The response to Reviewer #1:

We sincerely thank the reviewer for their constructive comments and valuable suggestions. Below, we provide specific responses to each comment, highlighted in blue.

1. I had a concern about the layout and structure of the article with most of the technical information (data and methods) in the appendix. I am still confused and compared to the letter format of other journals, there is more information on data and methods in them. I am not familiar with ACP letters, so I leave it to the editor to decide if there is enough information in the main article.

Thank you for your comment. We understand your concern regarding the placement of methodological details in the appendix. In ACP Letters, due to the strict word limit (a maximum of 2,500 words), many published papers present data and methods in the appendix rather than in the main text. For reference, we have cited similar examples below.

Given these limitations, we aimed to balance conciseness with methodological clarity. Our manuscript currently reaches 2,474 words (excluding references), which limits the extent of methodological discussion we can include in the main text. To ensure transparency while adhering to the journal's format, we have provided key methodological details in the appendix while keeping the main text focused on the primary findings and their implications.

While we recognize that some letter-format journals allow for more methodological content in the main text due to a higher word limit, we have followed ACP Letters' formatting conventions. We defer to the editor's judgment on whether the current level of detail is appropriate for this journal and are open to any recommendations for adjustments.

References

Diamond, M. S.: Detection of large-scale cloud microphysical changes within a major shipping corridor after implementation of the International Maritime Organization 2020 fuel sulfur regulations, Atmospheric Chemistry and Physics, 23, 8259–8269, https://doi.org/10.5194/acp-23-8259-2023, 2023.

Ploeger, F., Birner, T., Charlesworth, E., Konopka, P., and Müller, R.: Moist bias in the Pacific upper troposphere and lower stratosphere (UTLS) in climate models affects regional circulation patterns, Atmospheric Chemistry and Physics, 24, 2033–2043, https://doi.org/10.5194/acp-24-2033-2024, 2024.

Teixeira, J., Wilson, R. C., and Thrastarson, H. T.: Direct observational evidence from space of the effect of CO_2 increase on longwave spectral radiances: the unique role of high-spectral-

resolution measurements, Atmospheric Chemistry and Physics, 24, 6375–6383, https://doi.org/10.5194/acp-24-6375-2024, 2024.

2. I am still confused about the extension to the whole domain. It is still based on a hypothesis and the article is not very clear about this (or I miss the sentence). I agree with the authors that sulphate mass concentrations are mainly concentrated in major industrial regions, but the Arctic regions have shown that the effect of aerosols acting as CCN is particularly efficient in the Arctic (up to 8 times more efficient compared to mid-latitudes, see coopman et al., 2018).
Coopman, Q., Garrett, T. J., Finch, D. P., & Riedi, J. (2018). High sensitivity of arctic liquid clouds to long-range anthropogenic aerosol transport. Geophysical Research Letters, 45, 372–381. https://doi.org/10.1002/2017GL075795

Thank you for your insightful comment. We understand your concern regarding the extrapolation of our domain-average ERFaci to a global estimate. As noted in the manuscript, limitations in satellite observations prevent us from obtaining continuous and reliable data over land and polar regions (Jia et al., 2019; Gryspeerdt et al., 2022; Jia and Quaas, 2023). Under these circumstances, to account for the global-average ERFaci from our domain-average ERFaci, we use CMIP6 single-forcing experiments to derive a scalar multiplier (γ) based on the ratio of the multi-model mean of global-average ERFaci_true to domain-average ERFaci_true. Importantly, this approach inherently accounts for regional variations, as γ is derived from a multi-model estimate that includes polar aerosol-cloud interactions. Our analysis finds $\gamma = 0.86$, with a strong linear correlation ($r = 0.92$, $p < 0.001$), ensuring the robustness of our extrapolation methodology. This approach is detailed in Appendix A6 and illustrated in Figure A3.

To further validate our methodology, we perform a sensitivity test using observational dataset following Wall et al. (2022). In this alternative method, we assume that the albedo change associated with ERFaci is approximately uniform across the study domain and the entire globe. Under this assumption, γ is estimated as the ratio of global-mean to domain-mean insolation, yielding a central estimate of 0.92. Notably, this estimate is highly consistent with our model-derived value of 0.86, reinforcing the robustness of our extrapolation approach. Given this consistency, we adopt $\gamma = 0.86$ in this study.

We acknowledge that aerosol-cloud interactions in the Arctic may be more efficient per unit aerosol mass due to unique atmospheric conditions (e.g., pristine environment, polar night/day cycles). If the Arctic's high aerosol-cloud interaction efficiency were fully considered, our global ERFaci estimate could potentially be more negative. However, the efficiency estimates in Coopman et al. (2018) are based on a dataset with limited temporal (March to September, 2005–2010) and spatial coverage (north of 65°N over the ocean). Thus, incorporating these effects with arbitrary weighting (e.g., 2 to 8 times higher efficiency in the Arctic than in the mid-latitudes) could introduce additional uncertainties in our analysis. Nonetheless, we recognize the importance of Arctic aerosol-cloud interactions and their potential influence on global-mean ERFaci estimates.

To address your concern, we have revised the manuscript to clarify the potential uncertainties associated with Arctic aerosol-cloud interactions and their role in our extrapolation methodology. Specifically, we now state in Appendix A6:

"Additionally, following the approach of Wall et al. (2022), we conduct a sensitivity test for γ without relying on climate model results. In this alternative method, we assume that the albedo change associated with ERFaci is approximately uniform across the study domain and the entire globe. Under this assumption, γ is approximated as the ratio of global-mean insolation to domain-mean insolation, yielding a central estimate of 0.92. Notably, this value is highly consistent with our model-derived estimate of 0.86, supporting the robustness of our extrapolation approach. Given this consistency, we adopt $\gamma = 0.86$ in this study.

Even though our study domain captures the primary anthropogenic aerosol sources, particularly near major industrial regions in Eurasia and North America, and our multi-model mean extrapolation inherently accounts for aerosol-cloud interactions outside our domain, recent studies have highlighted their significant influence in polar regions (e.g., Coopman et al., 2018). Aerosol-induced cloud property changes in the Arctic may be more efficient per unit aerosol mass than at mid-latitudes due to the greater susceptibility of Arctic clouds to aerosols. Incorporating these effects could lead to a more negative global-mean ERFaci estimate. The role of Arctic aerosol-cloud interactions warrants further investigation, and future research incorporating more comprehensive observational constraints would be valuable."

Additionally, we have revised line 144 of the manuscript to explicitly state how our global estimate is extrapolated:
"To estimate global-average ERFaci_obs from our domain-average ERFaci_obs, we multiply our domain estimate by a scalar multiplier, γ, which represents the ratio of multi-model mean of global-average ERFaci_true to domain-average ERFaci_true (Appendix A6)."

References

Coopman, Q., Garrett, T. J., Finch, D. P., and Riedi, J.: High Sensitivity of Arctic Liquid Clouds to Long-Range Anthropogenic Aerosol Transport, Geophysical Research Letters, 45, 372–381, https://doi.org/10.1002/2017GL075795, 2018.

Gryspeerdt, E., McCoy, D. T., Crosbie, E., Moore, R. H., Nott, G. J., Painemal, D., Small-Griswold, J., Sorooshian, A., and Ziemba, L.: The impact of sampling strategy on the cloud droplet number concentration estimated from satellite data, Atmospheric Measurement Techniques, 15, 3875–3892, https://doi.org/10.5194/amt-15-3875-2022, 2022.

Jia, H. and Quaas, J.: Nonlinearity of the cloud response postpones climate penalty of mitigating air pollution in polluted regions, Nat. Clim. Chang., 13, 943–950, https://doi.org/10.1038/s41558-023-01775-5, 2023.

Jia, H., Ma, X., Quaas, J., Yin, Y., and Qiu, T.: Is positive correlation between cloud droplet effective radius and aerosol optical depth over land due to retrieval artifacts or real physical processes?, Atmospheric Chemistry and Physics, 19, 8879–8896, https://doi.org/10.5194/acp-19-8879-2019, 2019.

Wall, C. J., Norris, J. R., Possner, A., McCoy, D. T., McCoy, I. L., and Lutsko, N. J.: Assessing effective radiative forcing from aerosol–cloud interactions over the global ocean, Proceedings of the National Academy of Sciences, 119, e2210481119, https://doi.org/10.1073/pnas.2210481119, 2022.

3. I disagree with the authors about the convention of referring to SO42- as SO4 and think it should be changed throughout the text. I agree that SO4 could be clear to readers, but I do not think it is correct to write it in this way. I will follow the editor decision with this matter.

Thank you for your concern. Based on your suggestion, we have decided to use "SO_4^{2-}" instead of "SO_4" throughout the manuscript. We believe this revision enhances readability and aligns with conventional chemical notation. We appreciate your feedback.

4. I still have concerns about using only SO4 as an aerosol. On line 110 of the manuscript, the authors state that it is 64% smaller with activation than without. Taking into account other aerosols might potentially diminish the difference. The information with the AI is more impactful in this regards.

Thank you for your comment. We understand your concern regarding the use of SO_4 as the primary aerosol proxy. Our decision is based on its well-documented dominant role in aerosol-cloud interactions and its strong correlation with cloud droplet number concentrations compared to other aerosol types (Charlson et al., 1992; Stevens, 2015; McCoy et al., 2018). To provide a broader perspective and strengthen our analysis, we also included the Aerosol Index (AI) as an additional aerosol proxy. The consistency in ERFaci estimates derived from both proxies highlights the robustness of our findings.

Sulfate has been widely used in previous studies as a primary proxy for aerosol-cloud interactions, particularly in the context of radiative forcing (e.g., Wall et al., 2022; Gryspeerdt et al., 2023). Additionally, SO_4 concentrations at 925 hPa offer a more direct representation of CCN availability near the cloud base, which is crucial for assessing cloud microphysical processes (Painemal et al., 2017). In contrast, AI from MODIS represents a column-integrated aerosol quantity that does not account for vertical aerosol distribution, making it less precise in capturing aerosol-cloud interactions. Nevertheless, its inclusion in our study provides a complementary perspective on aerosol proxy.

While considering additional aerosol types could further refine the analysis, SO_4 remains widely used and physically relevant proxy for estimating ERFaci. Given the consistency of ERFaci

estimates across both SO_4 and AI, we are confident that our approach provides a robust and comprehensive assessment. We appreciate your feedback and believe that the use of these two proxies strengthens the reliability of our findings.

To further clarify this point, we have revised line 50 of the manuscript to emphasize the dominant role of sulfate in ERFaci:

"Sulfate aerosol is recognized as a dominant contributor to ERFaci as well as cloud droplet formation, alongside other aerosol types such as black carbon, organic carbon, sea salt, and dust (Charlson et al., 1992; Stevens, 2015; McCoy et al., 2018)."

Additionally, we have highlighted the column-integrated feature of AI in Appendix 1.3: "However, it is important to note that since AI provides column-integrated quantities and does not account for the vertical profile, it may not accurately capture aerosol concentrations in low-level clouds, which are the focus of our study."

References

Charlson, R. J., Schwartz, S. E., Hales, J. M., Cess, R. D., Coakley, J. A., Hansen, J. E., and Hofmann, D. J.: Climate Forcing by Anthropogenic Aerosols, Science, 255, 423–430, https://doi.org/10.1126/science.255.5043.423, 1992.

Gryspeerdt, E., Povey, A. C., Grainger, R. G., Hasekamp, O., Hsu, N. C., Mulcahy, J. P., Sayer, A. M., and Sorooshian, A.: Uncertainty in aerosol–cloud radiative forcing is driven by clean conditions, Atmospheric Chemistry and Physics, 23, 4115–4122, https://doi.org/10.5194/acp-23-4115-2023, 2023.

McCoy, D. T., Bender, F. A.-M., Grosvenor, D. P., Mohrmann, J. K., Hartmann, D. L., Wood, R., and Field, P. R.: Predicting decadal trends in cloud droplet number concentration using reanalysis and satellite data, Atmospheric Chemistry and Physics, 18, 2035–2047, https://doi.org/10.5194/acp-18-2035-2018, 2018.

Painemal, D., Chiu, J.-Y. C., Minnis, P., Yost, C., Zhou, X., Cadeddu, M., Eloranta, E., Lewis, E. R., Ferrare, R., and Kollias, P.: Aerosol and cloud microphysics covariability in the northeast Pacific boundary layer estimated with ship-based and satellite remote sensing observations, Journal of Geophysical Research: Atmospheres, 122, 2403–2418, https://doi.org/10.1002/2016JD025771, 2017.

Stevens, B.: Rethinking the Lower Bound on Aerosol Radiative Forcing, Journal of Climate, 28, 4794–4819, https://doi.org/10.1175/JCLI-D-14-00656.1, 2015.

Wall, C. J., Norris, J. R., Possner, A., McCoy, D. T., McCoy, I. L., and Lutsko, N. J.: Assessing effective radiative forcing from aerosol–cloud interactions over the global ocean, Proceedings of the National Academy of Sciences, 119, e2210481119, https://doi.org/10.1073/pnas.2210481119, 2022.

5. Some of the texts in the appendix are referenced in the main text : Appendix A, A1.2, A1.1 for example.

Thank you for pointing this out. We intentionally reference specific sections of the appendix, such as Appendix A3 and A4, in the main text to provide additional methodological details while keeping the manuscript within the word limit.

However, if your concern is that certain sections of the appendix, such as Appendix A1.1 and A1.2, which primarily contain data details rather than methodological explanations, are not explicitly referenced in the main text, this was a deliberate decision to enhance readability and avoid unnecessary redundancy. We sincerely appreciate your thoughtful feedback and believe that this approach does not compromise the clarity or comprehension of the main text.