken as proving the aerosol indirect effect.

Line 8. There is in fact no statistical difference in LWP among all the years since the uncertainty is about 20 g m-2. What is the uncertainty in COD and AOD retrievals? Are the differences statistically significant?

Line 15-20. There is no evidence to support this claim. It is not clear whether the differences are statistically significant. Furthermore, correlation does not imply causation. The same behavior can be explained by a larger incidence of convective events (which are not screened out) in the cleaner years which increase aerosol scavenging rates. Finally, there is no guarantee that conclusions based on process-based calculations apply to distributions of cloud properties.

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Lines 3-4. As mentioned above Re and Nd are not independent retrievals (which is also evident since the distributions are mirror images of each other).

Lines 4-5. Figure2 actually exhibits two modal distributions for COD and LWP under polluted conditions. This is a strong indication that other mechanism, rather than aerosol effects, may be responsible for the differences. My candidate is a higher frequency of convective events.

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Line 20. How is the vertical distribution of effective radius taken into account in this analysis?

Line 25. A correlation between LWP and Nd is actually indicative that effects other than the Twomey effect are at play.

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Line 20. Maybe MODIS and Calipso data could be used to better characterize the aerosol over the period of time studied, using this campaign as validation.