

The response to Reviewer #1:

The article "Observational Constraints Suggest a Smaller Effective Radiative Forcing from Aerosol-Cloud Interactions" by Park et al. quantifies the effective radiative forcing from aerosol-cloud interactions to better represent climate predictions. The authors used satellite observations, reanalysis and climate models to support their analysis and results, and they focus on the effect of aerosols on cloud droplet concentration and evaluate the role of aerosol activation, which is usually overlooked in current estimates of effective radiative forcing.

I think this topic is within the scope of the ACP, as it seems to be an important parameter that the scientific community should take into account. The hypothesis is well explained and we understand each step of the process, but there are parts that should be improved before publication on ACP. I think the main problem is the predominance of the appendix and important information should be included in the main article. I am also concerned about the omission of meteorological parameters in the study and I do not know how this affects the study of cloud controlling factors. I recommend the paper for publication after the major revisions I suggest below.

We sincerely thank the reviewer for their constructive comments and valuable suggestions. As our manuscript has been submitted to *ACP Letters*, which has strict word limits, we have prioritized presenting the main findings concisely. Where necessary, additional details have been included in the Appendix to provide further support for our findings. We kindly ask for your understanding in this regard. Below, we provide specific responses to each comment, highlighted in blue. All "L" references in our responses correspond to line numbers in the revised manuscript with highlighted changes.

Major Comments

1. The conclusion does not follow the ACP recommendations. The conclusion needs to be expanded.

"Every article must have a final section where the overall advances are concisely summarized and put in context. Although the results section may include some discussion, a synthesis and interpretation must appear in the final section. ACP expects that the concluding section will normally include the following components, although not necessarily in separate paragraphs:

** Summary: Summarize the main results and relate them to the objectives, questions, or hypotheses of the study. The summary should include the main quantitative results.*

** Synthesis/interpretation: Explain and interpret the results concisely to enable readers to make sense of them as a whole.*

** Comparison and context: Compare the results with previous studies to put them in context. Explain consistencies, inconsistencies, and advances in knowledge.*

** Caveats and limitations: State how these affect confidence in the overall results, and where*

future work is needed.

** Implications: Discuss what the results mean for our understanding of the state and/or behaviour of the atmosphere and climate, which is the main requirement for publication in ACP. The editor's acceptance/rejection decision will be strongly guided by this component of the concluding section.**

Thank you for your valuable feedback. In response, we expand the conclusion section to align more closely with the journal's recommendations. This revision now provides a comprehensive discussion of the uncertainties our study has, ensuring a balanced view of the findings. However, given the strict word limit of *ACP Letters*, it is challenging to provide more details. We appreciate your understanding regarding these constraints and thank you for your thoughtful review.

2. Aerosols have an effect on cloud properties, cloud droplet size and droplet concentration, but the effect is small compared to the effect of meteorological parameters. If the authors did not constrained the results of Fig. 1 by meteorological parameters, then the changes in N_d can be due to meteorological parameters and what is observed is (indirectly) the correlation between aerosols and meteorological parameters. If I understand correctly, these coefficients are used afterward in the Equation A6. This problem is taken into account for the cloud controlling factors but I am not sure about the impact for the study.

Thank you for your comment and suggestion. In the revised manuscript, we now account for environmental factors when assessing the activation rate of aerosols into cloud droplets. This ensures that meteorological influences do not confound the activation rate analysis, as reflected in L88-89, where we state: "To verify the key assumption while accounting for environmental influences, we performed cloud controlling factor (CCF) analysis (Appendix A3)". This approach results in consistent findings, supporting the robustness of our conclusions. Additionally, in the revised manuscript, we extend this consideration to include not only activation rate metrics but also the susceptibility and the resulting effective radiative forcing due to aerosol-cloud interactions (ERF_{aci}) estimates, particularly for those equations explicitly incorporating activation rate.

3. Section A3 : Cloud controlling factor analysis,

Have the authors attempted to perform a Variance Inflation Factors analysis to estimate the performance of the CCF (as done in Scott et al. 2020) to avoid any cross-correlation and ensure that the effect seen is due to aerosols only?

Thank you for your comment. In response, we perform a Variance Inflation Factor (VIF) analysis, following the methodology in Scott et al. (2020), to evaluate any potential multicollinearity within CCF analysis. The results indicate that the sulfate mass concentrations (SO₄) and aerosol index (AI) are independent of other environmental factors, supporting the validity of our approach and emphasizing that our estimates of ERF_{aci} are truly attributable to aerosols. We include these VIF results in Figures A1 and A2, along with corresponding explanations in Appendix A3.

4. L457: The authors state that their value is the same as the global value because the polar ocean surfaces are limited in area. Firstly, we can argue whether the region is indeed negligible in terms of area compared to the globe, but their impact could be significantly greater in these regions due to their specificities (polar night/day, ice surface, pristine conditions...). I am currently not convinced that the results can really be generalised to the globe.

Thank you for your comment. As noted in the manuscript, limitations in satellite observations prevent us from obtaining reliable data over land and polar regions (Jia et al., 2019; Gryspeerd et al., 2022; Jia and Quaas, 2023). However, we expand our study domain to cover the area between 60°S and 60°N, and our conclusions remain consistent within this extended area. We believe that excluding the polar regions does not compromise our conclusions, as sulfate mass concentrations are primarily concentrated in major industrial regions, such as East Asia and North America, which are well represented within our study domain.

Additionally, to address your concern, we adjust the text to avoid potential confusion regarding polar region contributions. We now highlight our extrapolation approach using CMIP6 single-forcing experiments, where we apply a scalar multiplier to the domain-average value to estimate a global-average value, as explained in Appendix A6 and illustrated in Figure A3.

5. A3, have the authors considered different cloud regimes as in Scott et al 2022?

Thank you for your comment. In this study, we focus specifically on non-obscured low clouds without distinguishing between cloud types, as our primary objective is to examine observationally constrained global effective radiative forcing from aerosol-cloud interactions and validate it through various estimation methods, based on CMIP6 models. Given the dominant contribution of low-level clouds to ERF_{aci}, focusing on these cloud types is sufficient to represent the ERF_{aci} value (Christensen et al., 2016; Bellouin et al., 2020; Forster et al., 2021). While considering different cloud regimes, as suggested by Scott et al. (2022), could provide valuable insights, the journal's word limit necessitated focusing on non-obscured low clouds to maintain clarity and conciseness. We agree that exploring cloud regime distinctions would be an excellent avenue for future research.

6. Statistical tests and quantification would be welcome to better assess the results instead of “significantly diminishes” for example, etc.

Thank you for your comment. We revise the manuscript to remove the term “significantly” where statistical quantifications are not applied. Additionally, we include *p*-values alongside *r*-values where correlation coefficients are reported for improved statistical rigor.

Minor :

The ACP guidelines mention : “*Appendices: all material required to understand the essential*

aspects of the paper such as experimental methods, data, and interpretation should preferably be included in the main text.” I have found that most ACP papers have data set and methods sections and usually an appendix with important information. I strongly recommend to include the data set and method sections in the main body of the paper and not in the appendix, following the ACP recommendations.

Thank you for your comment and recommendation. To address your concern, we specify the origin of each dataset in the main text for improved clarity. We considered moving the methods for estimating ERF_{aci} in CMIP6 models to the main text in accordance with ACP guidelines; however, due to the strict word limit for *ACP Letters*, we decide to retain the current format. We appreciate your understanding.

L14: “While some studies it is assumed”, I suggest “While some studies assumed”

Thank you for your suggestion. We revise the sentence to “While some studies assume...”.

L15: “Variation in sulfate aerosols”, do the authors mean sulphate aerosol concentration? It could also be aging, coating, etc that would change the aerosol-cloud interaction.

Thank you for your valuable comment. We were indeed referring to sulfate aerosol mass concentrations in the atmosphere. We revise the text to specify “sulfate aerosol mass concentrations” or “sulfate mass concentration” where relevant to avoid any potential confusion.

L15: I think Sulfates are SO₄²⁻ and not SO₄

Thank you for your comment. To clarify, we add a note in L66 stating, “sulfate mass concentration (SO₄; for simplicity, we omit its ionic form)...” to ensure readers understand the notation used throughout the text. Additionally, we note that Randles et al. (2017), the MERRA-2 reference paper, also uses sulfate as SO₄. Therefore, we believe this notation aligns with established conventions and will be clear to readers.

L20: It would be interesting to know how much, on average, the ERF is reduced and less uncertain. A quantification would increase the impact of the abstract.

Thank you for your comment. Given our use of two different aerosol concentration proxies, we instead add quantification of recent climate assessments. The revised sentence now states in L21 “Our results suggest a smaller and less uncertain value of the global ERF_{aci} ($-0.32 \pm 0.21 \text{ W m}^{-2}$ for SO₄, 90% confidence) than recent climate assessments (e.g. $-0.93 \pm 0.7 \text{ W m}^{-2}$, 90% confidence)...”.

L34: All aerosols do not act as CCN, some would act as INP, and some would not interact with clouds.

Thank you for your comment. You are correct that not all aerosols serve as CCN; some act as INP, while others may not interact with clouds at all. In this study, we address aerosol-cloud interactions in a broad context, primarily focusing on aerosols that contribute to CCN. Additionally, this study focuses specifically on aerosol-cloud interactions within low-level clouds, where ice formation is minimal, and CCN-related interactions are most prominent and well-documented (Christensen et al., 2016; Bellouin et al., 2020; Forster et al., 2021). This generalization is intended to streamline the discussion and maintain relevance to our specific focus on CCN-driven interactions.

L47-51: Citations are missing to support the text.

Thank you for your comment. We revise the sentence and add references to support the text in L52-55: “In some studies, the activation rate is not explicitly incorporated into the estimation process of ERF_{aci} as it is implicitly assumed to have a one-to-one relationship (e.g. Chen et al., 2014; Christensen et al., 2016; Douglas and L’Ecuyer 2020; Wall et al., 2022, 2023).”

L62: Have the authors constrained to consider situations where sulphate aerosols are the dominant aerosol type (e.g. more than 80% of the total concentration?) Other aerosols may not be as efficient CCN but they could still bias their results.

Thank you for your comment. The original sentence may have been misleading. Our intent was to convey that sulfate mass concentration is recognized as a primary contributor to cloud droplet formation among various aerosol types (Charlson et al., 1992; McCoy et al., 2018). To enhance clarity, we revise the sentence to state in L69-71: “SO₄ is recognized as a dominant contributor to cloud droplet formation, alongside other aerosol types such as black carbon, organic carbon, sea salt, and dust (Charlson et al., 1992; McCoy et al., 2018).”

L77: I do not think Nd is defined in the paper.

Thank you for your comment. We now define N_d in the introduction section at L51 to ensure clarity.

L81: I would not expect a one-to-one relation. As the authors mentioned it is related to the activation rate but this would mean that SO₄ is the only CCN.

Thank you for pointing this out. Of course, our intention is not to suggest that SO₄ is the only contributor to CCN, but rather to illustrate that not all sulfate aerosols convert to cloud droplets. Additionally, we stated in L69-71 that “SO₄ is recognized as a dominant contributor to cloud droplet formation, alongside other aerosol types...”, which we believe clarifies that SO₄ is not the only CCN. However, to avoid any confusion, we revise the text to replace “one-to-one relation” with the sentence in L92: “...underscoring that not all SO₄ in the atmosphere are converted into cloud droplets.” Furthermore, we add in L98: “This variation may be attributed to differences in local environmental conditions and the role of aerosols in which these clouds

occur (e.g. Douglas and L'Ecuyer, 2019, 2020).". These revisions aim to ensure clarity and prevent misunderstanding.

L82-84 : "show a notably weaker (...) coefficient", but the correlation coefficient, therefore shall we conclude anything from that ?

Thank you for pointing this out. To avoid confusion and improve clarity, we remove the correlation coefficient from Figure 1 and Figure S1, as it was not ideal for visually representing the activation rate.

Fig1: I am not sure what is the date range used by the authors to produce the plot.
Thank you for your comment. We add the date range (January 2003 to December 2019) to the figure caption for clarification.

L85: I do not find the results consistent between AI and SO₄, there are negative values and the results with large regression coefficients are not the same. Can the authors clarify what they mean by consistent?

Thank you for pointing this out. You are correct that there are differences between the AI and SO₄ results. While both exhibit similar regional patterns—such as higher activation rates in stratocumulus regions, consistent with SO₄ findings—the results for $\partial \ln(N_a) / \partial \ln(\text{AI})$ differ in some aspects. To clarify, we now state in L100-101: "Repeating our analysis using AI yields somewhat different results with those for SO₄ though still showing strong positive regression coefficients near continental coasts (Fig. S1)". Additionally, as noted in the manuscript, the differences in regression coefficients may arise from the use of column-integrated quantities (AI from MODIS), which do not capture the aerosol vertical structure. This clarification would help provide further context for the observed discrepancies.

L201 : "lower end", Do they authors mean "higher end" ?

Thank you for pointing this out. We revise the text to "higher end" and add "(less negative)" for additional clarification.

Equation A1: I am not sure to understand the equation, L_n is at the 2.5 horizontal resolution but L and U are at the native resolution but within the 2.5 degree grid cell, is it correct ?

Thank you for pointing this out. To improve clarity, we remove the expression "relative to the area of each grid box," as it causes confusion regarding the resolution of L_n, L and U in Equation A1. All variables are evaluated at the same 2.5° grid point resolution.

Equations A5 and A6, what is needed to account for LWP bins? If I understand, the authors have constrained for LWP, is 30 g cm⁻² bins sufficient? Have they tried smaller bins to see how the results change?

Thank you for your comment. Initially, we considered using liquid water path (LWP) bins to further specify cloud properties based on LWP. However, to maintain consistency and directly compare our results with those of Wall et al. (2022), which do not constrain LWP, we decide to exclude LWP in our equations. Even without this constraint, our results remain consistent, supporting the robustness of our findings.

L558 and L561, I cannot access the webpages:

<https://esgf559node.llnl.gov/projects/cmip6/> and
https://github.com/nicklutsko/Radiative_Forcing_Aerosol_Clouds

Thank you for your comment. I check the web addresses and, in place of the data from Wall et al. (2022) as we extend our domain area to 60°S and 60°N, I create a data repository for the relevant variables used in our study. The data are now accessible at <https://zenodo.org/records/14058556>.

I think all the appendices are not referenced in the main text. For example, I cannot see where sections A3 and A4 are referenced in the main article. It just says "In Appendix A". Again I think most of the part should be in the main text but for the remaining part, I suggest to clearly specify which part is referenced in the main text.

Thank you for pointing this out. We revise the manuscript to specify exactly which appendix sections are referenced in the main text to improve clarity.

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