Atmos. Chem. Phys. Discuss., 14, C8965–C8973, 2014
www.atmos-chem-phys-discuss.net/14/C8965/2014/

Atmospheric Chemistry and Physics Discussions

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

S. Li et al.

siwei.li@howard.edu

Received and published: 10 November 2014

We thank Referee #2 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.

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, 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.

Response: We only chose cloud samples which continued more than 30 minutes and with LWP is larger than 40 g/mˆ2 and smaller than 180 g/mˆ2. Re and Nd are dependent. Nd can be retrieved with known Re and COD under the assumption that clouds are adiabatic. The original data are collocated. The statistical analysis are based on the collocated original data. PM2.5 is used to instead of AOD in our analysis since Figure 2. The detailed answers are following.

Specific Comments:

Page 18944

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

Response: We change the sentence “Evidence of aerosol indirect effect is found where for polluted cases the mean and median values of COD and Nd distributions were elevated while the mean and median values of Re were decreased.” to “Evidence of aerosol indirect effect is found: in polluted cases the mean (median) values of Nd are elevated and the mean (median) values of Re are reduced, and consequently for given LWP, the cloud optical depths are increased compared to the clean cases.”

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

Response: Thanks, we corrected it in the reivsed version.

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

Response: Thanks, we change it to “a higher frequency of abundant aerosol loading (AOD>0.5)” in the revised version.

Line 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?

Response: Thanks, we changed the sentence as ““Evidence of aerosol indirect effect is found: in polluted cases the mean (median) values of Nd are elevated and the mean (median) values of Re are reduced, and consequently for given LWP, the cloud optical depths are increased compared to the clean cases.” ”

Line 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”.

Response: Thanks, we change it to “the dynamic differences between the two cases of influential factors on cloud properties were relatively controlled” in the revised version.

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

Response: Thanks, we rephrase it in the revised version as “Investigation of the radiative forcing from aerosol-cloud interactions (RFaci) which refers to aerosol effect on the concentration of cloud condensation (CCN) and ice nuclei and then on cloud albedo also known as the Twomey effect (Twomey, 1977) is crucial to estimates and interpretations of the Earth’s changing energy budget. (IPCC, 2013).”.

Page 18945.

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

Response: Thanks, we rephrase the sentences in the revised version as “A growing list of studies have employed space- and ground-based observations to provide convincing evidence of RFaci. By using satellite remote sensing, Han et al. (1998) showed that. . ..”.

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.

Response: Thanks, we change the AIE to the new definition RFaci in the revised version.

Page 18947

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 an 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.

Response: Yes, the COD and LWP are retrieved independently. In the retrieval of Nd, Nd is assumed constant in nonprecipitating adiabatic parcel. With this assumption, Nd can be presented by a function of LWP and COD. Re which is an equivalent vertically uniform parameter ("mean Re") can be also represented in terms of a function of LWP and COD. With retrieved Re and COD, Nd can be retrieved if the LWP is unknown. So we can say Nd depends on Re but not just depends on Re. Here Re is equivalent vertically uniform cloud droplet effective radius and Nd is cloud droplet number concentration.

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.

Response: Thanks for the comments. The Re and Nd have been explained above.

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.

Response: Thanks, the data are filtered by using cloud lasting time (30 minutes continuous cloud), LWP (40-180 g/mˆ2), COD (less than 100) and Re (5-15 um) based on the measurements we have at HUBC site to constrain the cloud samples and we only chose cloud samples in summer time to remove seasonality.

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.

Response: Thanks for the comments. The retrieval of Nd is developed with the assumption that the clouds are adiabatic. Min et al., 2012 introduced the adiabaticity in

the retrieval of Nd and showed the approved accuracy for Nd retrieval for sub-adiabatic clouds. Because of the limited measurements and retrieval method, Nd is retrieved with the assumption that the clouds are adiabatic or have similar adiabaticity in this investigation.

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.

Response: Thanks, we did screen high LWP (larger than 180 g/mˆ2) cases. (page 18948 line 27)

Line 25. Can the uncertainty in Re be quantified?

Response: Compared with eight aircraft in situ vertical profiles (constructed from measurements), Forward Spectra Scattering Probe (FSSP), the retrieved Re for single-layer warm water clouds agree well with in situ measurements, within 5.5% (Min et al., 2003). And the main uncertainty of retrieved Re come from the uncertainty of LWP which is mainly a systematic bias and moreover represents the preponderance of the total error. What we are interested in is the relative difference between clean and polluted conditions, so the consequence of systematic biases is therefore minimized.

Page 18948

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.

Response: The statistical analysis are based on the collocated original data. In the data, deep convective events have been screened out.

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

Response: The error of LWP retrieval consists of instrument error, errors associated with the climatological profiles used for the retrieval (given day to day variability of atmospheric temperatures from this climatology), and errors from the absorption model used to develop parameters for the retrieval algorithm (Turner et al., 2007). The former two errors are considered random errors and thus are minimized with increasing sample size of data. The component of the error that derives from the absorption model is considered a systematic bias and moreover represents the preponderance of the total error (Turner et al., 2007). It is the relative difference of cloud-droplet effective radius (re) vs. LWP for clear and polluted conditions that is crucial to the approach put forth in this study for investigating AIE; the consequence of systematic biases is therefore minimized. The uncertainty in COD is better than 5% and the error of retrieved AOD is below 0.004. The differences are 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.

Response: Compared to the measurements uncertainties, the differences are statistically significant. In our investigation, deep convective events (LWP >180 g/mˆ2) have been screened out. In the figure1, we firstly showed a potential RFaci and the detailed analysis are followed in the text. The distributions of cloud properties are based on the

collocated data which is used for process-based calculations.

Page 18948

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).

Response: Thanks, the meaning of Re and Nd have been explained above.

Lines 4-5. Figure 2 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.

Response: Both dynamical and microphysical factors may affect COD and LWP. It is possible that other mechanism exist. However in this investigation, we try to isolate aerosol's effect from the other impact by choosing cases with similar dynamical conditions but different aerosol loading. Deep convective events (LWP >180 g/mˆ2) have been screened out of the dataset.

Page 18949

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

Response: Thanks for the comments. Because of limitation of instrument, retrieval of vertical distribution of effective radius is not available at HUBC site. In this work, the Re we used is an equivalent vertically uniform parameter ("mean Re") retrieved from the measurements of MFRSR and MWR.

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

Response: We agree with referee that other effects are also at play. In this paper we mainly investigate aerosol's role besides other effects. We show that there is a cor-

relation between LWP and Nd and also the relationship (slope) changes with different aerosol loading. In other words, increase of aerosol can affect the other factors' impact on cloud properties.

Page 18950

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.

Response: Yes, agree with the referee, satellite data are very unique and useful. However MODIS only passes our site twice per-day and the space scale is much larger than ground based observation. Considering space and time resolution, we used ground based observations at our site in this work. Thanks for referee's valuable suggestions, we are considering to combine satellite data with our site data to extend our investigation to larger area and different climatological area in the future.

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