Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-36-RC1, 2020

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Quantifying Cloud Adjustments and the Radiative Forcing due to Aerosol-Cloud Interactions in Satellite Observations of Warm Marine Clouds" *by* Alyson Douglas and Tristan L'Ecuyer

Anonymous Referee #1

Received and published: 26 January 2020

The authors utilize remote sensing observations and a regime-based approach to isolate the effects of varying aerosol index on cloud microphysical (1st indirect effect) and cloud macrophysical properties (adjustments). The authors utilize regimes of above-cloud RH and stability. LWP is binned to account for variations in cloud state in each regime. The results show that in some regions adjustments and the first indirect effect have opposing signs. The authors also show that as LWP increases the radiative response to AI saturates. The analysis presented here satisfies the important problem of separating variability due to meteorology from aerosol-cloud interactions (aci). The

authors find a relatively weak ERFaci from warm-topped clouds over oceans, which appears to be due to dimming in regions in the equatorial Atlantic and Indian ocean.

While I appreciate that the authors are applying the methodology developed in a previous study, it is hard to understand what is being done and I think the authors could briefly summarize their methodology to allow readers to more efficiently refer to DL19. The description of the observational data sets could be much more substantial. It is confusing what observational and modelling data is being used for what. In some cases it appears that observational data sets that are not appropriate are being used, but it is hard to confirm this from the data section. One solution that might make this un-ambiguous would be to create a table of variables and data sources.

A critical issue with this paper is use of area-mean LWP (in-cloud LWP*CF) from microwave when the authors imply they are using in-cloud LWP based on wording in the paper (ln 153). From reading the discussion in DL19 I believe that scene-mean LWP from AMSR is just used to filter data into rough bins, and does not play a role in the analysis beyond this. While this is probably not a big problem, the authors may want to clarify what the footprints of the different data sets are that they are using, possibly with a diagram overlaid over an actual satellite image to allow readers who are less familiar with remote sensing to contextualize what is being shown, especially because the authors are using active instruments averaged along track with passive instruments. In particular, in this regard I am confused how the authors are overlapping along-track averaged CF from Cloudsat-CALIPSO with AMSR LWP and a diagram might be helpful. A nice image of the actual cloud field from MODIS on the background would be helpful to readers trying to contextualize the retrievals in terms of cloud features.

The authors ultimately present a correlative study to predict ERFaci (or at least ERFaci for warm-topped clouds over oceans- see comments below). Characterizing covariance is important but does not guarantee an accurate prediction. In the case of aerosol-cloud adjustments in particular, there is not a unique causality flowing from aerosol to cloud (Wood et al., 2012; Gryspeerdt et al., 2019). In this context, and because their ERFaci is rather weak compared to other studies it seems possible that their analysis conflates aci with precipitation scavenging and other confounders (Gryspeerdt et al., 2019), which would tend reduce correlation strength between aerosol and cloud amount (eg precipitation scavenging is strongest when there is a lot of cloud and there tend to be less cloud and more aerosol off the coast of continents). The authors need to either apply their analysis in a GCM simulating PI and PD (Gryspeerdt et al., 2017; Gryspeerdt et al., 2016; McCoy et al., 2019; Costa-Surós et al., 2019) to make sure that their analysis methodology has predictive power, or examine the response of cloud to some sort of transient change in aerosol (Malavelle et al., 2017; Toll et al., 2019) and make sure that their analysis trained over different data can predict the response to the transient change in AI. Without these falsification tests of their predictions, it is unclear what predictive use their correlation model has in that there is no way to falsify their predictions. Even an approximate calculation using model LWP, CF, SW, and AI without any complex output along the satellite overpass (which doesn't appear to be a major source of error compared to problems from low aerosol amount as shown in Ma et al. (2018)) would provide a much more powerful validation of what the authors are hypothesizing is the ERFaci.

The authors need to refer to their ERFaci as ERFaci_liquid-topped_over_oceans (or at least that is my take from the methodology and Eq 9). Is it possible to use this metric regarding warm-topped maritime clouds, and what they know about the relative occurrence of the clouds that this analysis is performed on, to allow them to extrapolate to global ERFaci? A similar strategy is employed in Costa-Surós et al. (2019) to related forcing over Germany to global forcing.

Specific changes:

Pg 1 ln 3: ERFaci is a combination of microphysical (RFaci) and macrophysical changes (adjustments) and the latter could be further split into changes in extent and thickness(Gryspeerdt et al., 2019; Gryspeerdt et al., 2017; Gryspeerdt et al., 2016). As written this implies that thickness stays constant and the only possible adjustment

is CF. I understand now that this is more like the intrinsic extrinsic separation in other studies (Christensen et al., 2017), but this would be better to clarify in the abstract.

Pg. 2 ln 40: The goals of DL19 overlap a lot with the goals of the present study. A sentence like 'The present study expand on DL19 in the following ways:' would be helpful. I believe the primary difference between these studies is the inclusion of adjustments, but it would be helpful to state that explicitly for readers to rapidly ingest what is happening.

Pg. 3 ln 85: It would help readers to quickly process what data sets are being used to describe what variable to use subheaders here (2.1 Data, 2.1.1 Warm cloud fraction). This is stylistic, but I found it hard to understand where precipitation measurements were coming from. I think that it would help a lot to have a table of what the precise data sets used are, especially since some of the remote sensing data sets being used may be inappropriate, but it is unclear if they are actually used (eg AMSR rain rates, although I believe these are not used despite being mentioned).

Pg4 ln 124: is the material not shown in the citation? If it's in the citation no need to put not shown here.

Pg 4 ln 125: Swelling is a key issue in trying to understand adjustments. I believe that swelling is not an issue for SPRINTARS because the model can be internally consistent, but an additional comment is needed about MACC aerosol swelling. It's unclear that MACC can fully correct for swelling given the very complex way that swelling occurs (Christensen et al., 2017; Twohy et al., 2009). This needs to be explained and caveated. Also, why mix MACC aerosol and MERRA2 meteorology? MERRA2 produces a very similar aerosol reanalysis to MACC and this would avoid confusing MERRA2 meteorology with aerosol reanalysis in a different framework. Also- how are SPRINTARS and MACC not sensitive to precipitation scavenging? Presumably both data sets have a precipitation sink of aerosol otherwise it would be very hard to maintain realistic aerosol.

Pg5 ln126: The MACC AI is effectively satellite AI because it is nudged to satellite radiances. At some point in this paper it is necessary to caveat the use of AI with the results of Ma et al. (2018), which found that satellite inability to detect low aerosol loading biases inferred aci.

Pg5 ln 129: It would be good to note that microwave LWP is area*in cloud LWP. In this context it is a little confusing relating this to Twomey on line 154 because that is for in-cloud LWP, not area mean LWP.

Pg.5 Ln140: This methods section is really short. I understand that the authors refer to DL19, but I think it would help readers evaluate this paper more quickly if a paragraph or so was taken to summarize DL19.

Pg. 5 ln 147: The authors refer to partitioning into precipitating and non-precipitating clouds. I am not clear how this is done. On line 245 it looks like 2C-RAIN-PROFILE is being used- this needs to be caveated that it will only see relatively heavy precipitation, but will miss light rain events. Other parts of the methodology makes it sound like AMSR-E precipitation is being used, which is problematic due to the AMSR-E precipitation just being a partitioning of condensed liquid by SST.

Pg. 6 Eq3-6: how do the authors account for CF being bounded between 0-1 in this calculation?

Pg. 7 Ln 197: The authors assert that they have accounted for the effects of precipitation on the aerosol-cloud-precipitation system. This is not supported by any evidence or citations, but is an important justification of the validity of the analysis presented here.

Pg. 8 ln 221: The authors assert that by binning LWP they reduce the chances of buffering. One thing that should be mentioned in this study is that AI and LWP will naturally anti-correlate due to precipitation and scavenging correlating with cloudiness (eg LWP or CF) (Wood et al., 2012) and due to air mass history leading to both drier

and more aerosol-laden air (Gryspeerdt et al., 2019). These non-causal relationships are not meaningful to ERFaci, but can substantially affect the covariability of cloud macrophysical properties and aerosol, and thus the inferred aci strength (McCoy et al., 2019). It is possible that the LWP binning and precipitation stratification reduce this effect. However, the authors must show some demonstration of the predictive ability of this method by either (1) applying it to GCM data (in this case SPRINTARS) and showing that their methodology when applied in a GCM can accurately reproduce the GCM response to enhance aerosol as in Gryspeerdt et al. (2016) or McCoy et al. (2019) – or – (2) examining one of the transient aerosol emissions identified in recent studies (Malavelle et al., 2017; Toll et al., 2019) and see if their characterization of sensitivity of cloud to aerosol has some predictive ability. Without this sort of test there is no guarantee that the inferred ERFaci_warm-topped_oceanic is accurate.

Pg 9 Ln 261: The authors find an ERFaci that at the very weakest end of what would be expected based on existing best-estimates (Bellouin et al., 2019). This is of course completely fine, but it would be interesting for the authors to add some discussion of why their forcing is so relatively weak compared to other empirical estimates of ERFaci from observations. The authors do cite the AR5 range, but this is for the range of GCM estimates, which may not be as appropriate to consider their results relative to as observational constraint studies. I suspect that this is partially because the authors are not really looking at ERFaci, but ERFaci_warm-topped_oceanic. As such I recommend the authors do not use the terminology ERFaci. In the interest of relating this result to forcing the authors could consider using this methodology applied to GCMs (as noted above I view this as a necessary condition to this analysis) and then using the GCMs to extrapolate this result to ERFaci as in Costa-Surós et al. (2019).

Pg 10 ln 300: An alternative explanation of the weakening precipitation effect in clouds with higher LWP may be that precipitation increases with LWP, which means that precipitation scavenging becomes larger, which in turn means that the true adjustment strength is obscured by non-causal covariance between aerosol and cloud macro-physics (see discussion in McCoy et al. (2019)).

Figure 7 and ln 456: The authors find a large ERFaci in the SH, which is really surprising given the very small change in anthropogenic aerosol in these regions. Figure 1 shows change in AI, but it is a bit hard to distinguish small changes from zero and the authors may want to consider some sort of log normalization to their color scale. However, strong ERFaci exists along a line around 40°S, which is hard to square with studies examining pristine days in the PD (Hamilton et al., 2014). That is to say, the pattern of ERFaci in this study is dramatically different than the RFari shown in, for example, aerocom (Myhre et al., 2013).

Figure 7: While I think it's good to pursue analysis to its conclusion by applying it to all data, I am surprised at the positive RFaci and CA in the tropics. Can the authors comment on whether their analysis is sensitive to retrieval errors in convective cloud? In particular, a positive forcing due to RFaci is quite unusual- while it may be due to biomass burning aerosol above cloud in some regions via semi-direct effects or blocking reflective light (so not really aci) (Bellouin et al., 2019), the appearance of a positive RFaci seems to be more related to SST, than aerosol type given its appearance over the tropics, and far away from strong aerosol sources.

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-36, 2020.

Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-36-RC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantifying Cloud Adjustments and the Radiative Forcing due to Aerosol-Cloud Interactions in Satellite Observations of Warm Marine Clouds" *by* Alyson Douglas and Tristan L'Ecuyer

Anonymous Referee #2

Received and published: 13 February 2020

Review of: Quantifying Cloud Adjustments and the Radiative Forcing due to Aerosol-Cloud Interactions in Satellite Observations of Warm Marine Clouds By Douglas and L'Ecuyer

This study uses satellite observations with the addition of model aerosol data and re-analysis meteorological data to calculate the effective radiative forcing due to cloud-aerosol interaction in warm clouds over the oceans. The authors decompose the forcing to two components: due to the Twomey effect (RFaci), and due to cloud adjustments (CA), which in this case include only changes in cloud cover (without including changes

in LWP). The analysis is conducted also regionally and as a function of LWP, inversion strength and RH in the free troposphere. The binning according to the last two criteria is done to account for the meteorological dependency. The calculation is also done separately for precipitating and non-precipitating clouds. I think that this paper presents some interesting results that worth being published. However, I think that the paper includes some limitations that are not all fully acknowledged in the manuscript. Hence, including a more comprehensive discussion about these limitations and maybe weakening the conclusions accordingly will improve the paper.

General comments

1) If I understand correctly, calculating the radiative forcing based on the multiplication of the susceptibility calculated in present day with the total change in AI between present day and preindustrial assume linearity of the susceptibility with time. As you show that the susceptibility is a function of the environmental conditions and it is known that the environmental conditions changed, it is not clear how valid is this assumption. In addition, I think that your calculation assumes that the frequency of occurrence of each bin of EIS, RH and LWP remain the same between PD and PI (as you do not account for changes in the frequency of occurrence -eq. 9). I can't see any reasons for that to be true. Hence, and because of the large uncertainty in PI aerosol conditions, it might be better to stay only with the susceptibility calculations and not present the forcing calculations. I leave it to the authors to decide. 2) Feedbacks between clouds and the environmental conditions are not discussed and accounted for sufficiently. It is known that the environmental conditions may change differently under different aerosol conditions. In particular, the EIS and RH (maybe not at 700mb but definitely below that) may be affected by the clouds feedback on the environmental conditions differently under different aerosol conditions. In addition, direct aerosol-radiation interaction may influence the environmental conditions. Hence, the binning according to the meteorological conditions may not be independent of the aerosol conditions. I suggest to add a discussion about that. In addition, the separation to precipitating/non-precipitating

conditions could be, under certain conditions, due to aerosol effect (total rain suppression could be found in shallow clouds under polluted conditions). This effect is not discussed and you treat it as if it was external. 3) Co-variability between aerosol and cloudiness and the uncertainty in the causality relationships are not discussed enough. I appreciate that binning the data according to EIS and RH at 700 mb may reduce the role of co-variability between aerosol and cloudiness. However, some co-variability may still remain. For example, it was previously shown (Nishant and Sherwood, 2017) that under some conditions, near surface wind speed have a positive correlation with both aerosol concentration and cloudiness (CF in this case). It is possible, and even expected, that wind speed will be partially corelative with EIS and RH but not sure to what extent. I suggest to add a dissection about those limitations. 4) Uncertainties due to the semi-direct effect are not mentioned. Form satellite observations it is impossible to distinguish between the aerosol microphysical effect and the semi-direct effect but the latter is very likely to affect your calculations. I suggest to include a discussion about that. 5) Referring to the forcing only from warm marine cloud as ERFaci and RFaci might be confusing with the total estimations for all cloud. I appreciate that you mention the focus on warm clouds over the ocean directly in the tile and in many other places but I still think that the use of general terms here could be confusing. 6) At many places along the manuscript you mention "buffering" as if it was an artefact that one should avoid in his analysis (i.e. "While LWP being held approximately constant accounts for some variability in the meteorology, explicitly holding the stability and free atmospheric contributions fixed within regimes of EIS and RH700 will further control buffering and modulation of λ by the environment."). I think that if indeed clouds under different aerosol levels change differently the environmental conditions to reduce the total aerosol effect, that is something important to understand. In addition, I think you don't properly define what you mean by "buffering". That term could be used to describe many mechanisms.

Specific comments:

L3: CA is not defined hear. Consider writing in full cloud adjustments.

L10: if RFaci and CA counteract and the total effect is small I would say that it could be attributed to damped susceptibility (or buffering). Why is it "erroneously"?

L21: what do you mean by "cloud forcing"? is it the cloud radiative effect? I think it is better not to use forcing here and stick with the common definition of radiative forcing.

L68-72: consider adding here that the sign of the effect was also shown to be a function of the background aerosol concentration.

L82: the non-monotonic response was shown for other cloud properties (such as cloud fraction and top height) as well as for precipitation. Hence, I don't understand why is it important to separate specifically this effect from the rest.

L105, L109, L141 and other places: again, maybe better to use radiative effect here instead of forcing.

L119: SPRINTARS was run (in the paper you are citing) in a T21 resolution ($\sim$5.6o) and hence is not "cloud resolving" at all.

L250-258: I couldn't really understand how the uncertainty was calculated. I think more details are needed for it to be reproducible. What is the magnitude of error added to PI and PD AI estimations? How did you choose this magnitude?

L265: you are comparing here the estimated forcing for only warm cloud over the ocean with the total estimation from the IPCC report. I don't think this competition is valid. In addition, in the introduction you cited a few papers showing that most of the ERFaci is coming from warm clouds over the ocean. How that can go together with the relatively low estimations you are getting for warm cloud compared to the total forcing?

L310: you cite here a paper focusing on deep convective clouds. Consider adding papers discussing warm cloud invigoration.

L 312: I don't understand the claim here. Why determining the casualty of aerosol effect on LWP is more difficult than for CF?

L320: why is that a sign of “buffering”? it just means that the aerosol effect is non-monotonic and change sign. The aerosol level at which the sign flip is a function of the environmental condition as was shown before.

L417: the possible change in precipitation could also be relevant between PI and PD making point 1 (general comments above) even more critical.

L426-430: you don’t mention here, at the beginning of the conclusion section, that these estimations are only relevant for warm cloud over the oceans. It could look like you are giving general estimations here.

L442: I think that this could also be due to the semi-direct effect of absorbing aerosols.

L445: again, if RFaci and CA counteract and the total effect is small I would say that it could be refer to as “buffering”.

Technical comments L102 and L107: ECWMF -> ECMWF? Anyway, should be written in full (and maybe also add a citation).

L401: “on the both the”

Reference Nishant, N., and Sherwood, S. C.: A cloudâĂŘresolving model study of aerosolâĂŘcloud correlation in a pristine maritime environment, Geophysical Research Letters, 44, 5774-5781, 2017.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-36, 2020.

Reviewer 2 Response

This study uses satellite observations with the addition of model aerosol data and reanalysis meteorological data to calculate the effective radiative forcing due to cloud aerosol interaction in warm clouds over the oceans. The authors decompose the forcing to two components: due to the Twomey effect (RFaci), and due to cloud adjustments (CA), which in this case include only changes in cloud cover (without including changes in LWP). The analysis is conducted also regionally and as a function of LWP, inversion strength and RH in the free troposphere. The binning according to the last two criteria is done to account for the meteorological dependency. The calculation is also done separately for precipitating and non precipitating clouds. I think that this paper presents some interesting results that worth being published. However, I think that the paper includes some limitations that are not all fully acknowledged in the manuscript. Hence, including a more comprehensive discussion about these limitations and maybe weakening the conclusions accordingly will improve the paper.
We thank the reviewer for taking time to read our manuscript and provide constructive feedback. We will now go through and address each of their points.

1) If I understand correctly, calculating the radiative forcing based on the multiplication of the susceptibility calculated in present day with the total change in AI between present day and preindustrial assume linearity of the susceptibility with time. As you show that the susceptibility is a function of the environmental conditions and it is known that the environmental conditions changed, it is not clear how valid is this assumption. In addition, I think that your calculation assumes that the frequency of occurrence of each bin of EIS, RH and LWP remain the same between PD and PI (as you do not account for changes in the frequency of occurrence -eq. 9). I can't see any reasons for that to be true. Hence, and because of the large uncertainty in PI aerosol conditions, it might be better to stay only with the susceptibility calculations and not present the forcing calculations. I leave it to the authors to decide.
We agree that the frequency of occurrence for each regime changes throughout time, however we believe that it is useful to provide an estimate of the ERFaci for warm clouds under the assumption that the cloud and environmental regimes have not changed enough to significantly alter the distribution of states across our relatively coarse LWP, EIS, and RH bins. A bin resolution of 10% in EIS, RH, and LWP is adopted to distinguish regimes in the present study. Even if these parameters have changed since pre-industrial times, it is unlikely that the changes are of a magnitude comparable to this bin resolution which would be required to substantially alter the distribution of states. *To place aerosol-cloud interaction effects into the context of other climate forcings, we feel it is important estimate how these susceptibilities, regime constraints, and regional variations combine to impact the overall estimated forcing. Never-the-less we focus on the sensitivities throughout the paper, with a majority of the results focused on how these change as a constraint is enforced. Future*

work will include evaluating if climate models accurately capture current regime trends and examining how regimes may have changed since the pre-industrial times.

2) Feedbacks between clouds and the environmental conditions are not discussed and accounted for sufficiently. It is known that the environmental conditions may change differently under different aerosol conditions. In particular, the EIS and RH (maybe not at 700mb but definitely below that) may be affected by the clouds feedback on the environmental conditions differently under different aerosol conditions. In addition, direct aerosol-radiation interaction may influence the environmental conditions. Hence, the binning according to the meteorological conditions may not be independent of the aerosol conditions. I suggest to add a discussion about that. In addition, the separation to precipitating/non-precipitating conditions could be, under certain conditions, due to aerosol effect (total rain suppression could be found in shallow clouds under polluted conditions). This effect is not discussed and you treat it as if it was external.
Feedbacks between the clouds, the environment, and the aerosol are much harder to constrain as these are non-linear, time dependent, and occur on multiple time scales. While we agree that aerosol may alter some environmental conditions, the degree to which aerosol impacts the environment is much less than the degree to which aerosol affects clouds and/or clouds affect the environment. The free atmosphere should be minimally impacted by clouds or aerosol, except for in the highest humidity regime, where deep convection detrainment may lead to local changes in the RH_{700}. We agree that aerosol-radiation interactions can alter the environment. We have added some caveats to the Methodology and Observations 2.3 Regimes:

> *"While binning our observations by environmental regime should control for some modulation the environment has on aerosol-cloud interactions, it does not fully capture aerosol-environment interactions. For example, in some regions such as off the coast of Africa, biomass burning results in smoke layers that absorb incoming radiation and warm the atmosphere (Cochrane, 2019). This could affect the humidity and temperature of the local environment. Environmental regime constraints would capture how the altered environment may regulate aerosol-cloud interactions, but separation into such regimes does not address how the aerosol has impacted the environment."*

3) Co-variability between aerosol and cloudiness and the uncertainty in the causality relationships are not discussed enough. I appreciate that binning the data according to EIS and RH at 700 mb may reduce the role of co-variability between aerosol and cloudiness. However, some co-variability may still remain. For example, it was previously shown (Nishant and Sherwood, 2017) that under some conditions, near surface wind speed have a positive correlation with both aerosol concentration and cloudiness (CF in this case). It is possible, and even expected, that wind speed will be partially correlative with EIS and RH but not sure to what extent. I suggest to add a dissection about those limitations.
We agree that not all covariability will be limited by our regime constraints as they are do not encompass all meteorological variability of the boundary layer nor control

for all processes that may impact clouds, aerosols, or their interactions. We have added to Methodology and Observations 2.3 Regimes:

> *"Using EIS and RH_{700} does not guarantee to limit all covariability between the environment, aerosols, clouds, and their interactions. Some covariability may still exist, such as surface winds that may affect both clouds and aerosol (Nishant and Sherwood, 2017). These constraints only account for the major environmental controls on clouds and aerosol-cloud interactions, some more minor or less* common environmental controls may still exert an influence on our results."

We have also added in Results and Discussion section 3.3 Constrained by local meteorology

> "It is possible with additional constraints, understanding how other components of the meteorology is affecting these terms would become more clear."

4) Uncertainties due to the semi-direct effect are not mentioned. From satellite observations it is impossible to distinguish between the aerosol microphysical effect and the semi-direct effect but the latter is very likely to affect your calculations. I suggest to include a discussion about that.

In Results and Discussion section 3.6 we do discuss how the semi-direct effect may be influencing our estimates of the RFaci. We have added, for further clarity, in Results and Discussion section 3.3. Constrained by local meteorology

> *"It is also possible lambda_RFaci is impacted by some semi-direct effects by smoke aerosol which would lead to a cloud dimming and positive susceptibility. Semi-direct effects are not accounted for by our methodology, however aerosol within the cloud layer could lead to cloud breakup processes, a dimmer albedo, and changes to the local environment by the absorbing aerosol."*

5) Referring to the forcing only from warm marine cloud as ERFaci and RFaci might be confusing with the total estimations for all cloud. I appreciate that you mention the focus on warm clouds over the ocean directly in the tile and in many other places but I still think that the use of general terms here could be confusing.

We agree with your suggestion. Sometimes it is easy to focus on warm clouds and forget other clouds are important to the climate too. We have changed ERFaci, RFaci, and CA to be $ERFaci_{warm}$, $RFaci_{warm}$, CA_{warm} in order to remind the reader these results are for only one cloud type.

6) At many places along the manuscript you mention "buffering" as if it was an artefact that one should avoid in his analysis (i.e. "While LWP being held approximately constant accounts for some variability in the meteorology, explicitly holding the stability and free atmospheric contributions fixed within regimes of EIS and RH700 will further control buffering and modulation of λ by the environment."). I think that if indeed clouds under different aerosol levels change differently the environmental conditions to reduce the total aerosol effect, that is something important to understand. In addition, I think you don't properly define what you mean by "buffering". That term could be used to describe many mechanisms.

We have added to Methodology and Observations 2.3 Regimes:

> *"Buffering can entail the cloud being too thick and impervious to changes due to aerosol due to its high LWP, offsetting and opposite reactions of the cloud resulting in reduced mean signal, or the environment acting to damp the cloud reaction, such as an unstable boundary layer reducing the impact of aerosol on cloud lifetime (Fan, 2016; Stevens & Feingold 2007)."*

We have also removed some references of buffering to simplify some explanations.
We have added to Results and Discussion 3.2

> *"Modulation may by the environment can include the amplification of the reaction through a stable environment further prolonging the cloud lifetime and therefore extent."*

In general, there are so many different environmental or liquid water path dependent processes that could affect aerosol-cloud interactions that to go through those all would be a review paper in itself.

Specific Comments
L3: CA is not defined hear. Consider writing in full cloud adjustments.
Added cloud adjustments before CA is used.
L10: if RFaci and CA counteract and the total effect is small I would say that it could be attributed to damped susceptibility (or buffering). Why is it "erroneously"?
We have removed buffering so as not to confused the reader in this aspect. The total susceptibility may be small, however that does not mean the individual components are each small or cause cooling. A point we aim to make is the idea that the ERFaci should be considered by its components in order to better understand all processes occurring.
L21: what do you mean by "cloud forcing"? is it the cloud radiative effect? I think it is better not to use forcing here and stick with the common definition of radiative forcing.
We have replaced cloud forcing with cloud radiative effect.
L68-72: consider adding here that the sign of the effect was also shown to be a function of the background aerosol concentration.
We have added "and the background state of the aerosol."
L82: the non-monotonic response was shown for other cloud properties (such as cloud fraction and top height) as well as for precipitation. Hence, I don't understand why is it important to separate specifically this effect from the rest.
We have added "in order to reduce the effects of this non-linear relationship on our results."
L105, L109, L141 and other places: again, maybe better to use radiative effect here instead of forcing.
We have replaced some instances of forcing with flux.
L119: SPRINTARS was run (in the paper you are citing) in a T21 resolution (~5.6o) and hence is not "cloud resolving" at all.
We have removed cloud resolving.
L250-258: I couldn't really understand how the uncertainty was calculated. I think more details are needed for it to be reproducible.

We have added: The regressions within all regime constraints, from only meteorological to regional, remain robust for all susceptibilities when 10% of the AI estimates were randomly assigned.

What is the magnitude of error added to PI and PD AI estimations? How did you choose this magnitude?

The PI aerosol error magnitude is calculated from the SPRINTARS data.

L265: you are comparing here the estimated forcing for only warm cloud over the ocean with the total estimation from the IPCC report. I don't think this competition is valid. In addition, in the introduction you cited a few papers showing that most of the ERFaci is coming from warm clouds over the ocean. How that can go together with the relatively low estimations you are getting for warm cloud compared to the total forcing?

Our estimates remain at the low end of observation-based estimates of ERFaci for warm clouds. IPCC estimates are primarily based on global climate models, which difference industrial vs. non-industrial runs.

It is possible that even these estimates of forcing are slightly different than the definition of forcing from the IPCC or model based studies which difference top-of-atmosphere forcings in polluted vs. non-polluted GCM runs.

In the methods section. Our methodology agrees with how others have calculated ERFaci from observations, however.

L310: you cite here a paper focusing on deep convective clouds. Consider adding papers discussing warm cloud invigoration.

We have added a citation to a warm cloud invigoration paper by Ilan Koren.

L 312: I don't understand the claim here. Why determining the casualty of aerosol effect on LWP is more difficult than for CF?

Research has yet to agree if there is some effect of aerosol on LWP (Toll et al. 2019), a large effect (Rosenfeld et al. 2019), or non-linear effects. As deriving a signal in liquid water susceptibility has proven difficult, we chose to focus only on cloud extent, where research has converged to more of an agreement.

L320: why is that a sign of "buffering"? it just means that the aerosol effect is nonmonotonic and change sign. The aerosol level at which the sign flip is a function of the environmental condition as was shown before.

We have replaced "buffering effect" with "the influence of the environment"

L417: the possible change in precipitation could also be relevant between PI and PD making point 1 (general comments above) even more critical.

We agree that precipitation, its effects on aerosol and the environment, and how this alters aerosol-cloud interactions is important. Since we addressed your first general comment, we believe this will now tie in well with our final statement in the precipitation section.

L426-430: you don't mention here, at the beginning of the conclusion section, that these estimations are only relevant for warm cloud over the oceans. It could look like you are giving general estimations here.

We have added "warm, marine cloud" before ERFaci in this first sentence. This, along with adding the warm subscript, should remind the reader these results are limited to only warm clouds.

Technical comments L102 and L107: ECWMF -> ECMWF? Anyway, should be written in full (and maybe also add a citation). *Fixed.*
L401: “on the both the”
Fixed to “on both the.”

Reviewer 1 Response

The authors utilize remote sensing observations and a regime-based approach to isolate the effects of varying aerosol index on cloud microphysical (1st indirect effect) and cloud macrophysical properties (adjustments). The authors utilize regimes of above cloud RH and stability. LWP is binned to account for variations in cloud state in each regime. The results show that in some regions adjustments and the first indirect effect have opposing signs. The authors also show that as LWP increases the radiative response to AI saturates. The analysis presented here satisfies the important problem of separating variability due to meteorology from aerosol-cloud interactions (aci). The authors find a relatively weak ERFaci from warm-topped clouds over oceans, which appears to be due to dimming in regions in the equatorial Atlantic and Indian ocean.

We would like to thank the reviewer for taking the time to read our manuscript and provide feedback and comments.

While I appreciate that the authors are applying the methodology developed in a previous study, it is hard to understand what is being done and I think the authors could briefly summarize their methodology to allow readers to more efficiently refer to DL19. The description of the observational data sets could be much more substantial. It is confusing what observational and modeling data is being used for what. In some cases it appears that observational data sets that are not appropriate are being used, but it is hard to confirm this from the data section. One solution that might make this un-ambiguous would be to create a table of variables and data sources.

We are only using satellite observations and reanalysis data intended to be paired with satellite observations (MERRA-2). To clarify what observations we are using, we have added to section 2.1 Data:

> *"Collocated satellite observations of cloud shortwave forcing, cloud fraction, and aerosol index are obtained by NASA A-Train satellites Aqua, CloudSat, and The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) from 2007 to 2010. The NASA A-Train is configured to maximize the synergy between different satellite products to improve our understanding of clouds, aerosols, and the environment (L'Ecuyer et al. 2011)."*
> *"2B-CLDCLASS-LIDAR combines CloudSat's CPR with CALIPSO lidar observations in order to discern even the thinnest clouds."*
> *"To broadly characterize large-scale environmental conditions, MERRA-2 temperature and humidity profiles are collocated by taking the environmental profile within 30 minutes of a CloudSat overpass and within ~1/2 degree latitude and longitude"*

A critical issue with this paper is use of area-mean LWP (in-cloud LWP*CF) from microwave when the authors imply they are using in-cloud LWP based on wording in the paper (ln 153). From reading the discussion in DL19 I believe that scene-mean LWP from AMSR is just used to filter data into rough bins, and does not play a role in the analysis beyond this. While this is probably not a big problem, the authors may want to clarify what the footprints of the different data sets are that they are using, possibly with a diagram overlaid over an actual satellite image to allow readers who are less familiar with remote sensing to contextualize what is being shown, especially because the authors are using active instruments averaged along track with passive instruments. In particular, in this regard I am confused how the authors are overlapping along-track averaged CF from Cloudsat-CALIPSO with AMSR LWP and a diagram might be helpful. A nice image of the actual cloud field from MODIS on the background would be helpful to readers trying to contextualize the retrievals in terms of cloud features.

We have added the caveats of the footprint discrepancies along with how close geometrically the footprints are. Added to section 2.1 Data:

> *"While the footprints of CloudSat and AMSR-E do not perfectly overlap, the AMSR-E LWP is used to establish a scene based constraint on the clouds in order to better consolidate our observations into regimes. AMSR-E's footprint is within ~2.5 km of CloudSat's track, meaning both sensors are observing the same, liquid clouds (Lebsock et al. 2014)."*

CloudSat observations are often combined with AMSR-E scene averaged LWP in a number of cloud and aerosol studies (such as L'Ecuyer et al. 2009 and Chen et al. 2014). Our study does not aim to understand how the LWP responds to aerosol, only to use LWP as a higher level constraint in order to partition warm clouds into characteristic regimes.

The authors need to either apply their analysis in a GCM simulating PI and PD (Gryspeerdt et al., 2017; Gryspeerdt et al., 2016; McCoy et al., 2019; Costa-Surós et al., 2019) to make sure that their analysis methodology has predictive power, or examine the response of cloud to some sort of transient change in aerosol (Malavelle et al., 2017; Toll et al., 2019) and make sure that their analysis trained over different data can predict the response to the transient change in AI. Without these falsification tests of their predictions, it is unclear what predictive use their correlation model has in that there is no way to falsify their predictions. Even an approximate calculation using model LWP, CF, SW, and AI without any complex output along the satellite overpass (which doesn't appear to be a major source of error compared to problems from low aerosol amount as shown in Ma et al. (2018)) would provide a much more powerful validation of what the authors are hypothesizing is the ERFaci.

A next step will be to find these same signals within output from a GCM, however that is beyond the scope of the current study. This study intends to only document how the observed brightness and extent of clouds respond as aerosol concentration increases, and how these signals depend on the environment and cloud state. These responses *are then used to derive an estimate of ERFaci that is consistent with the specific observations used. While similar methods can be applied to GCM output, the results do not provide a stringent test on the methodology since model responses will depend strongly on how the underlying processes are represented in the model. Non-linearities or stronger/weaker dependencies on environmental state may yield vastly different results that do not provide a useful assessment of the validity of the decomposition approach. Furthermore, any meaningful comparison of GCM output against observations is severely limited by mismatches in resolution between the large GCM gridbox and the fine-scale satellite observations (e.g. Kay et al, 2019).*

In principle, results from this study can be used to assess how well GCMs recreate the derived linearized relationships between aerosol, cloud brightness, and cloud extent under different environmental regimes but such an evaluation requires considerable additional effort and requires close cooperation with modeling groups to ensure appropriate interpretation of the results. It is acknowledged in the manuscript that our study merely aims to document the observed relationships in present climate, not to predict how these may have changed since pre-industrial conditions. Our study provides a benchmark of regimes to be used to evaluate how well updated parameterizations capture current signals.

Within section 2.4 Decomposing the ERFaci we point out that we do not use the lowest 12% of aerosol indices in order to reduce biases in regimes where the correlation between our aerosol proxy and CCN is expected to be weak.

The authors ultimately present a correlative study to predict ERFaci (or at least ERFaci for warm-topped clouds over oceans- see comments below). Characterizing covariance is important but does not guarantee an accurate prediction. In the case of aerosol-cloud adjustments in particular, there is not a unique causality flowing from aerosol to cloud (Wood et al., 2012; Gryspeerdt et al., 2019). In this context, and because their ERFaci is rather weak compared to other studies it seems possible that their analysis conflates aci with precipitation scavenging and other confounders (Gryspeerdt et al., 2019), which would tend reduce correlation strength between aerosol and cloud amount (eg precipitation scavenging is strongest when there is a lot of cloud and there tend to be less cloud and more aerosol off the coast of continents).

It should first be noted that our estimate of the warm cloud ERFaci is within the limits of uncertainty (±0.16 Wm^{-2}) of other observation based estimates such as Christensen et al. (-0.36 Wm^{-2}). To address potential biases due to scavenging effects, we explicitly control for precipitation using CloudSat observations that represent the most sensitive satellite-based metric for precipitation occurrence (Haynes et al, 2009). Separating precipitating from non-precipitating clouds in order to understand how precipitation scavenging and other processes that differ between the two alter their ERFaci reduces our decomposed estimate from -0.21 to -0.207 Wm^{-2}. If our estimates were highly affected by precipitation scavenging of aerosol, we would expect the difference between these estimates to be greater.

We acknowledge that our regimes do not capture all signals of covariability between the environment and aerosol and have added to section 2.2 Regimes:
"Using EIS and RH_{700} does not guarantee to limit all covariability between the environment, aerosols, clouds, and their interactions. Some covariability may still exist, such as surface winds affecting both clouds and aerosol (Nishant et al. 2017)."

The authors need to refer to their ERFaci as ERFaci_liquid-topped_over_oceans (or at least that is my take from the methodology and Eq 9).

We have changed all mentions of ERFaci to $ERFaci_{warm}$, RFaci has become $RFaci_{warm}$, and CA has become CA_{warm} in order to remind the reader these results only apply for warm-topped clouds. We have added mentions of our observations being limited to only marine warm clouds throughout section 2.1 Data.

Specific changes:
Pg 1 ln 3: ERFaci is a combination of microphysical (RFaci) and macrophysical changes (adjustments) and the latter could be further split into changes in extent and thickness (Gryspeerdt et al., 2019; Gryspeerdt et al., 2017; Gryspeerdt et al., 2016). As written this implies that thickness stays constant and the only possible adjustment is CF. I understand now that this is more like the intrinsic extrinsic separation in other studies (Christensen et al., 2017), but this would be better to clarify in the abstract.

We have added to the abstract intrinsic and extrinsic to make the connection to the study by Chen et al. 2014 and Christensen et al. 2017 adding next to the RFaci term intrinsic and the cloud adjustment term extrinsic.

Pg. 2 ln 40: The goals of DL19 overlap a lot with the goals of the present study. A sentence like 'The present study expand on DL19 in the following ways:' would be helpful. I believe the primary difference between these studies is the inclusion of adjustments, but it would be helpful to state that explicitly for readers to rapidly ingest what is happening.
We have added to section 1 Introduction:

> *"The present study expands upon work done in DL19 by specifying what aspects of the cloud lead to changes in the CRE, whether that be the brightness or cloud extent or both, and whether these changes can negate each other, such as when a cloud shrinks but the brightness increases."*

Pg. 3 ln 85: It would help readers to quickly process what data sets are being used to describe what variable to use subheaders here (2.1 Data, 2.1.1 Warm cloud fraction). This is stylistic, but I found it hard to understand where precipitation measurements were coming from. I think that it would help a lot to have a table of what the precise data sets used are, especially since some of the remote sensing data sets being used may be inappropriate, but it is unclear if they are actually used (eg AMSR rain rates, although I believe these are not used despite being mentioned).

We have added an additional paragraph in section 2.1 Data to clarify how we separated precipitating and non-precipitating clouds exactly.

> *"Clouds are separated into precipitating and non-precipitating regimes using CloudSat's 2C-PRECIP-COLUMN precipitation flag. Clouds with a 0 precipitation flag, no precipitation detected, are designated as non-precipitating. Precipitating clouds are separated using flag 3, where rain is certain (Haynes et al. 2009). Our precipitating clouds include a majority of the drizzling cases, as CloudSat's 2C-PRECIP-COLUMN's threshold for drizzle is -15 dB, which should capture all but the lightest drizzling clouds (Stephens et al. 2007)."*

Pg4 ln 124: is the material not shown in the citation? If it's in the citation no need to put not shown here.

The material is shown within the citation, we meant to say that we do not show these results within the current paper. We have removed not shown.

Pg 4 ln 125: Swelling is a key issue in trying to understand adjustments. I believe that swelling is not an issue for SPRINTARS because the model can be internally consistent, but an additional comment is needed about MACC aerosol swelling. It's unclear that MACC can fully correct for swelling given the very complex way that swelling occurs (Christensen et al., 2017; Twohy et al., 2009). This needs to be explained and caveated. Also, why mix MACC aerosol and MERRA-2 meteorology? MERRA2 produces a very similar aerosol reanalysis to MACC and this would avoid confusing MERRA2 meteorology with aerosol reanalysis in a different framework. Also- how are SPRINTARS and MACC not sensitive to precipitation scavenging? Presumably both data sets have a precipitation sink of aerosol otherwise it would be very hard to maintain realistic aerosol.

Our results shown do not include any MACC aerosol products. We removed the reference to MACC aerosol in order to not confuse the reader. We have done the same regime analysis with MACC and SPRINTARS AOD for the same time period in order to validate the sign of the regime signals derived here. We have removed the precipitation scavenging mention since SPRINTARS does include some type of precipitation sink for aerosol.

We have added to section 2.4 Decomposing the ERFaci:
"Aerosols swell in the vicinity of clouds, which increases their size and therefore affects the MODIS retrieval AI (Christensen et al. 2017). To assess how significantly this may affect results we have randomly added errors of 10% to our AI estimates and re-derived all signals with all regime constraints. Even with extreme amounts of error in AI, the signals within our environmental and LWP regimes are robust.

Pg.5 Ln140: This methods section is really short. I understand that the authors refer to DL19, but I think it would help readers evaluate this paper more quickly if a paragraph or so was taken to summarize DL19.

We have added to the methods:

> *"In DL19, environmental and cloud state regimes were imposed on a regional basis in order to identify regime specific behavior of aerosol-cloud-radiation interactions. Within each regime, we regressed the cloud radiative effect (CRE) against AI in order to find the susceptibility of warm cloud radiative properties to aerosol. We use these same susceptibilities within section 3.1 to quantify the total warm, marine ERFaci. DL19 found that the susceptibility varies regionally and by regime, however the $ERFaci_{warm}$ depends on the magnitude to which aerosol has increased since pre-industrial times. Further, the $ERFaci_{warm}$ does not diagnose what characteristics of the cloud are causing the effect, prompting us within this paper to decompose the $ERFaci_{warm}$ into the effects on the albedo and the effects on cloud extent."*

Pg. 6 Eq3-6: how do the authors account for CF being bounded between 0-1 in this calculation?

Our cloud fraction is the fraction of a 12 km x 1 km along track region covered in clouds according to CloudSat's 2B-CLDCLASS-LIDAR, which includes even the thinnest clouds not captured by CPR. Therefore, our cloud fractions should be between 0 and 1.

Pg. 8 ln 221: The authors assert that by binning LWP they reduce the chances of buffering. One thing that should be mentioned in this study is that AI and LWP will naturally anti-correlate due to precipitation and scavenging correlating with cloudiness (eg LWP or CF) (Wood et al., 2012) and due to air mass history leading to both drier and more aerosol-laden air (Gryspeerdt et al., 2019). These non-causal relationships are not meaningful to ERFaci, but can substantially affect the covariability of cloud macrophysical properties and aerosol, and thus the inferred aci strength (McCoy et al., 2019). It is possible that the LWP binning and precipitation stratification reduce this effect. However, the authors must show some demonstration of the predictive ability of this method by either (1) applying it to GCM data (in this case SPRINTARS) and showing that their methodology when applied in a GCM can accurately reproduce the GCM response to enhance aerosol as in Gryspeerdt et al. (2016) or McCoy et al. (2019) – or – (2) examining one of the transient aerosol emissions identified in recent studies (Malavelle et al., 2017; Toll et al., 2019) and see if their characterization of sensitivity of cloud to aerosol has some predictive ability. Without this sort of test there is no guarantee that the inferred ERFaci_warm-topped_oceanic is accurate.

We have added to section 3.2 Impact of LWP within the results:

> *"While regime constraints on LWP do reduce the covariability between aerosol-cloud interactions and the role LWP plays in buffering these interactions, it does not remove all sources of covariability between LWP, aerosol, the*

environment, and cloud properties. Aerosol has been shown to negatively correlate with LWP (Gryspeerdt et al. 2019). It is possible that this relationship, and the inherent relationship between the environment and LWP, could affect results shown."

Future work is planned to evaluate how regime-specific relationships compare to those derived via application of similar methods to GCMs, however, as noted above, uncertainty in the parameterization of aerosol-cloud interactions and their regime-dependence preclude drawing concrete conclusions regarding the validity of the methodology from such analyses. More importantly, the resolution of today's GCMs is not sufficient to accurately emulate the distributions of clouds and aerosols on the same scales as the observations so considerable thought and effort will be needed to ensure that the methods can be applied within a model framework in a meaningful way. We agree that the observations have caveats, which we have acknowledged within our manuscript, but we have thoroughly documented our methods, the underlying datasets used, and the analysis approach. As with any study, these choices can be debated and improved upon but the results presented here are (a) an accurate representation of the correlations that exist in the datasets employed; (b) reproduceable; and (c) accompanied by an appropriately large error bar. We believe these data and the analysis method described here represent the current state of the art given current Earth observing capabilities but acknowledge that these estimates will likely be refined in the future.

Pg 10 ln 300: An alternative explanation of the weakening precipitation effect in clouds with higher LWP may be that precipitation increases with LWP, which means that precipitation scavenging becomes larger, which in turn means that the true adjustment strength is obscured by non-causal covariance between aerosol and cloud macro physics (see discussion in McCoy et al. (2019)).

We have added to section 3.2 Impact of LWP:

> *"An alternative explanation is that thicker clouds with larger LWPs are more likely to precipitate, scavenging aerosol and weakening the susceptibility. Aerosol-cloud-precipitation interactions complicate cloud adjustment processes in higher LWP clouds; the true susceptibility may be masked by covariance between aerosol and precipitation in these clouds (McCoy et al. 2019)."*

Figure 7 and ln 456: The authors find a large ERFaci in the SH, which is really surprising given the very small change in anthropogenic aerosol in these regions. Figure 1 shows change in AI, but it is a bit hard to distinguish small changes from zero and the authors may want to consider some sort of log normalization to their color scale. However, strong ERFaci exists along a line around 40◦S, which is hard to square with studies examining pristine days in the PD (Hamilton et al., 2014). That is to say, the pattern of ERFaci in this study is dramatically different than the RFari shown in, for example, aerocom (Myhre et al., 2013).

Since our estimates of the ERFaci are weighted by occurrence, regions with the highest occurrence of warm clouds will have larger ERFaci. The southern hemisphere is known to have the largest occurrence of warm cloud decks, therefore our weighted ERFaci from observations will weight the southern hemisphere over the northern hemisphere. Further, the southern ocean may have a higher susceptibility due to their usual pristine conditions making them primed and highly sensitive to any changes in aerosol.

Figure 7: While I think it's good to pursue analysis to its conclusion by applying it to all data, I am surprised at the positive RFaci and CA in the tropics. Can the authors comment on whether their analysis is sensitive to retrieval errors in convective cloud? In particular, a positive forcing due to RFaci is quite unusual- while it may be due to biomass burning aerosol above cloud in some regions via semi-direct effects or blocking reflective light (so not really aci) (Bellouin et al., 2019), the appearance of a positive RFaci seems to be more related to SST, than aerosol type given its appearance over the tropics, and far away from strong aerosol sources.

A limitation of our data is that the cloud radiative effect can be reduced due to semi-direct effects not constrained by our environmental or LWP limits.
We have added to address that the semi-direct effect is not accounted for by our methodology and may result in a reduced RFaci, in Results and Discussion section 3.3. Constrained by local meteorology

> *"It is also possible lambda_RFaci is impacted by some semi-direct effects by smoke aerosol which would lead to a cloud dimming and positive susceptibility. Semi-direct effects are not accounted for by our methodology, however aerosol within the cloud layer could lead to cloud breakup processes, a dimmer albedo, and changes to the local environment by the absorbing aerosol."*

Quantifying Cloud Adjustments and the Radiative Forcing due to Aerosol-Cloud Interactions in Satellite Observations of Warm Marine Clouds

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Abstract. Aerosol-cloud interactions and their resultant forcing remains one of the largest sources of uncertainty of future climate scenarios. The effective radiative forcing due to aerosol-cloud interactions (ERFaci) is a combination of two different effects, how aerosols modify cloud brightness (RFaci, intrinsic) and how cloud extent reacts to aerosol (cloud adjustments CA, extrinsic). Using satellite observations of warm clouds from the NASA A-Train constellation from 2007 to 2010 along with MERRA-2 reanalysis and aerosol from the SPRINTARS model, we evaluate the ERFaci [..[1]]in warm, marine clouds and its components, the RFaci$_{warm}$ and CA$_{warm}$, while accounting for the liquid water path and local environment. We estimate the ERFaci$_{warm}$ to be -0.32 $\pm$0.16 Wm^{-2}. The RFaci$_{warm}$ dominates the ERFaci$_{warm}$ contributing 80% (-0.21 $\pm$0.15 Wm^{-2}), while the CA$_{warm}$ enhances this cooling by 20% (-0.05 $\pm$0.03 Wm^{-2}). Both the RFaci$_{warm}$ and CA$_{warm}$ vary in magnitude and sign regionally, and can lead to opposite, negating effects under certain environmental conditions. Without considering the two terms separately, and without constraining cloud-environment interactions, weak regional ERFaci$_{warm}$ signals may be erroneously attributed to a damped susceptibility to aerosol.

1 Introduction

Aerosol-cloud interactions (ACI) and their impact on cloud radiative effects are a vital component of Earth's radiative balance. Warm clouds, in particular, are susceptible to aerosols, and due to their prevalence and role as "Earth's sunblock", these interactions are critical for regulating Earth's surface temperature (Platnick and Twomey, 1994). Aerosols entering a cloud may become cloud condensation nuclei (CCN) initiating a domino effect wherein the cloud's droplet number increases, reducing the mean droplet radius, brightening the cloud's albedo, dampening its ability to precipitate, and, in theory, increasing its lifetime and radiative effect (Twomey, 1977; Albrecht, 1989). However, it remains unknown to what degree aerosols alter warm cloud radiative forcing as models and observations disagree. Global climate models are prone to uncertainty due to their dependence on parameterizations and inability to explicitly represent all scales of ACI, while satellite observations have poor temporal resolution, and natural covariances with the environment may influence warm cloud response to aerosol (Stevens and Feingold, 2009). In order to understand aerosol-cloud interactions and the resulting change in cloud radiative effect, observation-based

[1]removed: $_{warm}$

methods must address the inherent limitations of satellite observations by creating a framework to resolve the interplay between clouds, the environment, and aerosol-cloud interactions (Seinfeld et al., 2016).

Correctly quantifying the effective radiative forcing due to aerosol-cloud interactions (ERFaci) of warm clouds specifically is important to establish a climate sensitivity and identify cloud feedbacks (Bony and Dufresne, 2005; Rosenfeld, 2006; Boucher et al., 2013). It has been understood since the early 1990s that low, warm clouds play a leading role in determining future warming scenarios (Slingo, 1990). The micro- and macrophysical responses of warm clouds to ACI lead to numerous, poorly understood cloud feedbacks in the Earth system (Gettelman and Sherwood, 2016). Clouds do not exist in isolation (Stephens, 2005). Clouds are part of an interconnected system; changes to one aspect, such as particle size or liquid water content, has a ripple effect to other components of the Earth system. Likewise, clouds can be thought of residing in a "buffered system" where a clouds response to aerosol perturbations can be invigorated or diminished depending on the conditions in which it is initiated (Stevens and Feingold, 2009). These interconnections lead to a range of cloud responses to aerosol that depend on the local meteorology and cloud state (Douglas and L'Ecuyer, 2019). Both the short and long time scales of ACI and their radiative forcing are affected by the interconnections they exist in, meaning constraining the ERFaci of warm clouds must go beyond a single measure of the ERFaci globally and distinguish the individual components of the ERFaci, the radiative forcing due to aerosol-cloud interactions (RFaci) and cloud adjustments (CA). To account for the challenges in estimating the cloud radiative response to aerosol, we constrain the influences of the local meteorology and cloud state using a method developed in Douglas and L'Ecuyer 2019, hereafter DL19. The ERFaci$_{warm}$ is separated into the RFaci$_{warm}$ and cloud adjustments determined with constraints on meteorology following DL19 and estimates of each effect are presented to find the relative contributions of the RFaci$_{warm}$ and cloud adjustments to the ERFaci$_{warm}$. The present study expands upon work done in DL19 by specifying what aspects of the cloud lead to changes in the CRE, whether that be the brightness or cloud extent or both, and whether these changes can negate each other, such as when a cloud shrinks but the brightness increases.

Warm clouds, like marine stratocumulus and trade cumulus, are the prevailing cloud type over the oceans and dominate aerosol-cloud interactions (Gryspeerdt and Stier, 2012). Marine stratocumulus over the cold upwelling waters, such as off the west coast of Africa, persist for long periods of time in the stable, low marine boundary layers (Wood, 2012). Cumulus form from marine stratocumulus to cumulus transitions and in the equatorial region as trade cumuli (Sandu and Stevens, 2011). Warm clouds sheer abundance and bright albedo make them important to the radiative balance of Earth, and it should be no surprise that warm clouds contribute the largest amount of forcing to the ERFaci (Christensen et al., 2016). Marine stratocumulus have been the primary focus of aerosol-cloud-radiation interactions due to their sheet-like, "homogeneous" structure, pervasiveness ($\sim$25% of the Earth at any moment), location near anthropogenic continental emissions, and susceptibility to changes in their CCN (Hahn and Warren, 2007; Platnick and Twomey, 1994).

The warm cloud albedo has the largest response to aerosol compared to mixed phase or ice phase clouds (Christensen et al., 2016). Twomey was the first to hypothesize the high susceptibility of entirely liquid clouds to aerosol using a simple cloud model; work since then has confirmed this as the basis of RFaci (Twomey, 1977). Observation- and model-based studies focus on the albedo effect because it is a macrophysical manifestation of microphysical processes. An increase in CCN and decrease in mean droplet radius greatly increases the cloud albedo, and, as such, has significant implications for the radiative balance.

The radiative forcing of the albedo effect, or the sudden microphysical response to aerosol loading (RFaci), is dependent on the activation and eventual microphysical initiation of aerosol as cloud droplets, which can be influenced by local dynamics, the stability of the boundary layer, and the initial cloud state (Su et al., 2010). "Model" conditions simulated by Twomey only exist in the most pristine, stable southern oceans (Gryspeerdt et al., 2017; Hamilton et al., 2014). Depending on the region studied, aerosol can increase the cloud albedo as expected, or in certain cases, lead to a dimming effect, such as when aerosol loading reaches a critical point or the local meteorology regulates the sign and/or magnitude of ACI (Gryspeerdt et al., 2019b; Christensen et al., 2014). Studies conflict to what degree the RFaci dominates the ERFaci, in part because the cloud acts as a “buffered system” and mitigates the RFaci depending on the thermodynamic conditions, making the quantification of the RFaci particularly challenging (Goren and Rosenfeld, 2014; Feingold et al., 2016; Stevens and Feingold, 2009).

Efforts to understand the other component of the ERFaci, cloud adjustments, have been similarly clouded in uncertainty. Cloud lifetime and extent are highly susceptible to aerosol (Dagan et al., 2018). Models have shown that aerosol affects the distribution of liquid throughout the cloud and vertical motion within the cloud, greatly perturbing the cloud’s lifetime, precipitation, and extent (Ramanathan et al., 2001; Dagan et al., 2016). Aerosol can act to increase the lifetime of clouds through delayed collision coalescence, or decrease the lifetime through evaporation-entrainment and induced cloud feedbacks (Albrecht, 1989; Small et al., 2009). A satellite observation-based study of ship tracks showed clouds experience a expansion or shrinking of cloud extent depending on whether the clouds are at an open or closed state and the background state of the aerosol (Chen et al., 2015). The cloud adjustment response depends on the cloud state and a sequence of reactions dictated by the environment (Gryspeerdt et al., 2019b). As such, cloud adjustments remain the largest source of variability of ERFaci in global climate models (Fiedler et al., 2019).

To account for influences and variation in the ERFaci$_{warm}$, RFaci, and cloud adjustments, we constrain the liquid water path, relative humidity of the free atmosphere, and stability of the boundary layer and covariances between them before evaluating the susceptibility of the effect in the same fashion as DL19. These constraints are held fixed first on a global and then on a regional basis to diagnose regime specific then regionally specific responses. Finally, the decomposed ERFaci, or the sum of the RFaci and cloud adjustments, is found, with constraints on the environment and cloud state, for precipitating and non-precipitating scenes on a regional basis. Our methodology aims to reduce biases by accounting for the regionally specific aerosol and thermodynamic conditions (Feingold, 2003). The relationship between aerosol and cloud response has been proven to be sensitive to regional features like aerosol type or meteorology (Twohy et al., 2005; Chen et al., 2014)(DL19). Aerosol-cloud interactions experience a non-linear relationship with liquid water path therefore it is important to separate this complex relationship from ACI and the associated forcing in order to reduce the effects of this non-linear relationship on our results (Gryspeerdt et al., 2019b).

2 Methodology and Observations

2.1 Data

Collocated satellite observations of cloud shortwave forcing, cloud fraction, and aerosol index are obtained by NASA A-Train satellites Aqua, CloudSat, and The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) from 2007 to 2010. The NASA A-Train is configured to maximize the synergy between different satellite products to improve our understanding of clouds, aerosols, and the environment (L'Ecuyer and Jiang, 2011). Observations of marine warm clouds and aerosols from the Cloud Profiling Radar (CPR) and Moderate Resolution Imaging Spectroradiometer (MODIS) aboard CloudSat and Aqua, respectively, are utilized to evaluate the effects of aerosol-cloud interactions on the radiative properties of clouds including their albedo and extent.

CloudSat was launched to an orbit collocated with Aqua and other A-Train satellites in 2006. The CPR on CloudSat is a 94 GHz radar with a $\sim$ 1.7 km along track, 1.4 km cross track resolution, and 480 m vertical resolution (Stephens et al., 2018; Tanelli et al., 2008). A number of cloud properties can be inferred using the CPR backscatter including cloud top height, cloud type, and accompanying radiative effects.

An along track warm cloud fraction is defined using cloud top height from 2B-CLDCLASS-LIDAR and freezing level from 2C-PRECIP-COLUMN. 2B-CLDCLASS-LIDAR combines CloudSat's CPR with CALIPSO lidar observations in order to discern even the thinnest clouds. At each pixel, the cloud fraction is defined by the amount of cloud uptrack and downtrack of that pixel at a 12 km scale, chosen to approximate the scale of marine boundary layer processes and accentuate small scale changes in extent compared to other large sizes (e.g. 1° x 1°). Using a smaller scale such as 12 kms for cloud fraction will allow even minute changes in the cloud extent to be detected by our methodology; using a larger size such as 96 km ($\sim$1°) may diminish cloud breakup processes within large stratocumulus decks or minimize effects on trade cumuli. 2B-CLDCLASS-LIDAR includes collocated Cloud-Aerosol Lidar with Orthogonal Polarization (CALIPSO) satellite lidar backscatter measurements to identify thin, shallow clouds that may escape detection by the CPR (Sassen et al., 2008). Cloud top heights from 2B-CLDCLASS-LIDAR, defined using a combination of collocated lidar and CPR measurements, are required to be below the freezing level (Haynes et al., 2009). The freezing level of 2C-PRECIP-COLUMN is obtained from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses and is used to separate warm from mixed and ice phase clouds. Focusing only on warm phase clouds helps reduce the uncertainty associated with retrievals of mixed and ice phase clouds.

Cloud fraction is combined with shortwave top of atmosphere forcings from the CloudSat 2B-FLXHR-LIDAR product to approximate the effect of aerosol on albedo. 2B-FLXHR-LIDAR uses a combination of CPR and CALIPSO measurements along with MODIS cloud properties and atmospheric conditions from ECMWF as input to a radiative transfer model that computes top of atmosphere shortwave fluxes that have been shown to agree well with CERES observations (Henderson et al., 2013). The mean shortwave flux at the top of atmosphere is weighed by a mean incoming solar radiation at the top of atmosphere in our analysis to account for diurnal variation of incoming solar radiation not sampled by the sun-synchronous A-Train orbit.

We use aerosol index (AI) as a proxy for aerosol concentration from MODIS. The AI is the product of the Angstrom exponent, calculated using aerosol optical depth (AOD) at 550 and 870 nm, and the AOD at 550 nm. AI has been shown to have a higher correlation with CCN compared to AOD (Stier, 2016; Hasekamp et al., 2019). Cloudy scene AI is determined by interpolating between clear scenes along track. This interpolation may reduce the accuracy in completely overcast scenes, however for most scenes where cloud fraction is < 1, this interpolation should be sufficiently accurate. Aerosol swelling in high humidity environments also leads to some uncertainty in AI but but should be limited to select high humidity environmental regimes. Pre-industrial aerosol information is provided by Spectral Radiation-Transport Model for Aerosol Species (SPRINT-ARS), an atmosphere-ocean general circulation model (Takemura et al., 2000). Pre-industrial aerosol errors lead to the majority of uncertainty in ACI due to uncertainties in transport, source, and concentration of pre-industrial aerosol conditions (Chen and Penner, 2005).

The sign and regional variations in susceptibilities found using MODIS AI shown within this study were evaluated against susceptibilities found using MACC and SPRINTARS aerosol in order to qualitatively scrutinize any error due to aerosol retrieval [..[2]](Douglas, 2017). MACC and SPRINTARS provide independent aerosol estimates not susceptible to swelling, instrument sensitivity [..[3]]or retrieval error.. The fact that our results were qualitatively similar using modeled aerosol provides confidence that the derived susceptibilities shown are not simply an artifact of using satellite-derived AI.

The analysis is constrained to clouds with LWPs between 0.02 to 0.4 kgm^{-2} using the Advanced Microwave Scanning Radiometer for Earth Observing Satellite (AMSR-E), an instrument aboard Aqua that infers water vapor and precipitation amounts using six microwave frequencies over a [..[4]]$\sim$14 km area (comparable to the averaging scale of our cloud fraction) (Parkinson, 2003; Wentz and Meissner, 2007). While the footprints of CloudSat and AMSR-E do not perfectly overlap, the AMSR-E LWP is used to establish a scene based constraint on the clouds in order to better consolidate our observations into regimes. AMSR-E's footprint is within $\sim$2.5 km of CloudSat's track, meaning both sensors are observing the same, liquid clouds (Lebsock and Su, 2014). Imposing these LWP limits in place removes only $\sim$1% of observations leaving over 1.8 million satellite observations for analyses, but avoids possible skewing by extremely thick, bright clouds or extremely thin, dim clouds.

Environmental information to define local meteorological regimes is provided by the Modern-Era Retrospective analysis for Research and Applications, version 2 (MERRA-2) reanalysis (Gelaro et al., 2017). To broadly characterize large-scale environmental conditions, MERRA-2 temperature and humidity profiles are collocated by taking the environmental profile within 30 minutes of a CloudSat overpass and within $\sim \frac{1}{2}^{\circ}$ latitude and longitude. Vertical profiles of humidity and temperature are used to calculate the estimated inversion strength (EIS) of the boundary layer and the relative humidity at 700 mb (RH_{700}) to represent the humidity of the free atmosphere (Wood and Bretherton, 2006). By simultaneously stratifying the observations by LWP, RH, and EIS, the analysis directly accounts for covariability between LWP and the local environment by separately evaluating the susceptibility of each environmental regime within distinct LWP limits (Douglas and L'Ecuyer, 2019).

Clouds are separated into precipitating and non-precipitating regimes using CloudSat's 2C-PRECIP-COLUMN precipitation flag. Clouds with a 0 precipitation flag, no precipitation detected, are designated as non-precipitating. Precipitating

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clouds are separated using flag 3, where rain is certain (Haynes et al., 2009). Our precipitating clouds include a majority of the drizzling cases, as CloudSat's 2C-PRECIP-COLUMN's threshold for drizzle is -15 dB, which should capture all but the lightest drizzling clouds (Stephens and Wood, 2007).

2.2 [..[5]]Methodology

In DL19, environmental and cloud state regimes were imposed on a regional basis in order to identify regime specific behavior of aerosol-cloud-radiation interactions. Within each regime, we regressed the cloud radiative effect (CRE) against AI in order to find the susceptibility of warm cloud radiative properties to aerosol. We use these same susceptibilities within section 3.1 to quantify the total warm, marine ERFaci. DL19 found that the susceptibility varies regionally and by regime, however the ERFaci$_{warm}$ depends on the magnitude to which aerosol has increased since pre-industrial times. Further, the ERFaci$_{warm}$ does not diagnose what characteristics of the cloud are causing the effect, prompting us within this paper to decompose the ERFaci$_{warm}$ into the effects on the albedo and the effects on cloud extent.

The mean shortwave flux at the top-of-atmosphere from CloudSat's 2B-FLXHR-LIDAR along with our definition of warm cloud fraction from 60° S to 60° N are used to define the RFaci$_{warm}$ and cloud adjustment terms of the ERFaci$_{warm}$. We first calculate the ERFaci$_{warm}$ on a regional basis with regime constraints using estimates of the susceptibility of the warm [..[6]]CRE to aerosol from DL19 and pre-industrial and present-day AI from SPRINTARS. We then use a partial derivative decomposition to separate out the RFaci$_{warm}$ and cloud adjustment terms. These terms are evaluated globally as susceptibilities with constraints on the local meteorology and cloud state following the methodology of DL19. The RFaci$_{warm}$ and cloud adjustments are evaluated regionally with constraints on cloud state and local meteorology. The decomposed ERFaci$_{warm}$ is evaluated for precipitating and non-precipitating scenes to account for the potential effects of precipitation on ACI. Finally, the sum of the RFaci$_{warm}$ and cloud adjustments, the decomposed ERFaci$_{warm}$, is compared against the first estimate of the ERFaci$_{warm}$.

2.3 Regimes

Following DL19, the ERFaci$_{warm}$ and components are evaluated within a constrained space on both a global and regional scale. LWP is held approximately constant using a set of twelve LWP limits on a global basis and five LWP limits on a regional basis. This is in line with the original work of Twomey, who surmised that only for a fixed LWP will the cloud albedo increase in more polluted conditions. The local meteorology is defined by the stability of the boundary layer and the relative humidity of the free atmosphere. Both the stability, characterized by the estimated inversion strength, and the relative humidity of the free atmosphere, defined at the 700 mb level, have been shown to influence the sign and magnitude of the susceptibility of the CRE to aerosol (Wood and Bretherton, 2006; Ackerman et al., 2004; De Roode et al., 2014). The resulting regimes are used to minimize the effects of buffering, or reduced observed response, by the cloud state or surrounding environment to accurately isolate the susceptibility of the cloud to aerosol under controlled conditions. Buffering can entail the cloud being too thick and

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impervious to changes due to aerosol due to its high LWP, offsetting and opposite reactions of the cloud resulting in reduced mean signal, or the environment acting to damp the cloud reaction, such as an unstable boundary layer reducing the impact of aerosol on cloud lifetime (Fan et al., 2016; Stevens, 2007). Using EIS and RH_{700} does not guarantee to limit all covariability between the environment, aerosols, clouds, and their interactions. Some covariability may still exist, such as surface winds [..[7]]that may affect both clouds and aerosol (Nishant and Sherwood, 2017). These constraints only account for the major environmental controls on clouds and aerosol-cloud interactions, some more minor or less common environmental controls may still exert an influence on our results.

While binning our observations by environmental regime should control for some modulation the environment has on aerosol-cloud interactions, it does not fully [..[8]]capture aerosol-environment interactions. For example, in some regions such as off the coast of Africa, biomass burning results in smoke layers that absorb incoming radiation and warm the atmosphere (Cochrane et al., 2019). This [..[9]]could affect the humidity and temperature of the local environment. [..[10]]Environmental regime constraints would capture how the altered environment may regulate aerosol-cloud interactions, but [..[11]]separation into such regimes does not address how the aerosol has impacted the environment.

2.4 Decomposing the ERFaci

A Newtonian-based method is employed to represent the ERFaci_{warm} as a sum of its parts, the RFaci_{warm} and cloud adjustments. A positive ERFaci_{warm}, RFaci_{warm}, or cloud adjustment denotes a damped cooling effect of the cloud while a negative sign denotes an additional cooling due to aerosol-cloud interactions. If the shortwave cloud radiative effect is the product of the cloud fraction (CF) and the cloudy sky shortwave flux at the top-of-atmosphere (SW_{Cloudy}):

$$\mathrm{CRE} = \mathrm{CF} \times \mathrm{SW}_{\mathrm{Cloudy}} \tag{1}$$

then, taking the derivative of the CRE with respect to the log of aerosol index, we find the effective radiative forcing due to aerosol-cloud interactions (ERFaci) or the change in the CRE with respect to aerosol:

$$\mathrm{ERFaci} = \frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \times \Delta \ln(\mathrm{AI}) \tag{2}$$

where $\Delta\ln(\mathrm{AI})$ is the change in ln(AI) from pre-industrial to present-day conditions derived from SPRINTARS. SPRINTARS is a 3-D aerosol model that includes emission, advection, diffusion, chemistry, wet deposition, and gravitational settling of multiple species of aerosol driven by a general circulation model developed by the University of Tokyo (Takemura et al., 2000, 2005).

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All susceptibilities are found using MODIS AI, while only the $\Delta\ln(\text{AI})$ term uses SPRINTARS modeled aerosol. The lowest 12% of aerosol indices are ignored when determining a susceptibility, as these have been shown to have little to no correlation with CCN compared to higher indices (Hasekamp et al., 2019). Error in MODIS AI estimates adds the greatest source of uncertainty in the observationally based portion of this study, however, signals derived are all robust enough to be observed even when random error is added to 10% of the AI estimates. The regressions within all regime constraints, from only meteorological to regional, remain robust for all susceptibilities when 10% of the AI estimates were randomly assigned. The same relationships can be qualitatively observed when SPRINTARS [..[12]]AOD is used in lieu of MODIS AI (Douglas, 2017).

The susceptibility ($\frac{\partial \text{CRE}}{\partial \ln(\text{AI})}$) can be obtained directly from satellite estimates of top-of-atmosphere clear-sky and all-sky fluxes and aerosol index or further decomposed into separate albedo and cloud fraction responses using Equation 1. Applying the chain rule to equation 2, combined with the definition of CRE from Equation 1, gives:

$$\frac{\partial \text{CRE}}{\partial \ln(\text{AI})} = \frac{\partial \text{CF}}{\partial \ln(\text{AI})} \times \overline{\text{SW}}_{\text{Cloudy}} + \overline{\text{CF}} \times \frac{\partial \text{SW}_{\text{Cloudy}}}{\partial \ln(\text{AI})} \tag{3}$$

where the overbars represent means.

The sum of the right hand terms represent the decomposition susceptibility:

$$\text{Decomposition Susceptibility} = \lambda_{Sum} = \frac{\partial \text{CF}}{\partial \ln(\text{AI})} \times \overline{\text{SW}}_{\text{Cloudy}} + \frac{\partial \text{SW}}{\partial \ln(\text{AI})} \times \overline{\text{CF}} \tag{4}$$

The first term of Equation 4 represents the cloud adjustment susceptibility to aerosol, which to first order is the effect of aerosol on the cloud extent:

$$\text{Cloud Adjustment Susceptibility} = \lambda_{CA} = \frac{\partial \text{CF}}{\partial \ln(\text{AI})} \times \overline{\text{SW}}_{\text{Cloudy}} \tag{5}$$

The cloud adjustment forcing is the product of the cloud adjustment susceptibility λ_{CA} and the change in AI from pre-industrial to current times $\Delta\ln(\text{AI})$:

$$\text{Cloud Adjustment Forcing} = \lambda_{CA} \times \Delta\ln(\text{AI}) \tag{6}$$

The cloud adjustment susceptibility (λ_{CA}) is described by its most notable effect, the enhancement and sustainment of clouds as a result of precipitation suppression. We define the cloud adjustments as the product of the change in cloud fraction

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with respect to aerosol index and the mean cloud shortwave forcing. By multiplying by the mean cloud shortwave forcing, a change in cloud extent is converted to a change in the reflected shortwave. While this term does not explicitly account for precipitation, we separate clouds by rain state and determine the difference in the RFaci$_{warm}$ and cloud adjustments between precipitating/non-precipitating clouds; this difference is likely close to the overall effect of precipitation on aerosol-cloud-radiation interactions.

This cloud adjustment term accounts for the main process, the change in extent of clouds by aerosol, however many other studies define the cloud adjustment term by the change in LWP by aerosol. We choose to instead focus on the expansion or shrinking of clouds by aerosol and constrain any LWP effects. Research has yet to establish how and where LWP increases or decreases due to aerosol-cloud interactions; focusing on the changes to cloud extent reduces the error in the adjustment term due to this uncertainty.

The second term on the right hand side of Equation 4 represents susceptibility of warm cloud radiative forcing due to aerosol-cloud interactions (RFaci):

$$\text{RFaci Susceptibility} = \lambda_{RFaci} = \overline{\text{CF}} \times \frac{\partial \text{SW}_{\text{Cloudy}}}{\partial \ln(\text{AI})} \tag{7}$$

where the associated forcing is the product of the RFaci$_{warm}$ susceptibility λ_{RFaci} and the change in AI from pre-industrial to current times $\Delta\ln$(AI):

$$\text{Radiative Forcing due to aci} = \lambda_{RFaci} \times \Delta\ln(\text{AI}) \tag{8}$$

The RFaci$_{warm}$ susceptibility is the change in the shortwave effect owing to changes in cloud droplet radius, an immediate, fast response. The outgoing shortwave radiation for cloudy scenes depends on the cloud albedo; a brighter, whiter cloud will reflect more incoming solar radiation, increasing $\text{SW}_{\text{Cloudy}}$ at the top of the atmosphere. $\text{SW}_{\text{Cloudy}}$ is weighted by the annual solar insolation cycle in order to normalize the term and reduce the impact of changes in the incoming solar flux. RFaci$_{warm}$ is weighted by mean cloud fraction since the net effect of brighter clouds depends on how extensive they are.

Finally, to account for the dependence of each susceptibility (RFaci, CA, and total) on the meteorology and cloud state, each susceptibility (λs from above) is evaluated in distinct EIS, RH, and LWP regimes regionally. The product of each susceptibility and $\Delta\ln$(AI) is the resulting forcing of the aerosol-cloud-radiation interaction:

$$\text{Forcing} = \sum_{\text{l = 1}}^{\text{N}_{\text{Reg}}} \sum_{\text{k = 1}}^{\text{N}_{\text{LWP}}} \sum_{\text{j = 1}}^{\text{N}_{\text{RH}}} \sum_{\text{i = 1}}^{\text{N}_{\text{EIS}}} (\lambda_{i,j,k,l} \times \text{W}_{i,j,k,l}) \times \Delta(\ln(\text{AI})) \tag{9}$$

where $\text{W}_{i,j,k,l}$ is the weighting factor, N is the number of limits imposed, and λ is the susceptibility being evaluated (ERFaci$_{warm}$, RFaci$_{warm}$, or CA) regionally (N_{Reg}) with constraints on LWP, EIS, and RH_{700}. $\text{W}_{i,j,k,l}$ weights the ERFaci$_{warm}$, RFaci$_{warm}$, and cloud adjustments by the number of observations in each regime and also by the areal size of the region.

Constraints on LWP reduces the [..[13]]secondary effects of aerosol on LWP or LWP on susceptibility, as aerosol can result in thicker clouds and thicker clouds may have a damped reaction [..[14]]to aerosol. Constraining the meteorology separates signals forced by [..[15]]aerosol and the environment (Stevens and Feingold, 2009). On a global scale the approach outlined in DL19 identifies regime specific behavior; when applied on regional scales, the regimes allow a process level understanding of the mean regional behavior (Mülmenstädt and Feingold, 2018). This approach is optimal for our satellite based observations where larger scale parameters like AOD, AI, and cloud extent are less impacted by retrieval errors than specific properties of the aerosol.

The RFaci$_{warm}$ and cloud adjustment susceptibilities are first understood with limits on the environment and cloud states on a global scale. Their individual forcings are then found with constraints on the environment and cloud state regionally and contrasted against initial estimates of the ERFaci$_{warm}$ evaluated under the same constraints. The susceptibility estimates are not forcings. Forcings are the product of the susceptibilities (λ_{RFaci} or λ_{CA}) and the change in the aerosol index from pre-industrial times to current estimates (Δ ln(AI)). It is possible that even these estimates of forcing are slightly different than the definition of forcing from the IPCC or model based studies which difference top-of-atmosphere forcings in polluted vs. non-polluted GCM runs (Penner et al., 2011). The sum of these forcings, which we will term the decomposed ERFaci$_{warm}$, is contrasted against the simple expression for ERFaci$_{warm}$ evaluated directly using Equation 2. By separating out the individual components of the ERFaci$_{warm}$, the physical processes of aerosol-cloud-radiation interactions can be better understood. The difference between the ERFaci$_{warm}$ and the decomposed ERFaci$_{warm}$ represents uncertainty in the linear decomposition owing to covariability, non-linearity, and other effects not quantified by our approach. In reality, there should be a covariability term at the end of Equation 4 to relate how a change in RFaci$_{warm}$ may affect cloud adjustment processes or vice-versa, however a limitation of satellite observations are that they cannot temporally relate events meaning covariance between the two terms cannot be accurately quantified (Seinfeld et al., 2016). We focus on the main cloud adjustment, the effect of aerosol on the cloud extent/lifetime, however other cloud adjustment effects exist that our simple calculation of a decomposed ERFaci$_{warm}$ misses, such as how precipitation suppression directly leads to changes in cloud extent or how suppression could lead to a later invigorated state of the cloud and faster dissipation.

Precipitation is indicated by the 2C-RAIN-PROFILE rain rate along the entire 12 km track segment (L'Ecuyer and Stephens, 2002). The decomposition susceptibility is found for precipitating and non-precipitating scenes globally using equation 9. Only the decomposition terms are found separately for precipitating and non-precipitating pixels. The CERES footprint is larger than the CloudSat's, meaning while CloudSat could see an entire 12 km along track segment with no rain, the CERES footprint could still contain rain and influence the regression.

Uncertainty in each effect is found first by assuming the uncertainty in the observations lies in the AI, then by assuming a majority of the overall uncertainty in the ERFaci$_{warm}$ from error in the pre-industrial aerosol concentration estimates (Hamilton et al., 2014). Error is added randomly to AI to find how aerosol swelling or inaccurate retrievals of aerosol near cloud could

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alter susceptibility estimates. Aerosols swell in the vicinity of clouds, which increases their size and affects the MODIS retrieved AI (Christensen et al., 2017). To assess how significantly this may affect results we have randomly added errors of 10% to our AI estimates and re-derived all signals with all regime constraints. Even with extreme amounts of error in AI, the signals within our environmental and LWP regimes are robust. Uncertainty in the observations is most likely to come from the AI as CloudSat 2B-FLXHR-LIDAR fluxes have been shown to have at most $\sim$10 Wm^{-2} error in shortwave top-of-atmosphere fluxes (Henderson et al., 2013). The error from AI is then combined with randomly adding error to the pre-industrial AI estimates from SPRINTARS to quantify how error in the pre-industrial aerosol may lead to uncertainty in the ERFaci$_{warm}$, RFaci$_{warm}$, and cloud adjustments. Overall, the majority of uncertainty in any ERFaci estimate lies in the pre-industrial aerosol estimate (Chen and Penner, 2005; Carslaw et al., 2013; Stevens, 2013).

3 Results and Discussion

3.1 Estimate of the ERFaci

The warm cloud ERFaci, or the effective radiative forcing due to aerosol cloud interactions is -0.32 Wm^{-2} when found with constraints on the liquid water path, stability, and free atmospheric relative humidity applied regionally. As stated before, a negative ERFaci/RFaci/Cloud Adjustment denotes additional cooling due to aerosol-cloud interactions. Figure 1 shows each component of Equation 9 and the resulting regional distribution of the ERFaci$_{warm}$. The ERFaci$_{warm}$ is found applying Equation 2 regionally with regime constraints following DL19. This is within the range reported by the fifth IPCC report (-0.05Wm^{-2} to -0.95Wm^{-2}) but suggests the net cooling effect is toward the lower end of the expected range. Note, however, that this estimate neglects contributions from cold or mixed phased clouds and land regions (Boucher et al., 2013). This first estimate of the ERFaci$_{warm}$ represents the sum of all effects of aerosol on the warm cloud radiative effect with no distinction between the RFaci$_{warm}$ and CA$_{warm}$ and is representative of how aerosol-cloud interactions may be altering the current radiative budget (Carslaw et al., 2013).

As expected, marine stratocumulus decks in the Southeast Pacific and South Atlantic exhibit the largest ERFaci$_{warm}$, exceeding -3.0 Wm^{-2} off the coast of Chile. The peak cooling is observed in the southern hemisphere, where the marine stratocumulus cloud decks subsist due to the strong inversions and cool sea surfaces (Wood, 2012). The storm tracks region in the north Atlantic exhibit a slight cooling, as do the marine stratocumulus off the coast of California, however the southern hemisphere dominates the cooling effect. Some regions where dimming occurs are amplified by the change in emissions of the region, such as the Asian coast.

Interestingly, ACI is responsible for a net warming of as much as 0.6 Wm^{-2} in the tropical Atlantic and Indian oceans. Diagnosing the cause of this warming cannot be done through the ERFaci$_{warm}$, as it is impossible to accurately attribute it to a reduced albedo or cloud adjustment process. This signature, in particular, motivates decomposing the ERFaci$_{warm}$ into the RFaci$_{warm}$ and cloud adjustment components to allow the instantaneous albedo response to be separated from slower cloud processes. The physical processes resulting in a warming differ between the two components as the cloud adjustments are on a macrophysical scale while the RFaci$_{warm}$ is due to microphysical interactions between aerosol and CCN. The decomposition in Equation 3 allows the specific underlying physical processes responsible for this positive (warming) forcing to be assessed regionally.

The change in aerosol index is most notable off the coast of Asia and along the European coasts. Emissions from large coastal cities lead to large increases in AI, particularly changes in sulfuric aerosol (McCoy et al., 2017). The AI may have decreased off the coast of Australia due to the overall aerosol size increasing, which would decrease the Angstrom exponent and therefore AI (Carslaw et al., 2017). The northern hemisphere has had much larger changes in AI since pre-industrial times compared to the southern hemisphere due to the differences in anthropogenic activity between the two hemispheres. While the southern hemisphere has not experienced the same extreme changes in AI as the coast of Asia, the strong susceptibility of these warm clouds to aerosol combined with the local expansive clouds leads to a large cooling signal throughout the southern oceans.

3.2 Impact of LWP

Cloud LWP plays an integral role in modulating the strength of aerosol-cloud interactions. When first theorized by Twomey in 1977, the LWP of the cloud was considered to be constant as the first effect takes place. With this in mind, we first hold the LWP approximately constant and evaluate the decomposition susceptibility, Equation 4, within distinct LWP regimes. While both the RFaci$_{warm}$ and cloud adjustments are dependent on LWP, they appear to have inverse relationships (Figure 2). λ_{Sum} is found to increase with increasing LWP, reaching a peak susceptibility between 0.06 and 0.15 kgm^{-2} before asymptotically leveling off in the thickest LWP regime between 0.2 to 0.4 kgm^{-2}. For the lowest LWPs, the cloud adjustment susceptibility dominates. This reverses in slightly thicker clouds at around 0.08 kgm^{-2}. The RFaci$_{warm}$ susceptibility grows to $\sim$ 20 Wm^{-2}ln(AI)$^{-1}$ after 0.08 kgm^{-2}, while the cloud adjustment susceptibility damps and oscillates around 0 after 0.25 kgm^{-2}.

Thicker clouds are less susceptible to precipitation suppression, the key process to initiating many of the cloud adjustments (Sorooshian et al., 2009; Michibata et al., 2016; Fan et al., 2016). This is reflected in the very muted cloud adjustment susceptibility for higher LWPs past $\sim$0.1 kgm^{-2}. This inflection point is also where precipitation is more likely to occur in warm clouds and could be a sign of precipitation modulating the effects of aerosol on the cloud fraction (Lebsock et al., 2008; L'Ecuyer et al., 2009; Stevens and Feingold, 2009). An alternative explanation is that thicker clouds with larger LWPs are more likely to precipitate, scavenging aerosol and weakening the susceptibility. Aerosol-cloud-precipitation interactions complicate cloud adjustment processes in higher LWP clouds; the true susceptibility may be masked by covariance between aerosol and precipitation in these clouds (McCoy et al.). Precipitation would have an instantaneous effect on many cloud adjustment processes as major sink of liquid water within the cloud and therefore dampening process to other possible adjustments. Our framework for the cloud adjustment effect only considers processes which impact, either directly or indirectly, the cloud fraction. At higher LWPs, there are precipitation and other adjustment processes we do not account for that may later on change the radiative properties of the clouds, such as invigoration increasing the cloud depth and therefore both the longwave and shortwave cloud radiative effect (Rosenfeld et al., 2008; Koren et al., 2014).

Figure 2 confirms that LWP is intrinsically tied to the cloud albedo and extent necessitating the use of cloud state constraints on the decomposed ERFaci$_{warm}$. While a change in LWP is itself considered a cloud adjustment, it is harder to establish a causal relationship between LWP and aerosol than cloud extent and aerosol due to the manifold of environmental parameters LWP depends on. LWP being held approximately constant in some subsequent analysis should therefore reduce the impact of the LWP adjustment on cloud extent. While LWP being held approximately constant accounts for some variability in the meteorology, explicitly holding the stability and free atmospheric contributions fixed within regimes of EIS and RH_{700} will further control [..[16]]modulation of λ by the environment. Modulation may by the environment can include the amplification of the reaction through a stable environment further prolonging the cloud lifetime and therefore extent.

While regime constraints on LWP do reduce the covariability between aerosol-cloud interactions and the role LWP plays in regulating these interactions, it does not remove all sources of covariability between LWP, aerosol, the environment, and cloud properties. Aerosol has been shown to negatively correlate with LWP (Gryspeerdt et al., 2019a). It is possible that this relationship, and the inherent relationship between the environment and LWP, could affect results shown.

3.3 Constrained by local meteorology

When further separated by meteorological regimes defined by stability and RH_{700} of the free atmosphere, the influence of the environment becomes clearer as strong variations in both the sign and magnitude of RFaci_{warm} and CA_{warm} with environmental regime are evident (Figure 3). Both the RFaci_{warm} and cloud adjustment susceptibilities show warming responses in the most unstable, driest regimes. This is likely due to both the albedo and cloud extent being heavily influenced by entrainment-evaporation feedbacks (Small et al., 2009; Christensen et al., 2014). λ_{CA} shows a warming in the highest humidity, most stable regimes which may reflect cloud breakup processes like the stratocumulus to cumulus transition.

The total decomposed ERFaci_{warm} susceptibility, given by the sum of both the RFaci_{warm} and cloud adjustments within each individual stability and humidity regime, exhibits strong regime specific susceptibilities demonstrating the importance of understanding the total warm cloud radiative response to aerosol with consideration of the environment. Constraints on meteorology allow us consider how meteorology influences the cloud response to aerosol. Without these constraints, any derived susceptibilities could be attributed environmental responses. While cloud darkening occurs in only the most unstable regime ($<$ -1.8 K), λ_{CA} continues to show a warming response in moderately neutral environments ($\sim$2K). This suggests that the instantaneous response (RFaci) is more sensitive to local meteorology than the slower cloud adjustments.

The dominant cooling of λ_{RFaci} and λ_{CA} in stable regimes illustrates the potential of a stable inversion to strengthen ACI. The peak cooling of λ_{CA} occurs in a relatively dry atmosphere $\sim$27% RH_{700}. In this environment, the cloud extent rapidly increases as a response to aerosol, however the cloud is topped by a strong, stable inversion that prohibits much of an deepening of the cloud perhaps instigating the effect to push horizontally rather than vertically (Christensen and Stephens, 2011). λ_{RFaci} peaks in stable, but comparatively more moist environments where entrainment of moist air from the free atmosphere promotes activation of all available aerosol to CCN, rapidly increasing the albedo. This response may be similar to other regions where trade cumuli form and the FA is relatively moist (Koren et al., 2014).

Finally, while λ_{RFaci} shows less variation in sign, it exhibits more variation in magnitude between meteorological regimes indicating the importance of accounting for meteorological influences in order to capture this specific environmental regime

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dependence. It is possible with additional constraints, understanding how other components of the meteorology is affecting these terms would become more clear. It is also possible λ_{RFaci} is impacted by some semi-direct effects by smoke aerosol which would lead to a cloud dimming and positive susceptibility. Semi-direct effects are not accounted for by our methodology, however aerosol within the cloud layer could lead to cloud breakup processes, a dimmer albedo, and changes to the local environment by the absorbing aerosol.

3.4 Constraints on cloud state and local meteorology

As seen in Figures 2 and 3, the susceptibility of each component of the ERFaci$_{warm}$ varies with both cloud state and environmental regime. Therefore, when calculating each component of the ERFaci$_{warm}$, both the meteorology and LWP must be accounted for. To accomplish this, the RFaci$_{warm}$ and CA$_{warm}$ susceptibilities are found with constraints on both the LWP and environment (Figure 4). The shaded region of Figure 4 delineates the 10 to 90% range within each of the 11 cloud states of the susceptibility when further separated by the 100 environmental regimes used in Figure 3. Unlike Figures 2 and 3, λ is weighted by frequency of occurrence within each environmental state. This illustrates how the magnitude and sign of each susceptibility can vary by environmental regime even when LWP is held approximately constant. The weighted and summed susceptibility is -5.45 Wm^{-2}ln(AI)$^{-1}$ with constraints on LWP, stability, and RH$_{700}$ globally. This is slightly smaller than the susceptibility found in DL19, however that susceptibility took into account all changes in warm cloud CRE to aerosol while our decomposition only accounts for the two largest effects, the albedo and cloud extent susceptibilities to aerosol. The lowest LWP clouds ($\leq$ 0.1 kgm^{-2}) contribute most to the net susceptibility due to their abundance but also exhibit the widest range in susceptibilities across different meteorological states.

The two components exhibit different behavior in terms of susceptibility to cloud state (defined here by LWP). The cloud adjustment susceptibility is largest for the lowest LWPs, while the RFaci$_{warm}$ susceptibility peaks around 0.06 kgm^{-2} and gradually declines. This may represent a "sweet spot" of cloud albedo susceptibility. Up to 0.1 kgm^{-2}, aerosol are easily activated and there are few processes beyond entrainment and activation to reduce the concentration within the cloud layer. Beyond 0.1 kgm^{-2}, where the RFaci$_{warm}$ begins to decrease, the cloud may be influenced by precipitation formation, reducing the λ_{RFaci} within each environmental regime.

λ_{CA} decreases in magnitude with LWP. Higher LWP clouds, independent of the environment, may be less susceptible to lifetime effects, as was seen in Figure 2. Precipitation suppression, the main driver of cloud adjustments, becomes less likely as LWP increases (Fan et al., 2016; Sorooshian et al., 2009). The thinnest and smallest clouds may have the the largest potential to experience a enhancement effect.

3.5 Impact of precipitation and environment on susceptibility

Precipitation formation within the cloud and the environment surrounding modulate the susceptibility. When weighted by the relative frequency of occurrence, rather than overall frequency of occurrence, the susceptibility of precipitating clouds is shown to be much higher in some environments than non-precipitating clouds (Figure 5). Precipitating clouds in humid environments especially, defined as having a RH$_{700} > 44\%$, have a much greater susceptibility than any other regime of clouds.

Unstable clouds show a reduced susceptibility in all cases, with precipitating clouds showing a warming effect in these environments while non-precipitating clouds experience an extremely damped cooling effect. Unsurprisingly, in dry environments and stable environments, precipitation does less to magnify the susceptibility and the difference between precipitating and non-precipitating susceptibilities is reduced.

Precipitating clouds reduce the amount of aerosol available to interact with warm clouds through wet scavenging, yet still may induce several other processes within the cloud that stimulate a response Gryspeerdt et al.. These include stabilizing the boundary layer through virga, increasing the EIS and therefore susceptibility (Figure 3). Precipitation formation within the cloud induces vertical motion and mixing of the cloud layer, increasing turbulence and mixing of the layer which may increase activation of aerosol and therefore the response of the cloud. Further work must be done to resolve how and to what magnitude precipitation alters the warm cloud radiative susceptibility to aerosol.

3.6 Contribution of RFaci and cloud adjustments to global ERFaci

With these considerations in mind, the sum of the RFaci$_{warm}$ and CA, or the decomposed ERFaci$_{warm}$ as we will refer to it, is -0.26 $\pm$.15 Wm^{-2} found using Equation 9 (Figure 7). The components of the ERFaci$_{warm}$, the RFaci$_{warm}$ and cloud adjustments, are found using Equations 5 and 7 and shown in Figure 6. The ERFaci$_{warm}$ from Figure 1 is slightly larger in magnitude than the decomposed ERFaci$_{warm}$. Overall, their regional variations and magnitudes are extremely similar, suggesting the linear decomposition captures a majority of the ERFaci$_{warm}$. The southern ocean dominates the decomposed ERFaci$_{warm}$, as is expected based on the susceptibilities. The difference in overall magnitude stems from a stronger dimming effect evaluated in the decomposed ERFaci$_{warm}$ (Figure 6). In the decomposed ERFaci$_{warm}$, more regions experience a decrease in CRE with increasing AI compared to the ERFaci$_{warm}$. This may be due to the definition of the decomposed ERFaci$_{warm}$ that allows either the RFaci$_{warm}$ or CA$_{warm}$ to reduce cooling.

A reduced albedo, or positive RFaci, has been noted by other observation based studies before (Chen et al., 2012). A positive RFaci$_{warm}$ can be caused by multiple processes. A semi-direct effect, where non-activated aerosol acts to decrease the total albedo of the cloud in the case of smoke, reducing the CRE of the cloud and therefore the RFaci$_{warm}$ (Johnson et al., 2004). A decrease in the RFaci$_{warm}$ may also be due to any changes to the distribution of liquid water throughout the cloud layer. In certain environmental conditions, an increase in aerosol may lead to sedimentation within the cloud throughout the entrainment zone, which could decrease the cloud albedo and therefore CRE (Ackerman et al., 2004). If these two effects combined under the "perfect storm" of aerosol and environmental conditions, the RFaci$_{warm}$ would have a large, positive effect.

The cloud adjustment term likewise undergoes a positive, or damped cooling, response in certain regions. A reduced cloud fraction due to aerosol-cloud interactions has been noted before by others (Small et al. (2009), Gryspeerdt et al. (2016)). Chen et al. (2014) noted a decrease in LWP due to an increase in AI in their observationally based study, while other studies have indicated the LWP response and therefore cloud fraction response can be either positive or negative (Gryspeerdt et al., 2019a). Any process that alters the cloud's liquid water path, such as evaporation-entrainment, may lead to a decrease in cloud fraction given certain environmental conditions.

The discrepancy between the two estimates of ERFaci$_{warm}$ (0.065 Wm^{-2}) may be cloud adjustment effects or covariance between RFaci$_{warm}$ and CA$_{warm}$ not captured by the simple regression employed here. The error between the two lies well within the bounds of error of both estimates (±.16 and ±.15). While cloud extent changes are the dominant cloud adjustment effect, changes in liquid water path due to precipitation suppression will have an impact on the radiative forcing as well. Future work on understanding and evaluating the ERFaci$_{warm}$ must include other cloud adjustments and explicitly account for covariance between the RFaci$_{warm}$ and cloud adjustments. Although they occur on different time scales, the RFaci$_{warm}$ could be thought of as reactive to cloud adjustments. So while the cloud adjustment process may take hours, an albedo adjustment occurs simultaneously.

3.7 Regional variation due to precipitation

Figure 5 clearly demonstrates that precipitation plays a leading role in modulating the magnitude of aerosol-cloud interactions and their resultant forcing. The contribution of precipitating and non-precipitating clouds to the ERFaci$_{warm}$ is presented in Figure 8. Precipitation has a large impact on [..[17]]both the RFaci$_{warm}$ and cloud adjustment processes, indicated by the difference in global magnitudes between the decomposed ERFacis when separated by precipitation (-.21 Wm^{-2}) and not separated by precipitation (Figure 7 -.26 Wm^{-2}). Precipitating clouds exhibit different microphysical processes and therefore pathways of aerosol-cloud interactions that lead to an increased susceptibility (-43. Wm^{-2}ln(AI)$^{-1}$ vs. -30. Wm^{-2}ln(AI)$^{-1}$ weighed individually). However, on average only ∼30% of warm clouds observed by CloudSat are precipitating, leading to a smaller net contribution to the total ERFaci$_{warm}$ shown in Figure 8. If in future climates, warm clouds rain more frequently, it is possible that the decomposed ERFaci$_{warm}$ could increase due to precipitating clouds higher susceptibilities, given the environmental conditions (EIS and RH) remain constant.

In regions where trade cumulus are more prevalent and the marine boundary layer is more unstable, precipitation clouds have the capacity to greatly decrease the cooling due to ERFaci$_{warm}$ (Figures 5, 8). However, this positive ERFaci$_{warm}$ is balanced by their expansive cooling throughout the southern ocean. More regions experience a cooling due to ACI when clouds are precipitating than not precipitating. Further, due to wet scavenging of aerosol, it is possible that precipitating clouds could reduce semi-direct or direct effects and remove aerosol that could otherwise warm the atmosphere. The possible feedbacks or consequences of changes in precipitation require further research, especially since precipitation is heavily controlled by aerosol type as well as concentration.

4 Conclusions

The global distribution of [..[18]]the warm, marine cloud ERFaci and its components, the RFaci$_{warm}$ and cloud adjustments, are found with constraints on cloud state and local meteorology following the methodology of DL19. The total effective radiative forcing due to aerosol-cloud interactions is -0.32 ±0.16 Wm^{-2}. The radiative forcing due to aerosol-cloud interactions is -

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[18] removed: ERFaci$_{warm}$

0.21 ±0.12 Wm^{-2}. The forcing due to cloud adjustments from aerosol-cloud interactions is -0.05 ±0.03 Wm^{-2}. In all cases, constraining the environment and cloud state are found to be critical for reducing error in misrepresenting aerosol-environment effects as aerosol-cloud interactions. Our estimations of the $ERFaci_{warm}$, as a sum and/or single term, agrees with other estimates of the warm cloud $ERFaci_{warm}$ such as Chen et al. who estimated -0.46 Wm^{-2}, and with Christensen et al. who estimated -0.36 Wm^{-2}. The latter further showed liquid clouds dominate the $ERFaci_{warm}$ over mixed-phase and ice phase aerosol-cloud-radiation interactions. Thus changes in the warm cloud susceptibility to aerosol perturbations could substantially alter the radiative balance due to the warm cloud dominance of the $ERFaci_{warm}$.

Regionally, the $ERFaci_{warm}$ derived from the linear decomposition into $RFaci_{warm}$ and cloud adjustments agrees moderately well with that derived directly from the SW CRE, proving our method of decomposing the $ERFaci_{warm}$ to the first order components does capture the main effects adequately. Globally, the $ERFaci_{warm}$ is dominated by the $RFaci_{warm}$, however the cloud adjustment term is found to contribute $\sim\frac{1}{5}$ of the total forcing. The cloud adjustments vary regionally in sign and magnitude, meaning in some regions the two effects are additive, while in others they may cancel each other out. In the south Atlantic, both effects lead to a warming, or positive, forcing as clouds both shrink and dim in this region, most likely due to the prevalence of a drier free atmosphere and unstable boundary layer in this region. In the tropical Pacific, clouds dim while the cloud extent swells, leading to an overall muted cooling effect. Regions like this where the two signals have opposing signals are prime examples of why a decomposition of the $ERFaci_{warm}$ into its components is necessary. The muted signal in the tropical Pacific would most likely be attributed to [..[19]]offsetting reactions in the $RFaci_{warm}$ and CA_{warm}, as this region shows a damped signal of $ERFaci_{warm}$, if not for the knowledge that the $RFaci_{warm}$ and CA_{warm} have opposing responses in this region.

It is possible our simple methodology to evaluate cloud adjustments underestimates the possible forcing due to other adjustment processes or the possible covariance with the $RFaci_{warm}$. If the difference between the $ERFaci_{warm}$ and the sum of the $RFaci_{warm}$ and cloud adjustments is assumed to arise from the missing forcing from other adjustments, the total contribution of the CA_{warm} to the $ERFaci_{warm}$ would increase to -0.11 Wm^{-2}, or nearly a third, of the -0.32 Wm^{-2}. This would be consistent with a recent estimate by Rosenfeld et al. which found the relationship between Nd and cloud fraction, when constrained by LWP, still had a significant signal. Cloud adjustments are found to dominate over the $RFaci_{warm}$ at the lowest liquid water paths. Thus in regions or climate conditions that support enhanced prevalence of thin clouds, the cloud adjustment term would increase at the expense of the $RFaci_{warm}$.

The southern hemisphere dominates the global $ERFaci_{warm}$ due ubiquitous marine stratocumulus in the South Pacific and South Atlantic. The northern hemisphere storm tracks region in the North Atlantic and marine stratocumulus off California exert $\sim\frac{1}{5}$ the magnitude of forcing observed from the southern hemispheres pristine warm clouds. Warm clouds in the southern hemisphere are predisposed for aerosol-cloud-radiation interactions.

Cloud adjustments and $RFaci_{warm}$ varying regionally in sign and magnitudes implies that there are regions and conditions where the two components could effectively cancel each other out, thwarting short term, observation-based attempts at discerning a noticeable change in cloud radiative effects due to aerosol. Moreover, the character of the clouds does not remain

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constant. Aerosol interactions that result in brighter clouds covering a smaller area, or dimmer clouds covering a larger area, represent important physical responses that may be masked by direct assessment of ERFaci$_{warm}$ from CRE alone. In these regions especially, care should be given to discerning which effect is dominating and to what magnitude.

Data availability. All data used is publicly available. Satellite observations are available as stated in the acknowledgements.

Author contributions.

Competing interests. The authors have no competing interest that are present

Acknowledgements. This work was supported by NASA CloudSat/CALIPSO Science Team grant NNX13AQ32G and through the Jet Propulsion Laboratory, California Institute of Technology CloudSat grant G-3969-1. Special thanks to Toshihiko Takemura at Kyushu University for the SPRINTARS modeled AI. All CloudSat data is available from the CloudSat Data Processing Center (www.cloudsat.cira.colostate.edu). MODIS, AMSR-E, and MERRA-2 Reanalysis data is available from the NASA Earth Data repository (www.earthdata.nasa.gov). SPRINTARS aerosol information is available at the SPRINTARS archive (https://sprintars.riam.kyushu-u.ac.jp/).

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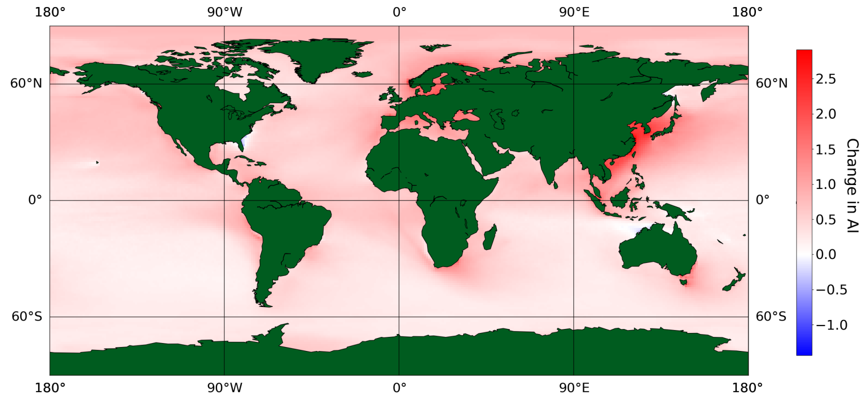

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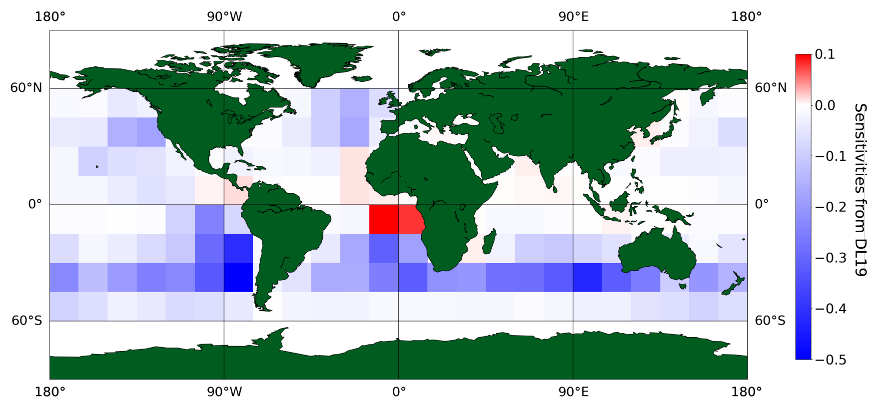

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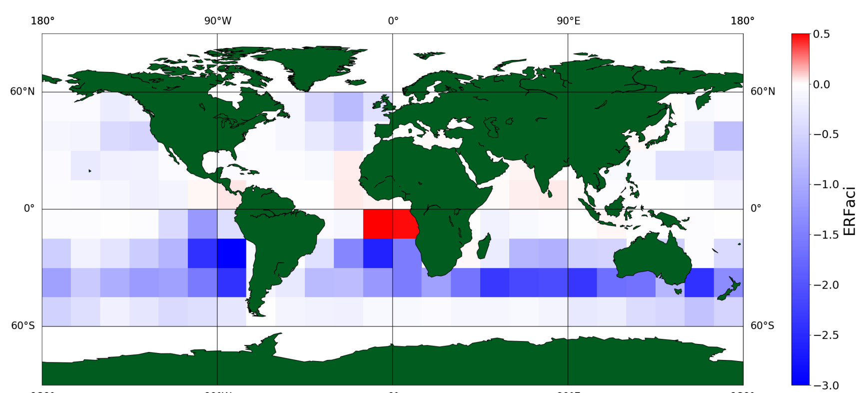


(c)

Figure 1. The change in aerosol index from SPRINTARS from the pre-industrial to present day (a), $\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})}$ adapted from DL19 (b), and the associated ERFaci_{warm} found using Equation 2 found with constraints on LWP, EIS, and RH_{700} (c, -0.32 $\pm$.16 Wm^{-2}) using susceptibilities from DL19 (b) without areal weighting.

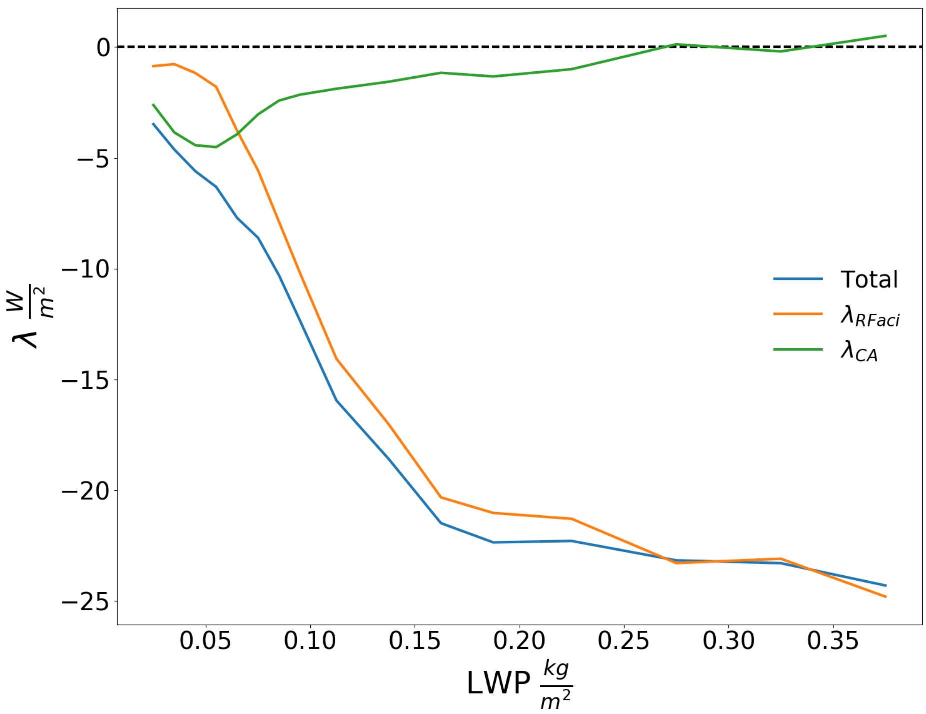
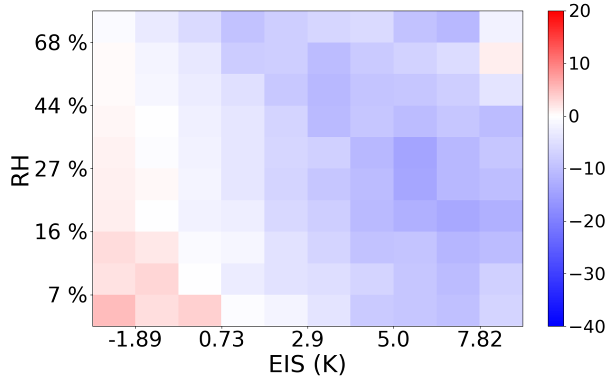


Figure 2. The RFaci$_{warm}$, cloud adjustment, and sum of the two susceptibilities, decomposition susceptibility, found within regimes of cloud state defined by LWP. The total decomposition susceptibility is -7.04 $\mathrm{Wm}^{-}2\mathrm{ln}(\mathrm{AI})^{-}1$.



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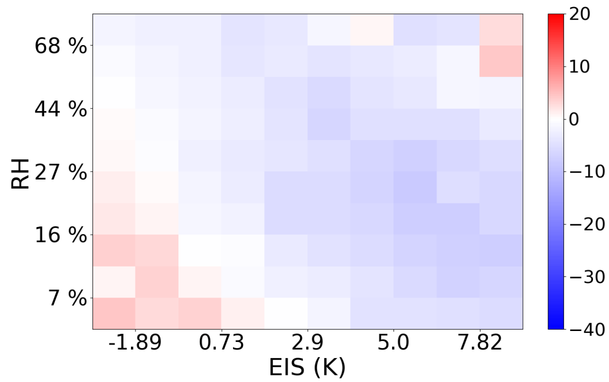

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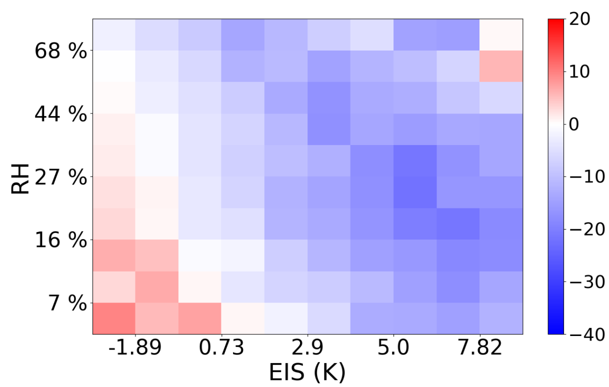

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Figure 3. Variations in the a) $RFaci_{warm}$ susceptibility (-5.26 $Wm^{-2}ln(AI)^{-1}$), b) cloud adjustment susceptibility (-2.88 $Wm^{-2}ln(AI)^{-1}$), and c) the sum of the two susceptibilities, the decomposed $ERFaci_{warm}$ susceptibility (-8.22 $Wm^{-2}ln(AI)^{-1}$) with meteorological regimes defined by EIS and RH_{700}.

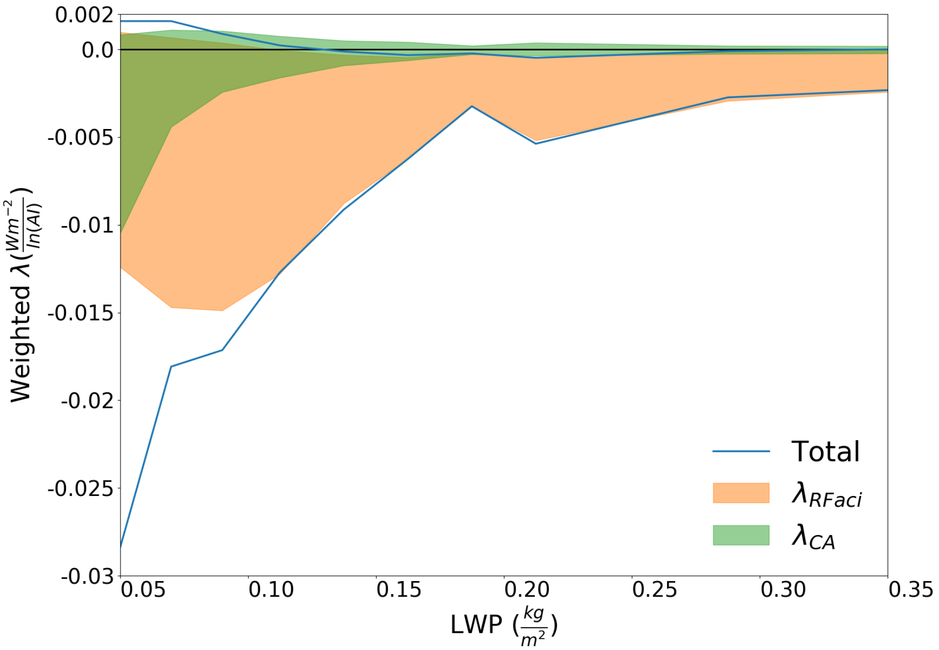


Figure 4. 10 to 90% range of the decomposition for 11 cloud states when found within 100 environmental regimes of EIS and RH_{700}. The $RFaci_{warm}$ (orange fill, λ_{RFaci}) and cloud adjustment susceptibilities (green fill, λ_{CA}) total -4.18 $Wm^{-2}ln(AI)^{-1}$ and -1.26 $Wm^{-2}ln(AI)^{-1}$, respectively. The sum of the two from 10 to 90 percentiles, the decomposed susceptibility (blue line), totals -5.45 $Wm^{-2}ln(AI)^{-1}$.

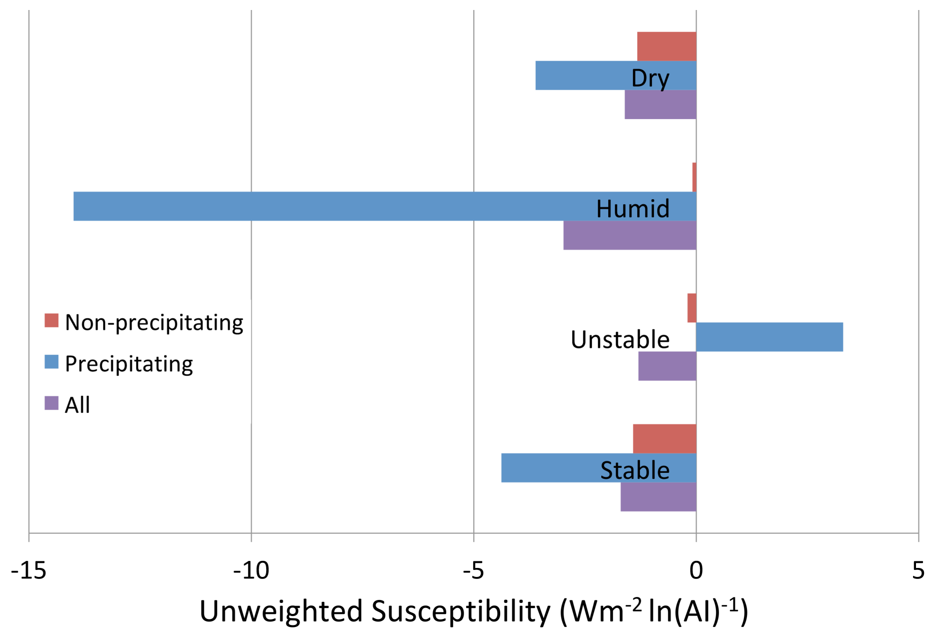
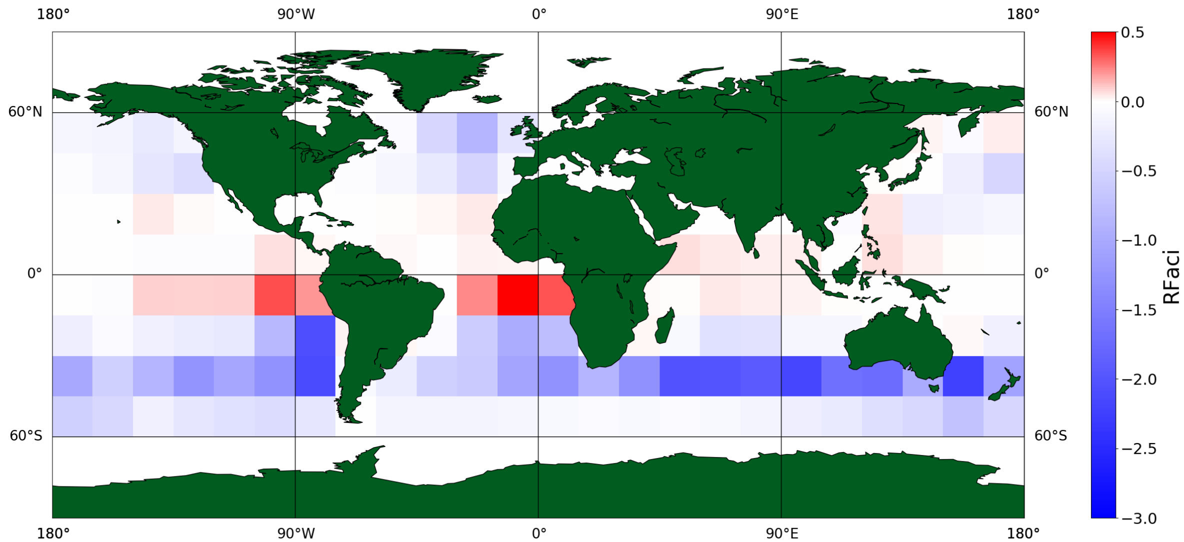


Figure 5. Globally summed and relatively weighted susceptibilities for different conditions when found within regimes of EIS, RH, and LWP on a regional basis.



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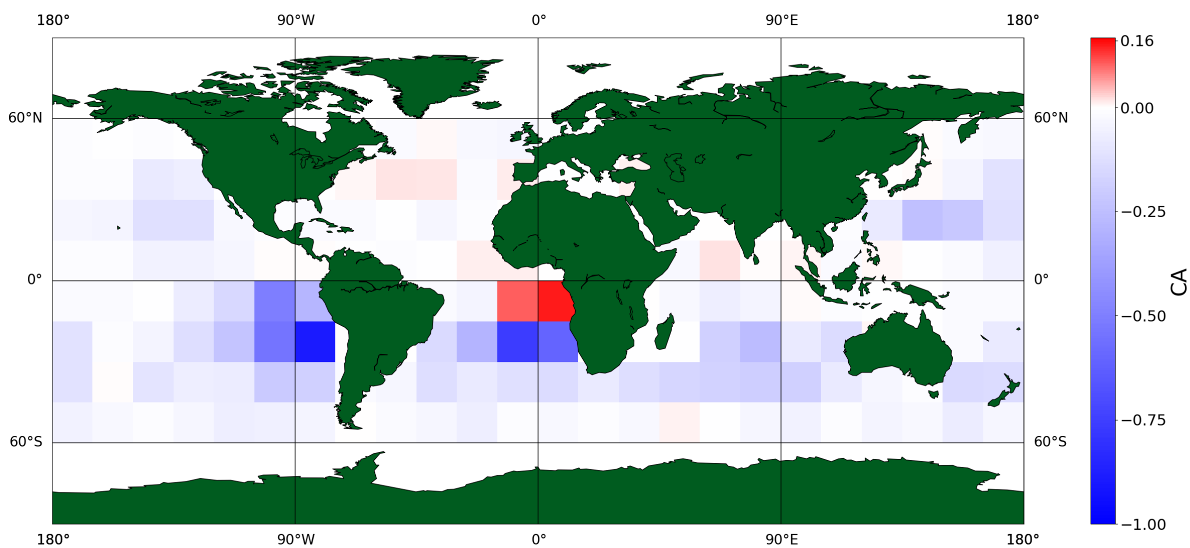


(b)

Figure 6. The radiative forcing due to aerosol-cloud interactions (RFaci) (top, -0.21 $\pm$.12 Wm^{-2}) and cloud adjustments (bottom, -0.05 $\pm$.03 Wm^{-2}) found on a regional basis with constraints on LWP, EIS, and RH$_{700}$ without weighting by area. Note the colorbar for CA$_{warm}$ (bottom) is 1/3 of the magnitude of RFaci$_{warm}$ (top).

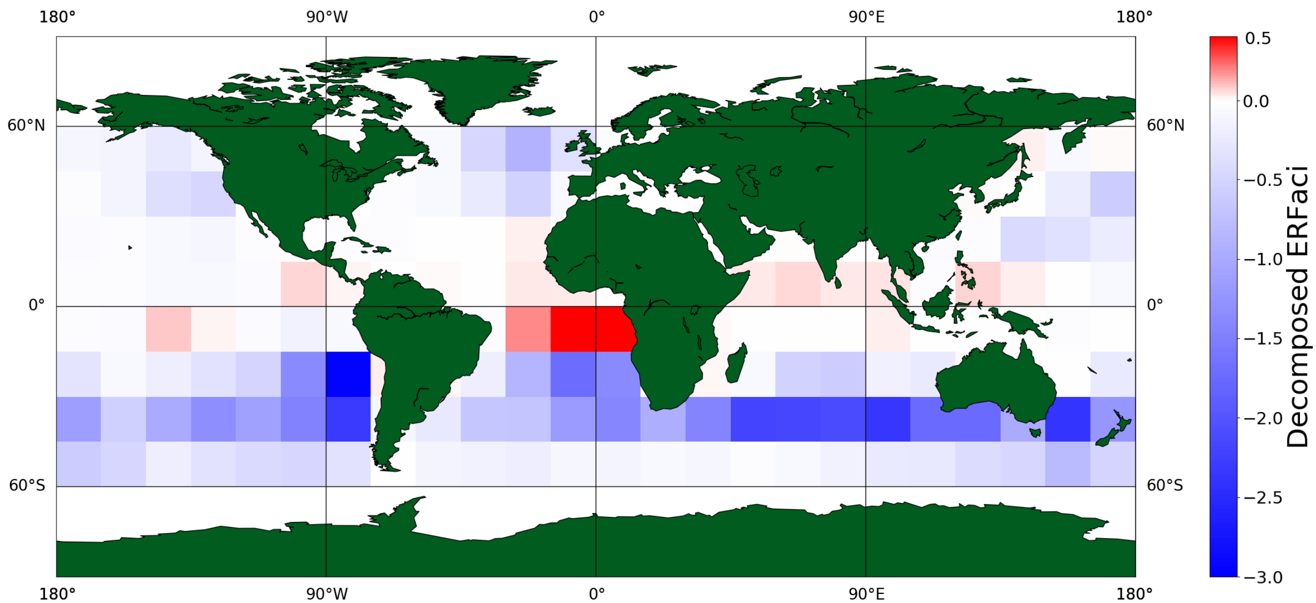
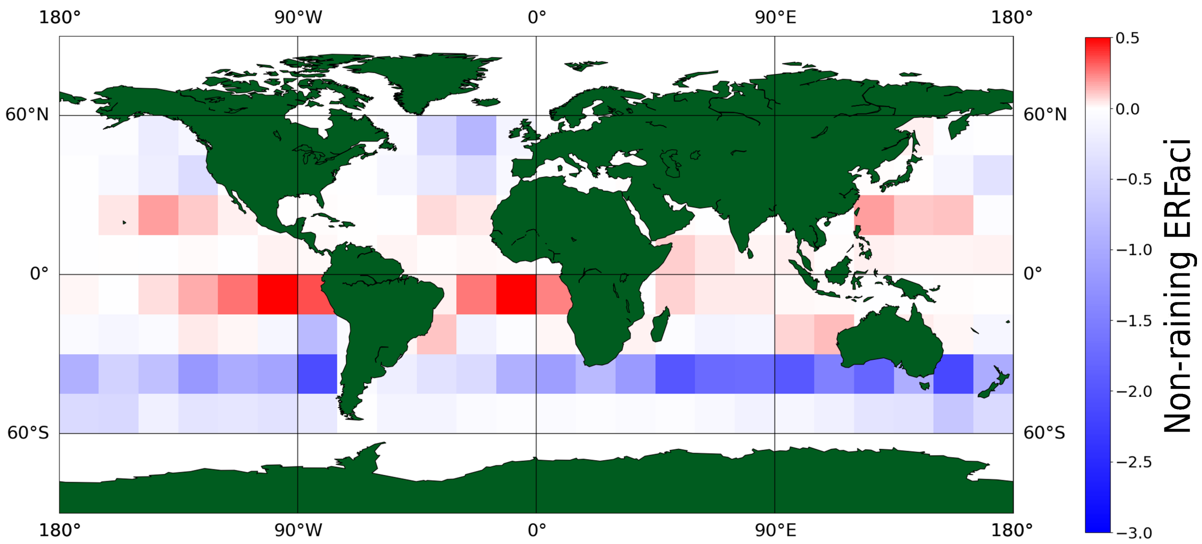


Figure 7. The ERFaci$_{warm}$ found as a sum of the RFaci$_{warm}$ and cloud adjustments (Figure 6) with constraints on the LWP, EIS, and RH$_{700}$ on a regional basis (-0.26 Wm^{-2}) without areal weighting.



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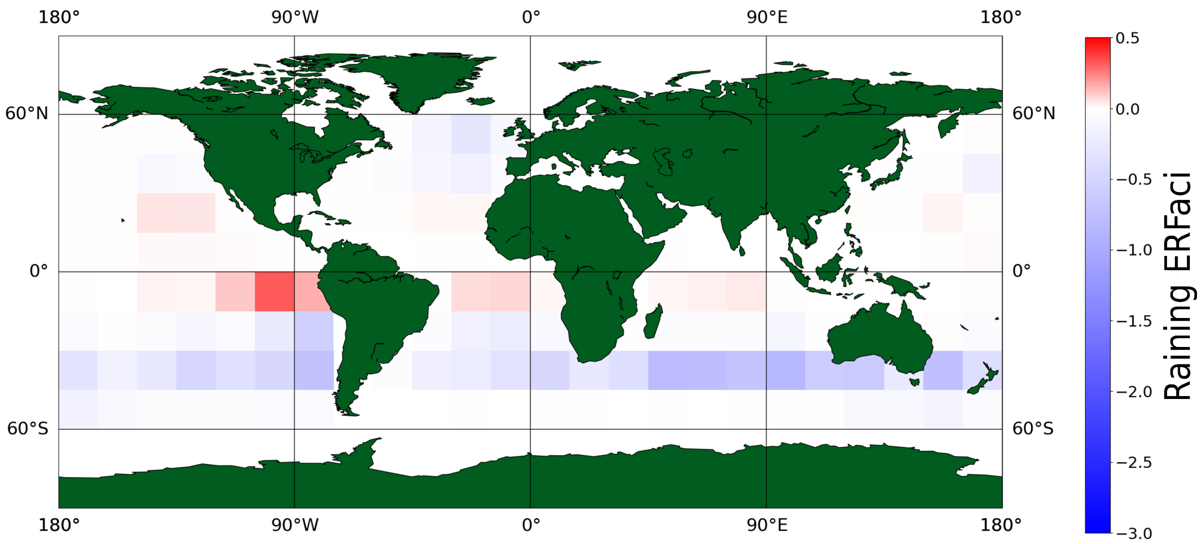

(b)

Figure 8. The decomposed effective radiative forcing due to aerosol-cloud interactions found as a sum of its components on a regional scale within regimes of EIS, RH, and LWP for a) non-raining clouds (-.147 Wm^{-2}) and b) raining clouds (-.06 Wm^{-2}).