Review of Christiansen et al.

This manuscript takes a look at some of the potential biases that arise when satellites are used to validate/inform the modeling/interpretation of ground aerosol measurements (specifically for the continental US and the MODIS satellite sensors). Valid aerosol satellite retrievals for scenes impacted by clouds are challenging, even in the free troposphere (e.g. above the cloud deck) and when looking specifically at ground data, they basically result in a large fraction of measurements being discarded. Christiansen et al ask the question how this missing data impacts aerosol composition and one related quantity, aerosol liquid water (ALW), using IMPROVE data for the former and ISORROPIA runs to estimate the later. They conclude that there is indeed a significant bias in both quantities across most seasons in the US.

Since clear-sky AOD (which is impacted by both dry aerosol concentration and ALW) is one of the preferred ways to evaluate GCM performance over large domains, the bias described in this paper is very relevant to the ACP readership. The chosen approach is simple, elegant and reasonably robust. While it has been used before to explore the PM2.5 bias in the CONUS (Christopher and Gupta, 2010), this manuscript improves on the method, uses more recent data and more advanced MODIS products and adds seasonality and ALW to the mix. So it will certainly be a useful addition to the literature. Not unlike Reviewer #1, however, I found some aspects of the logical flow of the current manuscript confusing, so here are some suggestions that hopefully will improve the presentation of what is overall very nice work.

Major comments:

- There are two interrelated questions that the manuscript is currently trying to addresses:
    a) Does the presence of boundary layer clouds impact aerosol composition on the ground in rural US locations and if so, how?
    b) Does the aerosol in cloudy satellite scenes look different than clear sky scenes and soes this result in a systematic bias for AOD analysis that only use clear sky scenes?

  While most of the manuscript focuses on a), b) is repeatedly mentioned in the abstract and conclusions, although never really directly addressed. Now, if (a) is indeed the main objective, the current approach is a little bit less than ideal. To wit:
    - If I follow correctly, the cloudmask used in this manuscript does NOT differentiate between low and high clouds (although this information is provided by MODIS). If the goal of this paper is (b), that is a valid choice. But if the goal is to actually explore how boundary layer clouds affect chemistry, this should be certainly changed, and the analysis repeated with low clouds exclusively. I would expect that this will actually strengthen the trends observed and won't be statistically too costly, especially in the Western US.
    - While relating the cloudmask to the most often used AOD sensor is certainly very useful to explore (b), it results in a pretty clear bias since AQUA and TERRA overpasses over the CONUS are only once a day each, and none of them after 2 pm (this info is missing currently from the manuscript and needs to be added for context), while the compositional data is taken for a 24 h period. Especially for the late Spring and Summer, where both the maximum cloud and photochemistry activity is later in the day, this will

lead to some misclassification. So again, if the impact of BL cloud chemistry is the focus, using data from a geostationary satellite (e.g GOES16 or Himawari-8, both of which have similar pixel sizes to MODIS) for full day coverage would make more a lot more sense and result in likely more robust results that using MODIS.

On the other hand, if (b) is the main objective, as Reviewer #1 mentioned in his review, the current manuscript does not really try to relate the changes in PM2.5 and ALW to an actual bias in the MODIS product.

From my reading of the manuscript, the goal seems to be to answer (a) and use (b) for illustrative purposes, which is a good choice in my opinion, especially since

- the authors have already published a manuscript with a similar approach where they look in more detail at the AOD impacts (Christiansen et al, 2019) that could be mentioned in some more length in the conclusions, and
- as discussed both in the manuscript and below, there is just too much uncertainty in the IMPROVE data to attempt closure, especially since FT aerosols are not included and likely play a role (again as discussed in Christiansen et al, 2019)

But in that case, I would certainly ask the authors to please clarify and illustrate the trade-offs in their choices of satellite products. Switching to geostationary data is probably outside the scope of this project, but the option should be discussed (and maybe considered for future analysis of similar dataset).

- Related to the previous point, the current manuscript spends considerable space talking about cloud affecting the FT aerosol, which is not really the focus of the paper (e.g. not clear to me why the fact that most aerosol measurements from aircraft exclude the cloud itself is relevant to this work, which deals with the aerosol below), so I would consider shortening those sections (mostly the intro) significantly to avoid confusion and for better flow. It also never really mentions the most important factor that matters for what the authors are exploring here, aerosol ground concentrations, namely clouds blocking any retrieval below cloud height. It is true that the MODIS team has done a great job trying to retrieve AOD ABOVE cloudy scenes, as the manuscript mentions, but below is just not feasible if it's too cloudy, one of the reasons why the author's analysis is so valuable: it's not faulty data, but missing data that they are addressing.
- I would strongly suggest to move the PM2.5 analysis at the beginning of the discussion, since it is the simpler metric to start with, and then go into the detailed compositional/ALW analysis. Most importantly, since this type of analysis has been done before, both in Christopher and Gupta with older MODIS data product and , for non-cloudy data using the new algorithm in Chudonowski et al, 2013, it would actually allow for a direct comparison of the observed PM2.5 with previous studies, which would significantly strengthen the analysis. It would also, to the extent possible, be useful if the authors could comment on why Christopher and Gupta came to the opposite conclusion.
- The speciated IMPOVE data is certainly not ideal for calculating ALW. The authors have done one sensitivity analysis to investigate the impact of dust. However, a similar analysis for the missing ammonium and OA is missing.
    - The impact of ammonium, since it will be associated with nitrate and sulfate, will, in the absence of good acidity measurements, just be a positive offset on the current trends,

as the authors write. Still, since for neutralized aerosol it is going to add significant mass and ALW to the analysis, it would be nice to add this explicitly to Fig S2

    - I agree with Reviewer #1 that not estimating OA ALW is an odd choice, since OA has a clear seasonal trend that will in some cases make the observed trends likely stronger and there are good published estimates for kappa for oxidized aerosol over the continental US (e.g. Brock et al, 2016 and Shingler et al, 2016). Obviously fresh POA is an issue, but since the analysis is restricted to rural sites there should be very little urban POA. Fire POA if needed can be just filtered by either the MODIS fire flag or some simpler compositional metric (e.g. f(OA)>0.8). So I would suggest that – at least as an additional sensitivity study - the authors add OA ALW, using an OA/OC of 2.1 (used for OOA in newer GCMs like Geos-Chem and CESM2) outside of fire days and a kappa value of ~0.1 (for OA). This would be very close to the assumptions that a modeling study would do and hence give a good sense of the possible effect

- I agree with Reviewer #1 that changing the format of Fig 3 to 7 (and Fig S4) to a single map with barplots for the four seasons (or some variation of this general idea) would make it much easier to take in the information conveyed in those graphs quickly, and would highlight the seasonal trends much better than the current format.
- The effect of wet deposition is likely significant and I would not expect it to be countered by evaporation (at least outside of cloud detrainment). Its sign is opposite to the trends shown in this paper and hence it does not affect the significance of the conclusions. Still, since the authors have the reanalysis meteorology, a simple sensitivity study looking at the differences between cloudy vs cloudy + rainy days would give the reader a better sense of how large the problem is.

Minor comments:

- Line 24: For OA and sulfate, Ervens et al 2018 have recently looked at FT cloud chemistry as well, please add.
- Line 24-25: Clouds do not redistribute aerosols and gases, convection (or convective clouds, if you will) does. Please rephrase.
- Line 40: This is wrong as written. RH dependent lab studies have been done since the ‘60s! I am assuming the authors are referring specifically to environmental chamber work exploring aerosol chemistry, where it is true that many older studies were done under dry conditions, but that has changen in the past decade, see e.g. Petters et al, 2017 or Schwantes et al 2019 for recent examples. So please revise accordingly.
- Line 97: It is not clear to me how 17x500m = 50 km, please explain. It would also help to mention some version of this is standard practice for most satellite/ground comparisons and that e.g. Christopher and Gupta, 2010 used 5x5 grids with a resolution of 4 km.
- In that context, it would be worth citing Twohy et al, 2009 that the effect of clouds on AOD extends at least 20 km farther than the actual cloud, and that using the larger detection area will hence certainly improve the accurate detection of *cloud free scenes*
- Line 215: Biogenic emissions have a strong dependence on ambient/soil temperature. Cloudy scenes are likely colder, so in the absence of data this effect could be either sun or temperature
- Line 217: While there might be some uncertainty in OA ALW (mostly related to the O/C ratio of OA) for the inorganic concentrations at this rural locations the overall effect is not that large. So while this statement might be true in other domains, I would revise it for the CONUS.

- Line 240: Jefferson et al show f(RH) that are significantly higher than the values reported in Table 1. Again, it would increase confidence in the analysis if it could be shown that adding ammonium and OC does result in roughly similar growth rates.

References:

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