Comment on acp-2021-999

Anonymous Referee #2

Referee comment on "Addressing the difficulties in quantifying droplet number response to aerosol from satellite observations" by Hailing Jia et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-999-RC2, 2022

Review of "*Addressing the difficulties in quantifying the Twomey effect for marine warm clouds from multi-sensor satellite observations and reanalysis*"

The article studies the effect of aerosols on liquid cloud droplet concentration using several space-based instruments (active and passive) to describe cloud and aerosol properties and reanalysis to retrieve information on sulfate. The authors investigate the potential biases that are usually overlooked by most of the studies quantifying this effect. The considered biases are the updraft, precipitation, retrievals of AOD and droplet concentration by satellite observations, and vertical co-location of aerosols and clouds. The sensitivity of the cloud droplet number is retrieved and quantified considering different regimes constrained for the potential biases individually.

The paper addresses relevant scientific questions within the scope of ACP and presents ideas using pre-existing data and methods but used for new scientific questions which I find particularly interesting. It presents different conclusions on the potential biases when studying the aerosol impacts on liquid cloud properties. The results are interesting for the scientific community and the paper indicates the credit to related work and the motivation of their new contribution. The abstract reflects the contents of the paper.

However, I have some concerns that need to be addressed. I was hoping for an S value with and without constrains for all the biases to evaluate the impacts all together, it would help to motivate future studies. I think the method section needs more details: There are many datasets used and many constrains, but it is difficult to understand what is used for each section. Considering the lack of my understanding in the method, I cannot assess that the results are sufficient to support the conclusions and it would be difficult for fellow scientists to reproduce the study. Also, the study refers to the Twomey effect, but the authors are not looking at the changes in radiative properties. I find the term of Twomey effect in the title and through the text misleading. Most of the discussions are in the section "*results*" and not "*discussion*" and the "*discussion*" section is more an outlook, but otherwise the paper has a good structure. For all the different reasons mentioned above, I

suggest major revisions before accepting the article for publication. I described below the different comments that I think are needed to improve the paper.

Major revisions

- As suggested in my introduction, the study claims to deal with the Twomey effect, but the Twomey effect refers to the change in cloud radiative properties due to CCN. The change in cloud droplet number is one of the causes. The cloud droplet concentration can be linked to the Twomey effect only if the water content is maintained constant. I do not think the present study is constraining for water content therefore the change in cloud droplets cannot be related to the Twomey effect here. I understand that biases on S would impact the quantification on the Twomey effect but claiming that "*the measure of the Twomey effect (S)*" on line 378 is wrong. I suggest limiting the reference to the Twomey effect as a motivation and removing it from the title because it is not what the paper is quantifying.
- I have several problems with the method in section 2:
  - There are a lot of datasets, but I am still confused about which dataset have been used for which part of the study. Maybe it is ok in the section, but I suggest to explicit the datasets used for each part of section 3 (as it is done for section 3.2 on lines 215-218).
  - Line 112: I do not understand how aerosol and cloud properties are collocated, since the aerosols are not retrieved if the pixel is cloudy. Can the authors explicit how they deal with that? The closest pixel is mentioned in one of the sections, but I do not know if this is the method for the entire study.
  - When MISR and Terra are used, I am guessing that A-train observations are not used, but I am not sure. Can the authors clarify?
  - Line 117: "*the lowest 15%*", is this threshold based on Ma et al. (2018) or is it an ad-hoc choice? If the latter, the authors should specify why 15% is chosen. Also, the lowest 15% can be important as it represents a transient mode and potentially the highest impact of aerosols on cloud properties. Did the author study how it potentially impacts the results?
  - Line 151: about dividing the dataset into 20 bins of AI/AOD, considering the median, and doing the fit, I understand the idea but I think the dataset loses a lot of information doing that. Also, if the dataset is large enough, the outliers will be removed since most of the points are going to be around the correct value. A statistical test is better than considering the medians. The difference between the blue and white lines in the example (Figure 1) are very similar and does not convince me that the method chosen by the authors is better than considering every data point.
- The authors are mentioning the uncertainties from space-based observations and how to reduce them (e.g., looking at certain solar zenithal angles...), but I doubt that the uncertainties are reduced to 0. However, remaining uncertainties are neither included in the results nor discussed by the authors. I think a discussion is missing about the uncertainties in the retrievals but also on the methods: The authors could have retrieved uncertainties on S with the 95% confidence interval on the fit considering the entire dataset (and not only the medians over the 20 bins) as it is done by many previous studies on the aerosol impacts on liquid clouds. Considering uncertainties on the fit would have been relevant when comparing the sensitivity values, for example in lines 222, the difference between 0.45 and 0.56 in S might not be statistically significant, I am not convinced by the comparison made if uncertainties are not provided.

- I do not understand for which type of clouds the study is designed.
  - The CBH is used as a proxy for the updraft and seems to be designed for cumuliform clouds as stated in lines 55 and 169 but in the method section, I do not see any constrain for avoiding other type of clouds. Therefore, I am wondering if the proxy for the updraft is relevant in most of the observed pixels.
  - There are some specifications referring to adiabatic situation (line 120 "*adiabatic approximation*"), to cumuliform clouds (line 55), or to convective clouds (line 169), but there is no constrain for parameters to limit the study to these situations. Therefore, I am not sure that the proxies used by the study are relevant. There is a threshold on considering single layered clouds (line 123), but I am not sure it is enough.
  - Some results are associated to "*stratus clouds*" (line 177). I guess that there is no threshold on the type of considered clouds, but then I do not understand how to use linear correlation only applicable to convective clouds and the discussion is about effects from stratus clouds. The authors should clarify that.
  - Another example on line 278, where the authors compare DELTANd and DELTANall, these values might be retrieved for different clouds, for open/closed cells, convective clouds, I do not understand how these two quantities can be compared without further consideration on the type of clouds. The differences in S can be explained by the biases, as suggested by the authors, but also by different cloud types, meteorology… The authors acknowledge it on line 410 "*CF also covaries with cloud dynamics*". Can the authors explain how they can certify that the observed differences are due to the biases and not due to different environments?
  - There is a cloud regime dataset from MODIS observations, did the authors try to use that to separate the different effects? (Naeyong Cho, Jackson Tan and Lazaros Oreopoulos, L. (2021), MODIS Cloud Regime Level-3 Daily 1 deg x 1 deg, Goddard Earth Sciences Data and Information Services Center (GES DISC), 5067/MEASURES/MODISCR/EQANGD/DATA301)
- I understand that the authors cannot study all the potential biases on the aerosol-cloud interactions, but I am wondering how did they chose? They considered the updraft velocity, but another important meteorological parameter is the humidity for which important effect on S has been demonstrated by previous studies.
- Some pixels can be mixed phase clouds but detected as purely liquid by the algorithm, impacting the effective radius, optical thickness, and Nd. I do not know the temperature range on the study, but does the dataset have liquid pixels potentially contaminated by ice?
- Result section, there are many discussions on the result section which should belong to the discussion section (e.g., from line 176 to line 182, from line 184 to line 191, from line 204 to line 213, from line 237 to line 244, from line 331 to line 339).
- The authors decided to study different biases on the aerosol-cloud interactions separately and I am wondering if the biases are not correlated with each other. For example, on section 3.2, the impact of precipitation on S is highlighted but it could also be due to a correlation between precipitation and the CBH (or CGT). Did the authors try to study the effect of precipitation on S constraining for CBH and/or CGT for example? Same apply for the other biases.
- Section 3.3 I am confused by this section, and I am not sure to understand the results and the discussion about it, can the authors rephrase this section?
  - I am skeptical about looking at aerosols next to clouds in general: The presence of a cloud means that the conditions are different than where there is clear sky. How can the authors make sure that the aerosols, meteorological parameters are the same between clear sky and cloudy sky?
  - Also, the authors mentioned that studies on 3D effect and aerosol swelling next to observed clouds are lacking but there are two references about this subject that I think are relevant to this subject and do not appear on the present article:
    - https://doi.org/10.5194/acp-17-13151-2017
    - https://doi.org/10.5194/acp-17-13165-2017

- Line 264 "*It is also noted that SAOD shows first an increase and then decreases from the second DELTA L bin*", I do not see what the authors are refereeing to, or is it from the third bin?
- Line 276 "*highly depends on the retrievals bias in clouds*", since there is no information on method biases, I believe this statement is too strong.
- Line 307, "*The strength of the Twomey effect derived on a basis of column-integrated aerosol quantity…*", I am confused because on line 294, the authors said that SO4C AND SO4S mimic the column integrated, and here only SO4C shows a large slope so how is it directly linked to this? and SO4S does not show the trend described on line 307.
- Along the article, different methods are employed to explain the different biases, I was expecting at the end a value of S considering all the possible biases, (precipitation, too close to the cloud, ...) for latitude bands for example and/or season, but no. Each paragraph is developed individually and at the end the discussion does not bring all of them together.
- Figure 1:
  - Why the blue and white lines do not go until AI=1. The authors mention that the lowest values of AI are removed, but they do not refer to large values of AI (AI>0.5).
  - Also, in the plot of PDF Vs AI, I do not understand why the PDF is almost equal to 0 for AI ~0.5 whereas on the upper plot the PDF of the data are clearly greater than the 0 (might even be higher than the maximum at AI ~0.8).
  - I think it would be better to indicate the value of the regression to see the difference between the blue and white lines and discuss about it, there is nothing about it in the data and method section.
  - Why the study uses the blue line instead of the white lines? The authors could infer the uncertainty through the 95% confidence interval using the white lines.

Minor revisions

- All along the text, there are several words which are unnecessary in my opinion (e.g., line 112 "*basically*", line 175 "*It is clear that*", line 175 "*evident*", line 183 "*remarkably*", line 219 "*as expected*", line 220 "*much*", line 226 "*evidently*", line 229 "*obviously*", line 232 "*clearly*", line 271 "*serious*", line 275 "*sharply*", line 293 "*practicall*y", line 319 "*much*", line 341 "*clearly*"), and sometime they imply that something is evident but it is not the case (in my opinion).
- Line 7 "*consistent with stronger aerosol-cloud interactions at larger updraft velocity*", this is not the case for every type of clouds (e.g., arctic stratus).
- Line 25 "*This study will*", I suggest moving this sentence to the last sentence of the paragraph.
- Line 30 "*As reviewed recently…*", I do not understand in which context the present study is related to Quaas et al. (2020), will they consider the same biases, new ones… I think, the authors could clarify this part in the introduction. It makes sense afterward but not reading the introduction for the first time.
- Line 55 "*their strong correlation illustrated by in-situ observations of cumuliform clouds*", this sentence is important as it is a key correlation used through the study, so I think the authors could elaborate a bit more on the limitations of it.

- Line 116 "*a standard deviation higher than the mean val*ue", does this threshold come from somewhere specific?
- Line 117 "*the lowest 15%*", is this threshold based on Ma et al. (2018) or is it an ad-hoc choice. If the latter, the authors could specify why 15% is chosen.
- Lin 123 (Feingold et al., 2021), it should not have parenthesis.
- Line 140, is there a reason why "*MYD06 5-km*" is not written "*MYD05 5x5 km2*" to be consistent with "*CloudSat data at a 1.4x2.5km2*".
- Line 146, the authors mention that they considered sulfate from MERRA-2, I am wondering if they considering other species.
- Line 162 "*Nd is essentially a function of both CCN and updraft*", a citation should be added here.
- Line 190 "*Sai is consistently higher than SAOD*", not always as for CGT~900m
- Line 195 "*They proposed…*", Who are they? Are they Reutter and al.? If so, they should not be put in parenthesis in the sentence before.
- Line 199 "*proxy of updraft (CBH/CGT)*", are the quartiles enough to discriminate the updraft regimes described by Reutter et al.? The quartiles defined regimes based on how likely an updraft regime occurs defining different regimes, but I am not sure that they separate in the regimes described by Reutter et al., I am not sure we are in category b.
- Lines 201-203, *"As illustrated in the …"* I am confused by this sentence; can the authors rephrase this sentence? What I understand:
  - At low AI, the updraft should have limited impact on Nd (case a from Reutter et al.), but looking at the plot 3c and i, the updraft has a strong impact.
  - On the opposite, at high AI, the updraft should most likely have a strong impact, but looking at the plot, the values are messier, and I do not observe a strong dependence on Nd with AI here. Can the dependence be quantified by the authors?
  - Maybe it is irrelevant, but I am wondering why the authors based their regimes on CGT and not the ratio CGT/AI?
- Line 228 "*appears to strengthen the aerosol-Nd relationship*", can the author develop a bit more on this?
- Section 3 "*AOD*", the authors mentioned earlier in the text that they would not consider AOD and prefer the use of AI, but they use AOD in this section. Why changing the considered parameter?
- Lines 264 to 268, I do not understand this part, can the authors rephrase it?
- Line 282 "*it is clearly illustrated that CF regulates the negative correlation between DELTAL and DELTANd*", can the authors provide more information on that part?
- Line 287 "*Given that CF also correlates closely with cloud dynamics*", the correlation presented by the authors are based on the medians which are not very significant in my opinion, 2D histogram and regression on the entire dataset would be better.
- Line 295 "*commonly used*", can the author support this with citations? Maybe some references on the use of satellite observation combined with models to study aerosol cloud interactions are missing here.
- Line 296 "*is considered to be more relevant to the amount of CCN*s", can the author provide a citation for this statement?
- Line 302 "*pre-binned method*", is it the method described in Figure 1? If this is the case, can it be explicit? I am still skeptical, and I would prefer statistics on the entire dataset and not on the medians (as done in Table S1).
- Line 302 "*binend*"-> "binned".
- Line 320 "*such as western North Pacific and the Atlanti*c", this is true for SO4B but not for SO4C (also East coast of south America and South Africa), or maybe I am misunderstanding.
- Line 321 "*the spatial CV of SO4C exhibits a much smaller (0.88) value than those of SO4B and SO4S (1.84 and 1.79)*", are these values averaged over North Pacific and Atlantic, if this is the case the authors should specify the limit in lat/lon of the considered box.
- Line 330 "*loose correlation (R<0.3) …*", can the authors quantify or rephrase that

because I am not convinced especially for r(SO4B, SO4S) which seems high when the ratio is small.

- From line 331 to line 339, I find the conclusions on this discussion very strong. I think it should be at least quantified to support that.
- Line 406 "*In terms of aerosol…*", is this sentence part of point 3 or point 4?
- Line 419 "*SO4B-S-C*", can the authors specify again on the different quantity in the conclusions?