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Comment on acp-2022-642

Anonymous Referee #1

Referee comment on "Uncertainty in aerosol–cloud radiative forcing is driven by clean conditions" by Edward Gryspeerdt et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-642-RC1, 2022

Review of Uncertainty in aerosol-cloud radiative forcing is driven by clean Conditions by Edward Gryspeerdt, Adam C. Povey, Roy G. Grainger, Otto Hasekamp, N. Christina Hsu, Jane P. Mulcahy, Andrew M. Sayer, and Armin Sorooshian

This short letter describes analysis of satellite and global model output data to show that uncertainty in the aerosol impact on clouds is largely driven by situations with little aerosol, i.e., relatively clean conditions. The behavior of clouds in clean conditions is found to be responsible for much of the diversity in both model and observational estimates of the Twomey forcing (radiative forcing from aerosol-cloud interactions, RFaci). This is an important and novel result, which provides another piece of evidence that understanding aerosol-cloud interactions in pristine conditions is important for quantifying aerosol forcing on climate (see e.g., Carslaw et al., 2013; or McCoy et al., 2020). As such, I strongly recommend publication as a letter in ACP. I provide some comments and suggestions for revision, but in my view the manuscript requires only modest changes.

The cloud droplet concentration susceptibility to aerosol (beta = $-d\ln N_d/d\ln A$) is typically established in observations by correlating cloud droplet concentration with an observed aerosol parameter (AOD, AI, CCN, etc…). Observations in the paper, and in some previous studies, indicate that the relationship between N_d and A is not well explained using a simple power law. The relationship tends to flatten for high A, where there are physical arguments for saturation based on droplet activation theory. Here, however, the authors demonstrate here that the N_d-A relationship also tends to flatten for low A, a result which is shown to hold regardless of whether A is represented by aerosol index (AI), sulfate mass loading from reanalysis, or an observational proxy for CCN concentration.

The paper demonstrates that if beta is determined using only relatively polluted aerosol levels, then there is excellent agreement between models (beta exhibits only weak intermodel spread). This gives some confidence that the models are accurately representing Nd variability and its dependence on aerosol when aerosol loadings are relatively high. However, if the entire range of A is considered, the values of beta exhibit a wider spread, but the full range beta is far better correlated with RFaci in the models, indicating the importance of being able to accurately quantify the Nd-A relationships across the entire range of A. The wider spread of model-derived beta values indicates greater intermodel uncertainty and therefore a relatively poor constraint on RFaci. The observational estimates of beta for different choices of the A variable also disagree more strongly when the full range of A is considered, which may indicate problems constraining aerosol properties in clean air masses. A variety of reasons could help explain this, including low signal to noise ratio in aerosol retrievals for low A. Future global aerosol measurements will need to embrace the need to function well at lower aerosol loadings than they currently do.

My only significant comment pertains to the distinction between ERFaci (Twomey + adjustments) and RFaci (Twomey only, no adjustments). This distinction should be clarified early on. Since only the term "radiative forcing" (RFaci) is used throughout the manuscript, I am assuming that the model simulations used in this study *do not* include adjustments to aerosol. This distinction is quite important, because recent studies appear to suggest that adjustments, and especially cloud cover adjustments, may be carrying a large fraction of the effective radiative forcing (Chen et al., 2022). Other studies/models show weaker adjustments. So, although beta is perhaps the leading source of uncertainty in RFaci, it is not clear that it is as important for ERFaci. The authors provide a comment on this on line 34-36, no quantitative correlation between susceptibility and ERFaci is provided to firmly establish this.

The distinction seems again to be blurred in Line 4: "….the diversity of which explains much of the variation in radiative forcing in global climate models." This suggests that role for cloud adjustments in driving variation in radiative forcing in models is small, or is that irrelevant here because radiative forcing does not include cloud adjustments?

Line 21: Zelinka et al. (2014) examines ERFaci, not RFaci. Is this distinction important?

References

McCoy, I. L., McCoy, D. T., Wood, R., Regayre, L., Watson-Parris, D., Grosvenor, D. P., Mulcahy, J. P., Hu, Y., Bender, F. A.-M., Field, P. R., Carslaw, K. S., & Gordon, H. (2020). The hemispheric contrast in cloud microphysical properties constrains aerosol forcing. Proceedings of the National Academy of Sciences, 117(32), 18998–19006. https://doi.org/10.1073/pnas.1922502117