Reviewer 1 Response

The paper investigates the effect of aerosols on cloud radiative effect while taking into account the covarying influence of meteorological factors. The sensitivity of the cloud radiative effect to aerosols is derived by sorting the data by LWP, stability and entrainment. The data is retrieved from satellite observations and reanalysis, with AI serves as a proxy for the aerosols load in the atmosphere. The results show that the global aerosol indirect effect is over estimated when not accounting for the covariability. This is probably due to buffering of the clouds response by meteorology.
We thank the reviewer for taking the time to read and comment our paper. We will go through now and address each comment below.

The authors use the term "inverse Twomey effect" which sounds physically strange. I think that the darkening of the clouds, which refers here as "inverse Twomey effect", is the response of the LWP. The LWP decreases when cloud droplets are smaller due to evaporation (entrainment), resulting in less bright clouds. This explanation is also given in the literature that the authors cite. In addition, the "inverse Twomey effect" gets much attention in the paper, perhaps more than it should. It seems to be a rather minor effect as it occupies only a small fraction of the overall samples, as shown in most of the figures.
We originally chose the term "inverse Twomey effect" as the clouds darkening go against the common assumption of the first indirect effect, however you are correct and this may have been an poor choice of words. The microphysical pathway to the darkening is not the same as the Twomey effect. We have revised our study to show that there is a general darkening effect, but the source of the darkening, whether it be a reduced cloud fraction or reduced albedo, remains unknown.
To avoid any confusion over the Twomey effect and what we were calling the "inverse Twomey effect," we changed all references of "inverse Twomey effect" to darkening or warming.

The authors write: "Constraining aerosol-cloud interactions using the local meteorology and cloud liquid water". It sounds like LWP is not part of the meteorology. However, meteorology determines boundary layer depth, and therefore also the cloud depth and LWP. Furthermore, moisture, which also controlled by meteorology in part, can alter cloud base height, and thus LWP. The authors should make it clear what they mean by meteorology.
While boundary layer depth determines the maximum cloud depth, there are variations in the LWP of warm boundary layer clouds. Decoupling, cloud breakup, and

precipitation can alter the LWP of the cloud independent of the boundary layer height. We therefore wanted to account for these processes separately from the influences of the meteorology like stability and entrainment of free atmospheric air.

We agree there should be more clarity on the difference between liquid water path and local meteorology. We have added "While the stability and entrainment directly affect the LWP, we consider the LWP separately from the local meteorology as it represents the cloud thermodynamics more than the local environmental conditions." in section 2.4.2 Cloud States page 7, line 9 to address the connections. We believe it is very common to use the term local meteorology or meteorology and not imply liquid water path.

The terminology used along the manuscript is inconsistent. For example, the authors use the term stability for both low level stability and inversion strength (though are similar). The same with entrainment and RH, cloud regimes and cloud states/morphologies. This is confusing.
We agree that the terminology should be explained and remained more consistent. We have clarified what some statements may mean in the methodology and have stuck with a consistent terminology for each type of regime.
We have added "Here, EIS is calculated using MERRA-2 temperature and relative humidity profiles and indicates the stability of the boundary layer." To section 2.4.1 page 6, line 23.
We have added to the section 2.4.2 page 7, line 1 "Although there are other definitions of cloud regimes and cloud states used in other studies (e.g. Oreopoulos et al. (2017)), throughout ours cloud state or cloud morphology refers to the set of observations binned by liquid water path." to inform the reader of the wording we have chosen for the study.
We have added to section 2.4.1 page 6, line 29 "When referring to the effects of entrainment, it means the effects of RH." to inform the reader in the methods that the relative humidity reflects the effects of entrainment on the cloud.

Instead of using aerosols indirect effect and CRE, the authors are encourage to use the IPCC new terminology that more clearly distinguishes the key mechanisms by which anthropogenic aerosols alter the energy balance of the earth (e.g., https://doi.org/10.1175/AMSMONOGRAPHS-D-15-0033.1).
In our study, we are only finding the sensitivity of the clouds, not the ERFaci. We chose to focus on the methodology of distinguishing the signal of the warm cloud CRE to aerosol from other factors in this study, not to determine the radiative forcing of aerosol-cloud interactions. Our terminology is consistent with others in the field and

we chose not to use IPCC terminology because we are not quantifying a forcing, only a sensitivity.

The captions are short and do not provide sufficient information to understand the Figures without digging into the text. Also, the captions sometimes do not present all the subplots in some of the Figures.
We agree our captions were too brief. We have added more detail to the captions to explain every part of the plot(s) shown.

"Local Meteorology" seems to be a key factor in the study (the authors chose to have it in the title). I think that this point is not enough explained in the introduction and should be emphasized more in the conclusions.
We agree that local meteorology should be focused on more in the introduction and have added "Constraining the local meteorology, or the characteristics of the environment around the cloud, as well as cloud type can significantly alter the magnitude of the AIE compared to single, unconstrained global linear regression (Gryspeerdt et al., 2014)." to page 2, line 16.

Specific Comments

P1 L24 Provide a reference.
We have added (Albrecht, 1989) as a reference for that statement.

P2 L1-2. This is a 1.5 line paragraph. Perhaps you can discuss here the relative contribution of the cloud life time effect and cloud albedo effect.
These two lines are part of the first paragraph of the introduction. The ACP Discussion formatting makes it seem like it is a separate paragraph.

P3 L17. I'm not sure a paper from 2014 can be considered "recent"
We have removed recent from that sentence.

P3 26. Decoupling between cloud and ocean? Provide references here and in the following sentences to establish the relationship between RH and decoupling.
We have added "...by increasing the temperature and humidity gradients at the cloud top (Lellewen 2002)." to explain how RH affects the decoupling process.

P3 L33. I would change effective radius to droplet size, and LWP to optical thickness.

We have changed the sentence to "In his original work, Twomey postulated that cloud albedo ought to increase with aerosol provided LWP is held fixed, after 10 all, albedo is dependent on the optical depth and effective radius." replacing LWP with optical depth.

P4 L1. AMF?

We have expanded this acronym to "Amospheric Radiation Measurement Mobile Facility."

P4 section 2.2. Please provide the spatial resolution of the data. Is the data from the different instruments is co-located to a single resolution?

We have added to section 2.2 Cloud "All data is interpolated down to CloudSat's ~1km footprint." Further, in section 2.1 Data we state "The A-Train is a series of synchronized satellites which allow for collocated observations from a variety of instruments (L'Ecuyer and Jiang, 2011)."

Why did you decide the upper threshold of LWP to be 400? Did you use optical thickness threshold to avoid additional uncertainties (see e.g. https://doi.org/10.1002/qj.2405).

We chose a limit of 400 because it removes outlier cases of convective warm clouds and other thicker clouds that are not the focus of this study. Less than 5% of warm clouds in our dataset had an LWP above 400. Additionally, having 400 as an upper limit reduces the impacts of warm rain on aerosol-cloud-radiation interactions.

If I understand correctly, the CF is determined based on a 12 km segment (P4 L29). A single open cell for example can cover 12km, which would give 100% CF, while the clear area in between cells would give 0% CF. Scaling is very important in determining the CF. You also exclude clouds with LWP

We have explained how we quantify cloud fraction further in section 2.2 Cloud page 5, line 9:

"An along-satellite track cloud fraction is determined by finding the average number of warm cloud pixels that satisfy these criteria (seen by CloudSat or CALIPSO, below the CloudSat determined freezing level, and LWP between .02 and .4 kg) over each 12 km segment of the CloudSat track on a pixel by pixel basis, a scale that represents both the local scale length of the boundary layer and field-of-view used to define cloud radiative effects from Clouds and the Earth's Radiant Energy System (CERES) (Oke, 2002)."

Our cloud fraction is pixel by pixel, meaning that as cloudiness changes at a 1km scale, the cloud fraction increases or decrease by 1/12th.

Make it clear in the second eq. that the F_all sky is only for the SW.
We have changed our statement to "It is easy to show that for the shortwave radiances:" *before F_all sky equation on page 5.*

P5. The Equations have no numbering.
The equation numbering appears on the far right of the page. There is no way to change the formatting of this as it is set by the ACP Discussion Paper template.

P5 section 2.3. You use AI as a proxy for aerosols. However, AI is retrieved only where there are no clouds. This is something that should be discussed.
We have addressed this in section 2.3 Aerosol by adding to page 6, line 9 "While AOD and the Angstrom exponent from MODIS are not available in cloud scenes, the collocated dataset interpolates these between clear sky scenes in order to infer an AI in cloudy scenes."

P5 L22. "The cloud sensitivity" suppose to be cloud albedo sensitivity?
We have clarified this further in section 2.5 Sensitivity page 7, line 26 by adding "he warm cloud radiative sensitivity to aerosol, or λ, is defined as the linear regression of the shortwave CRE against ln(AI). While other studies have called similar metrics a susceptibility, we use the term sensitivity." This is not the cloud albedo sensitivity as ours can include effects on cloud extent/lifetime. To delineate a cloud albedo sensitivity, the indirect effect/ERFaci would have to be separated by its parts, the RFaci and cloud adjustments.
We have clarified throughout the study that we are deriving the warm cloud radiative sensitivity to aerosol.

P6 L6 "inversion strength" is first mentioned here, which seems to be equivalent to the stability.
You are correct. We have added "Stability of the boundary layer is indicated by the EIS." to section 2.4.1 Environmental Regimes to clarify this.

P6. What do the numbers above the sigma mean?
The numbers above sigma represent the number of regimes. I.e. we use 7 cloud state regimes, 10 regimes of EIS, and 10 regimes of RH in equation 6, while in equation 7 the number of regimes is reduced due to sampling on a regional vs. global basis to 4 cloud state regimes, 5 regimes of EIS, and 5 regimes of RH. This is common notation when using sigma (Σ) notation of summation.

P7 section 2.6. The cloud regimes are simply LWP bins? Definition of cloud regimes is far more complex (e.g. https://doi.org/10.1002/2016JD026120).

We understand that other studies have defined cloud states/regimes differently than other studies and have added to address this in section 2.4.2 page 7, line 6 "Although there are other definitions of cloud regimes and cloud states used in other studies (e.g. Oreopoulos et al. (2017)), throughout our results and analysis, cloud state or cloud state regime will refer to observations binned by liquid water path."

P7 L30 "Low LWP clouds are less sensitive to aerosol" - but it is the thinnest clouds that response the strongest to the Twomey effect.

We have rephrased our statement to reflect that based on our results, the thinnest clouds showed the lowest sensitivity. We have added to page 10, line 13 "From Figure 2, the lowest cloud states are less sensitive to aerosol, with a steep increase at ~.8 kg/m2." This is a result seen in our analysis based on observations with minimal constraints, unlike the model Twomey used which was idealized and did not include processes that could reduce the CRE of extremely thin clouds.

P8 L9-10 Define the LWP bins.

The limits of the LWP bins can be seen on the figures and would add very little if explicitly stated in the text. We have added to further clarify how we established these limits on page 7, line 15

"The number of LWP bins decreases from global to regional analysis due to sampling; on a global scale, seven LWP regimes are used, while on a regional scale, only four LWP regimes are used. Limits are placed to separate out the signals of low LWP clouds vs. high LWP clouds, as low clouds may be affected by evaporation-entrainment feedbacks while high LWP clouds may be affected by precipitation (Jiang et al., 2006; L'Ecuyer et al., 2009). While the environmental regimes are established on a percentile basis, cloud state regimes are set by having an increased number of bins for the lowest LWP clouds and a bin limit always set at 150 g to delineate clouds which are extremely unlikely to precipitate (< 150 g/m2) and clouds more likely to precipitate (> 150 g/m2) (L'Ecuyer et al., 2009)."

Do you do any significant tests?

Yes, to include the regime in analysis it must have at least 100 observations and a Pearson correlation coefficient greater than .4. These criteria are also in place when the sensitivity is found on a regional basis, where the environmental regimes are more likely to have less than 100 observations or a worse linear fit. We have added to section 2.5 Sensitivity page 7, line 31 "The sensitivity is only included if there are 100 observations within the regime and the linear regression Pearson correlation coefficient is greater than .4."

Figure 6 panel h is not mentioned in the caption. What does the color bar mean?

The colorbar for panel h is the summed, weighted sensitivity. We have added to caption for Figure 6: "Panel (h) is the summed, weighted sensitivity within each environmental regime. The weighted, summed sensitivity is -10.6 Wm−2/ln(AI) (sum of panel (h)). Note the colorbar for panel (h) is adjusted due to weighting."

P14 L1. This sentence needs context and further discussion, rather than just stating it.

We have chosen to remove this sentence.

P16 L5 "top" of what?

We have changed this to "top panel of Figure 8."

P16 L7 "stability, entrainment and cloud morphology" are equal to EIS, RH and LWP?

Yes you are correct, we use the terms stability, entrainment, and cloud morphology interchangeably with EIS, RH, and LWP respectively in the discussion. We have addressed this through earlier comments and clarified our terminology in the Methods section.

P16 L18. An explanation regarding the relationship between entrainment and particle size is needed here.

We have added to the Discussions section Page 19, line 23 "Entrainment of drier air will force evaporation, decreasing particle size, while entrainment of moister air could have no effect or a reverse effect, increasing the number of CCN within the cloud."

Considering adding figure 9 to figure 8.

We separated them to help the reader focus on figure 9, where all constraints are in place, rather than only a panel of figure 8. Figure 9 is the final focus of our discussion and therefore is better suited to be its own standalone map, rather than a panel of figure 8.

P18 L16-18. I'm not sure about the context of Jiang et al. 2006 here. In their study the additional aerosols were related to enhanced evaporation, which limited the cloud life time. The study was focused on cumulus field.

We chose to cite Jiang 2006 as it was one of the first studies to theorize an entrainment-evaporation feedback. While their findings were limited to cumulus, this does not mean the process could apply to other warm clouds like the thinner cloud states of our study. We have added to the Discussions page 20, line 7 "…which would be *the result of forced evaporation and reduced particle size. The reduced particle size would affect the lifetime of the cloud as well as the cloud albedo, reducing the sensitivity of the warm cloud radiative effect to aerosol loading as seen in our results for some unstable, dry regions (Jiang 2006)."*

P18 L19. How turbulence decreases the activation efficiency of aerosols? Turbulence can also lead to secondary nucleation due to super saturation fluctuations.

Turbulence and higher in cloud updraft speeds can increase the efficiency of aerosol activation under certain conditions. Stable boundary layers have almost a "cap" at the boundary layer top, which acts to dampen cloud growth. Unstable boundary layers are less likely to have the "cap," meaning more turbulence and higher updraft speeds lead to higher cloud tops with possibly the same amount of activation. We have added to page 20, line 4 "Unstable conditions lead to strong vertical mixing and a reduced aerosol sensitivity, as activation favors strong vertical mixing in a stable environment. Unstable local meteorologies alter the conditions of aerosol activation (Cheng 2017)." to explain the role stability plays in modulating aerosol-cloud interactions.

P18 L26. You mention here that wind speed can affect cloud cover. Why didn't you include also wind speed in your parameters?

We chose to use only EIS and RH as constraints on local meteorology as they are the strongest modulators with CRE along with LWP. During initial analysis, using multivariate linear regressions, we found the highest correlations and amount of variance explained with EIS, RH, and LWP than suface wind. We have added to page 20, line 14 "Surface winds were not included in analysis because the dependence of the warm cloud radiative response to aerosols depends most on LWP, RH, and stability, with only some regions showing a dependence on surface winds in our initial analysis." to explain this reasoning.

P19 L19-21. I would expect decreasing stability not to decouple clouds from the surface due to more mixing. Also note that decoupling that occurs when the stability is increased can inhibit cloud breakup (https://doi.org/10.1029/2018GL078122). Please clarify. Where is the role of aerosol here?

The decoupling process occurs when warm marine boundary layer clouds move from a stable to less stable environment. A less stable boundary layer is more likely to have a higher boundary layer top height, increasing the chances of the cloud becoming decoupled from the surface. We have added to explain this process further to page 20, line 34 "The negative sensitivities seen in the unconstrained top panel of Figure 8 are *likely a result of this process, which happens simultaneously with a reduced stability, and epitomize how a single linear regression of warm cloud CRE against ln(AI) can capture meteorological effects when unconstrained (Wyant 1997)."*

P20 L10-11. It would be helpful to reference the relevant figures here and in the last paragraph where the sensitivities are given.
We have added the appropriate figure references to the Conclusions section.

Reviewer 2 Response

In general, I like the paper's approach to the critical problem of understanding aerosol impacts on clouds, and how they carefully dealt with co-varying meteorology. I also like how they combine measurements from different sources to get a broader picture of the system and to constrain the observations better. This paper has potential to be very important and useful for/referenced by a host of other studies using a similar methodology in the future.
We thank the reviewer for taking the time to read and comment on our paper. We will first address the major points, and then the specific comments.

Substantial extra work is required to clarify and explain the methodology. Another person attempting to replicate this study could probably not do so, as it is written now, and this makes it difficult for a reviewer to fully judge the value of the work. Unfortunately, by the time I reached section 3.5, the cumulative uncertainty I had with the methodology was large enough for me to have strong reservations about my ability to judge the meaningfulness of this section. That said, I do believe that if the authors answer the questions in the specific comments fully, I can better judge of the work in the next revision.
We agree that more information should be added to help those would like to reproduce our study. The methodology section has been expanded upon. We hope that by addressing the questions outline below further rectify this issue and help the reviewer and future readers understand how they could implement a similar methodology.

There is a possibility that serious errors were made regarding Fig. 5 that might have a large impact on the results. For the reasons listed in the specific comments below, I request that the authors double-check their results carefully, and if necessary, re-run the analysis.

There is an inherent relationship between the estimated inversion strength (EIS) and the relative humidity of the free atmosphere (RH). To alleviate any misunderstandings of the two meteorological variables, the relationship between EIS and RH has been explained in more detail in the Methods. The EIS depends in part on the height of the 700 mb isobar, which would directly depend in part on the relative humidity of the free atmosphere (define as 700 mb). There is some covariance between these parameters that we have now tried to address. Figure 5 is correct as it simply shows that marine warm clouds exist within environmental regimes of EIS and RH. There are well known phenomenon controlling each that lead to a relationship between the two that is not the focus of this study as could be explained further in "On the relationship between stratiform low cloud cover and lower-tropospheric stability" by Wood and Bretherton 2006. All following analysis and figures are correct according to our observations and reanalysis used.

A variety of confounding factors were ignored here, including the difficulties in co- locating aerosols and clouds based on AI, errors and biases in near-cloud satellite aerosol detection, and potential confounding influences of aerosol semi-direct and direct effects. These should be addressed and acknowledged.

These are now more distinctly addressed and acknowledged in the methods section. We agree there is some measure of uncertainty when using satellite observations to understand cloud and aerosol as clouds invariably affect near-cloud aerosol.

Throughout the paper the conclusions tended to be a bit overstated. It should be made clearer in the manuscript that the study only focuses on a subset of data, and that this subset is not necessarily broadly representative of all conditions.

The focus of the study and the conclusions drawn from the results do only apply to warm marine clouds, however these clouds are vital to understanding many different parts of the climate such as the sensitivity and radiative balance as mentioned in the introduction. A significant source of error in the IPCC's climate sensitivity is from the indirect effect. I have added more reminders in the introduction and methods that this study applies only to warm marine clouds. The importance of understanding and quantifying the warm cloud indirect effects is widely accepted. Twomey's 1977 study of the impact of pollution on Earth's albedo has been cited over 2000 times, while Albrecht's later study in 1989 has been cited over 3400 times. Aerosol impacts on continental and poleward clouds are offset by the brighter surfaces and therefore reduced impact of the indirect effect in these regions.

Specific Comments

Title, abstract, and conclusions: I suggest that the title and abstract better clarify the focus on warm marine clouds, and contain some stronger hints of the large remaining uncertainties (e.g., by adding "Better Understanding Aerosol-…." to the title). The reason for this suggestion is that large groups of clouds were excluded in this study, and there were some fairly major inherent uncertainties in the methodology. The study focused on daytime, single-layer, warm clouds, and as best I can tell, it only includes clouds with latitudes < 60^o and over the ocean. LW forcing at night was excluded entirely. Therefore, the study cannot address several important complex cloud-radiation interactions related to aerosols. For example, the method presented here would not address ice nucleating effects or seeding effects in multi-layer clouds, which can be quite complex. The results may also not be applicable to terrestrial areas where, for example, diurnal effects of heating can be much more variable.

This is true. As stated above, warm marine cloud systems are known to exert a strong influence on climate sensitivity, but these are certainly now the only cloud type on Earth. The title has been adjusted to: "Understanding Shortwave Aerosol-Cloud-Radiation Interactions in Marine Warm Clouds Using Local Meteorology and Cloud State Constraints." We have also identified the exact clouds we are studying in the abstract.

Introduction/Methods: Please clarify to the readers why RH and LWP, which are not independent variables, are considered separately, and essentially independently, in this study.

The relative humidity of the free atmosphere (defined as 700mb) and the liquid water path from AMSR-E are independent variables. The RH is primarily a function of the vertical motion in the free atmosphere and large-scale circulations, while the LWP is primarily a function of cloud depth, stability, in-cloud microphysical processes, and other boundary layer conditions. While there may be some relationship between these quantities, both can independently modulate aerosol indirect effects … two clouds with distinct LWP may respond differently to aerosols even in similar RH environments. Thus RH does not directly control the LWP of a cloud or completely define how the SW cloud radiative effect varies with aerosol concentration.

Methods: Please add a table where readers can quickly find what subset of clouds were included in the study. From Figures 8 and 9, it appears that terrestrial clouds and clouds poleward of 60o were eliminated. However, this is not explicitly stated in the paper. A concise central location to find which

latitudes, LWPs, and temperature levels, etc. for the subset of clouds assessed in this study would be useful.

We have added on page 5, line 2 "between 60◦N and 60◦S."

This information is provided in section 2.2 Cloud. We state in the first line of this section "…restrict analysis to single-layer, marine warm clouds between 60◦ N and 60◦ S" and "satisfy these criteria (seen by CloudSat or CALIPSO, below the CloudSat determined freezing level, and LWP between .02 and .4 kg)" when explaining the observations chosen for analysis. We feel this is too little information to warrant adding an entire table to the manuscript.

Section 2.2 or 3.5: A map of the frequency of observations of the subset of clouds compared to all clouds observed in the region would be very helpful for interpreting the relevance of this study. Are the types of clouds studied here more common in some locations than in others, and is there any geographic bias in Figures 8 and 9?

The focus on the study is to reduce the impact of influencing factors like RH, LWP, and EIS on estimating the warm cloud indirect effect. The frequency of clouds is not important, only the sensitivity of certain cloud regimes to aerosol. Including a map of cloud fraction or frequency would convey the message that the frequency is what determines the warm cloud indirect effect, when our study is focusing on how specific regimes of warm clouds independent of frequency can dominate the warm cloud radiative sensitivity to aerosol. Other studies on warm clouds note their prevalence globally.

We have added to the Introduction page 1, line 17 "These clouds are most prevalent off the western coasts of continents as marine stratocumulus, as trade cumulus near the tropics, and as stratus in the storm track regions (Ackerman 2018)."

p. 4, l. 27: "*An along satellite track cloud fraction is determined by finding the average number of warm cloud pixels that satisfy these criteria (seen by CloudSat or CALIPSO, below freezing level, and LWP greater than 20 g m-2)*"

What is meant by "below freezing level"? Is that determined from MERRA2 temperature profiles below 0 oC? Please clarify.

Freezing level is determined by the CloudSat 0^{o} isotherm from ECWMF-AUX product. Below freezing level means the entire cloud observed by CloudSat and other satellites collocated with CloudSat was contained to the layer at or below freezing level. The focus of our study is on liquid containing clouds only, not mixed phase or ice. Therefore, by limiting to clouds below freezing level, we guarantee the clouds do not contain ice or supercooled liquid.

As I interpret it, this suggests that the altitude or pressure level where the results are obtained varies by profile, and that this altitude would probably vary quite a bit over latitude and surface type. If this is the case, how do cloud altitudes in the study vary?

Clouds do vary with altitude, however by focusing on maritime liquid clouds, the variation will be limited by the boundary layer height. This varies with EIS, which we account for in our regime framework. In essence, by accounting for EIS, we are also accounting for any effects of cloud top height. Further, when the sensitivity is calculated on a regional basis, this will further constrain any small variations in height.

Can we rule out that vertical variation in the clouds being studied would not add substantial error or bias the results (e.g., by introducing different aerosol types at different levels, or horizontal/vertical winds, etc.)?

We cannot rule out that regional variation exists in aerosol type or cloud type, which is why the results are eventually found on a regional basis to account for some of this bias.

We have added to section 2.2 Clouds "All observations are restricted to below the freezing level of CloudSat which is determined using an ECWMF-AUX collocated reanalysis dataset and set where ECWMF determines the 0^{o} isotherm." And further on in the same paragraph we remind the readers again that observations are "below the CloudSat determined freezing level" to clarify that it is below the freezing level determined by CloudSat and not MODIS. We have also added that "Marine warm clouds fitting these parameters reside within the boundary layer." to the end of the Cloud section in the Methods to clarify these will be low-level, boundary layer clouds.

p. 5, l. 3: "The shortwave cloud radiative effect (CRE) is then defined in terms of the all sky and inferred clear sky forcings from CERES and cloud fraction from CloudSat." How is CloudSat cloud fraction defined? Which conditions are included in "all sky" conditions? From what I understand, situations when there are multi-layer clouds, and clouds below freezing temperatures, etc. are excluded. If this is correct, then the term "all sky" may be a little confusing, and perhaps other wording would be better.

Yes, as acknowledged elsewhere, this analysis is only for warm maritime clouds. The set of observations our analysis is based on is explained in detail in section 2.2 Clouds. To remind the reader that our analysis is for only a subset of clouds, we altered all "CRE" to "warm CRE" and "cloud" to "warm cloud." This is consistently mentioned further now in the results, discussion, and conclusions sections as well.

p. 5, l. 3: "*The shortwave cloud radiative effect (CRE) is then defined in terms of the all sky and inferred clear sky forcings from CERES and cloud fraction from CloudSat*." How is CloudSat cloud fraction defined? Which conditions are included in "all sky" conditions?

Cloud fraction is defined in Section 2.2 "Cloud"

> *"An along-satellite track cloud fraction is determined by finding the average number of warm cloud pixels that satisfy these criteria (seen by CloudSat or CALIPSO, below freezing level, and LWP greater than 20gm2) over each 12 km segment of the CloudSat track, a scale that represents both the local scale length of the boundary layer and field-of-view used to define cloud radiative effects from Clouds and the Earth's Radiant Energy System (CERES) (Oke, 2002)"*

From what I understand, situations when there are multi-layer clouds, and clouds below freezing temperatures, etc. are excluded. If this is correct, then the term "all sky" may be a little confusing, and perhaps other wording would be better.

We have added "All-sky radiances from CERES are not restricted to any type of scene and include the raw radiances observed by CERES." to section 2.2.

p. 5, l. 15: Which version of MODIS is used, and why? What is the resolution of these data and how does that relate to the cloud resolution?

We have added to section 2.3 Aerosol "MODIS AI is derived from the auxiliary dataset (MOD06-1km-AUX) developed from the overlap of the CloudSat CPR footprint and the MODIS cloud mask at pixel level."

Section 2.3: MODIS has a variety of known issues with reliably detecting aerosols near clouds. What kind of cloud screening was used, and how sensitive are the results to this choice? It would probably be useful to note in the paper that binning the data by RH conditions could create some biases, due to aerosol swelling near high-humidity conditions typical near clouds.

We have added

> "While AOD and the Angstrom exponent from MODIS are not available in cloudy scenes, the collocated dataset interpolates these between clear sky scenes in order to infer an AI in cloudy scene. For lower cloud fraction scenes, this interpolation is more accurate, however it is possible that in higher cloud fraction scenes, the accuracy of AI is reduced. This is a source of uncertainty within our results, but with constraints on cloud state, the error of this interpolation method should be reduced. Binning by relative humidity

when evaluating the sensitivity should reduce some bias from aerosol swelling in humid environments.

to section 2.3 Aerosol.

Equations 2-5: On the first read-through, I was quite confused about the upper limits of summation (e.g., the number 7 in equation 2). Justifying the specific choice of those numbers might make more sense in a later section (e.g., section 2.5) than in section 2.4, where they first come up. Therefore, I suggest the authors make the equation more generalizable by having the upper limit of summation be a variable, to be assigned a value later when explanation for that value can be more logically provided. This might also help if others want to cite this method in future work, but want to use different numbers of states for their specific application. Also, please specify earlier on in the text what the upper limit of summation represents, as this was not clear in these equations and in section 2.4 in general. Moving the following text from p. 7 into section 2.4 where these limits are first introduced could help: "*The regime bounds depend on the resolution used, which is varied to establish the degree to which environmental factors must be constrained to accurately characterize sensitivity*".

We have added to section 2.5:

"Where the numbers for summation come from i.e. the number of regimes of LWP/EIS/RH."

"Where N_k is the number of observations of cloud state k"

"Where $N_{i,j}$ is the number of observations within each environmental regime:"

"Where $N_{i,j,k}$ is the number of observations within each environmental regime when constrained further by each of the state regimes k."

We have also replaced the 7, 10, and 10 with LWPs, RHs, and EISs in the summation equations to clarify what bins are being summed.

Further, we have added to section 2.4.2 "The number of cloud states can be varied. In our results, we evaluate the efficacy of increasing and decreasing the number of cloud states."

p. 6, l. 6: How is estimated inversion strength calculated? (note, some information is provided later, on p. 7, l. 2, but this information is not fully descriptive).

Added from Wood and Bretherton 2006 the equation for EIS to section 2.4.1 Environmental Regimes.

$$EIS = LTS - \Gamma_m^{850}(z_{700} - LCL)$$

P. 7, l. 4: "*The relative humidity at 700 mb is used as a measure of the effect of entraining free tropospheric air.*" As I understand it, the RH at on vertical level is assumed to be representative of the whole vertical column up to the freezing point, or at least to provide important information for the whole column. However, RH at 700mb will be most relevant for clouds in that general altitude range. Will this bias the results, or add error? What is the variability in cloud locations? This was not quantified. Why not just use RH at the appropriate altitude ranges where the cloud layer is found?
700 mb is the most common level used to represent the free atmosphere. Boundary layer clouds entrain free atmospheric air, so using a level like 700 mb ensures we're getting an accurate picture of the air entering the cloud layer without any contamination from the cloud layer itself in the relative humidity (Karlsson, 2010).

p. 7, l. 5: "*All observations within the 5% - 95% percentiles of both EIS and RH are partitioned into regimes*." As I understand it, one nice thing about taking the weighted mean is that you can use all of the data, and still get representative results. Thus, I don't understand why these data were excluded in the first place? (from the above statement, I believe the excluded data would equal between 10-20% of their subset of observations?) Was a similar procedure was not followed for LWP, and if so, why not?
The tail ends of the stability and humidity spectrums were removed because we found they biased the results to the extremes. A similar approach was taken for LWP by limiting it to 20 – 400 g/m2. These results still apply for the vast majority of warm clouds.

p. 7, l. 5: "*Environmental regime limits are defined such that there are the same number observations within each percentile of either EIS or RH. The regime bounds depend on the resolution used, which is varied to establish the degree to which environmental factors must be constrained to accurately characterize sensitivity.*" I am very confused by this part of the methodology. Please define in the text what is a bound and what is a limit. I am guessing that the "regime bounds" are the same thing as the upper limits of summation in equations 2-5? Is the "regime limit" the lengths of the [i,j,k, or l] bins in equations 2-5 or something else? I also think the wording of "each percentile," which in general usage implies 1 of 100 equal groups in a dataset, may also be incorrect because it does not seem consistent with the rest of the sentence and equations 2-5. Did the authors mean bins instead of percentiles? If so, what are these bins, specifically, how were they chosen, and how does the choice of spacing affect the results? I was also confused as to why one would want to group the same number of observations within each EIS or RH

percentile [or bin?], if the results are going to be weighted later?
Section 2.4.1 Environmental Regimes has been edited for clarity. We have also added "For example, with 100 environmental regimes, the observations will be binned from by 10 percentile limits of both EIS and RH. Within each row of RH within the regime framework, there are the same number observations as within each column of EIS; however within each individual regime of both EIS and RH, the number of observations is dependent on the distribution of both EIS and RH."

p. 7, l. 19: Did the authors mean "...*warm, single-layer, marine cloud SW*..."? It doesn't appear that they looked at terrestrial clouds, and they stated that they excluded multi-layer cloud cases.
We have changed it to "single-layer, marine warm cloud" in multiple places throughout the text to clarify and remind the reader the results are for a subset of clouds only.

Fig. 1: Please describe in the Figure caption what the red lines and blue dots represent (blue dots are presumably l, but it is best to be completely clear). In the caption, l is referenced. To avoid confusion, please state which l is being referenced (so far l0, lLWP, lENV, lBOTH, and lALL have been defined, but no l without a subscript). Please also specify which l is being discussed in the rest of the paper as well as the symbol is used frequently. Please clarify that the R2 value in Fig. 1 is describing the blue points and not the underlying distributions of all the data, because the largely overlapping red bars would suggest that in fact the correlation of the more raw data before that averaging happens is much smaller. Attaching a p-value to this and other similar figures appearing later in the paper seems appropriate.
All figure captions have been edited for clarity. In figure 1, we have added "with the red lines representing the standard deviation within each bin of ln(AI) and the blue dots representing the mean SW CRE for each bin." which also addresses how the red lines were calculated.
All lambdas have been subscripted with the correct identifier (0, LWP, ENV, BOTH, ALL).

Figure 2a: What cloud states are included here, and how were they derived and chosen? Some explanatory information is provided in section 3.2, but only after this Figure is referenced, which makes things confusing for the reader. It would be best if the figure could be a standalone item without requiring substantial reference to the text. The y-axis label for Fig. 2a seems to be missing, and only the units are provided. The text explaining Fig. 2a is not explicitly identified in the caption.

We have added to 2.4: "While the environmental regimes are established on a percentile basis, cloud state regimes are set by having an increasing number of bins for the lowest LWP clouds and a bin always set at 150 g/m2 to have a defined boundary between clouds which are extremely unlikely to precipitate (<150 g/m2) and clouds more likely to precipitate (>150 g/m2)."
We have added a better label to the y-axis of figure 2.
We have added more description to the caption of figure 2.

p. 7, l. 29: It might be helpful to reference which equation was used to derive the -13.12 value.
We have added where -13.12 came from.

p. 8, l. 4: "*Constraints on LWP limit these influences.*" This is already a well-known work that has previously established this finding.
You are correct. We have added a citation to work by Feingold on LWP constraints. Further citations are mentioned in the discussion as well already.

p. 8, l. 6: why were 3,7,11, and 23 divisions chosen?
We have clarified in section 3.2

> *"We will be using seven cloud states throughout our global analysis as it appears to capture the impacts LWP has on the sensitivity while allowing ample sampling for further division of observations throughout environmental regimes. The number of cloud states are steadily increased from 3 to 7 to 11 to 23 because those follow a progressive increase in the number of bin limits from 4 to 8 to 12 to 24 limits, respectively."*

Section 3.2: The term "cloud state" is commonly used throughout the paper, and it is the focus on section 3.2. However, cloud state is not explicitly defined in the paper, as far as I can tell, and this is very confusing for the reader. The data in section 3.2 mostly revolve around clouds binned by LWP. Is it possible to just use LWP, instead of "cloud state"? Another minor suggestion: the authors might consider changing "cloud regimes" to "LWP bins" (if this is correct). That would be a lot easier for a casual reader of the paper to understand.
We have changed Cloud Regimes (2.4) to Cloud States and added "Cloud states are defined as a range of liquid water paths, such that the liquid water path is held ostensibly constant."

Fig. 3a: Where is the caption text describing Figure 3a? Please clarify where the -11 value in the Fig. 3 caption comes from in relationship to these figures.

Is it based on the weighted mean of the data in Fig. 3a? Where is the label for the z-axis in Fig. 3a?

Added to caption: "When weighted and summed following equation (3), λ_{ENV} is 11.Wm−2ln(AI)."

Also added to end of caption: "...where the red lines represent the standard deviation of the SW CRE within each ln(AI) bin and the blue dots represent the mean SW CRE for each ln(AI) bin**"**

Figs. 3b and 3c: Please state in the caption how moist and dry environments are defined. Are these figures examples of data within individual grid cells from Fig. 3a? If so, please state that. The red bars seem to suggest that there may be no significant differences between any of the ln(AI) values within Fig. 3b or Fig. 3c, including at very high ln(AI) values and very low ln(AI) values?

Added to figure 3 caption: "unstable (~1K), dry environment (< 10% RH)(b) and stable (~6K), moist environment (>30% RH)"

The red bars are the standard deviation within each ln(AI) bin, while the blue dots are the mean warm CRE for each ln(AI) bin, as now explained in the caption. The difference between high low ln(AI) environments is focused on the mean not deviation. There is ~20 W/m2 difference in the dry, unstable case between the high and low and ~ 35 W/m2 difference in the moist, stable case. The differences are significant enough to have slopes of 10 and -25 W/m2ln(AI) for each case respectively.

p.10, last line: "*To account for the local meteorology, warm clouds are separated into 100 environmental regimes*..." This method seems to closely parallel the methodology of previous work (e.g., Chen et al. (2014)). It would be appropriate for the authors to reference such work here.

You are correct. We have added a citation to this work here. "This approach is similar to other approaches taken to estimate the indirect effect such as by Chen et al. 2014." Chen et al. (2014) is also currently cited in both the introduction and discussion sections.

Section 3.3: Since some of the cells in the figure probably have much greater sample sizes in the natural environment than others, to me, the weighted mean is probably more meaningful than the findings of individual grids, and I think it would be appropriate to stress this more in the paper.

We have added to section 3.3: "*The results focus on contrasting individual regimes, while the discussion focuses on contrasting constraints and the weighted, summed sensitivities.***"**

Our discussion section focuses on contrasting the weighted, summed values while the results focuses on how the methodology can identify regime specific responses.

p. 11, l. 3: "*The highest sensitivity is observed in stable regimes (EIS > 5.0) with a moderately dry free atmosphere*." And p. 11, l. 8: "*Above 1 K, λ increases with increasing RH, while in less stable environments, RH plays only a secondary role in modulating the sensitivity*." In Fig. 3a, I don't see evidence so far of there being higher sensitivity in drier environments, or of the latter statement at all. Were Figs. 3b,c supposed to be referenced here? Please provide more information to substantiate these statements.
To highlight the differences in section 3.3 we have added "The less stable regimes in figure 3 exhibit almost no variation in unstable regimes, varying by only ~1 W/m2ln(AI) while more stable regimes can vary by >10 W/m2ln(AI)."

Fig. 4: In the caption, sensitivity of what? Again, here, I think it would be really useful to note which of the grid cells are significant. Sample number in each grid cell will go down as resolution increases, and that would presumably impact the weighted mean values presented and discussed with respect to this figure, so significance would be a useful metric to help evaluate these results.
We have changed the caption beginning to "The sensitivity of the warm cloud CRE to aerosol found using equation 3 for environmental frameworks of…"

Fig. 5: This figure seems very likely to have an error. I do not see how the clouds at two extreme EIS and RH values can have the highest frequency of occurrence. If the x- and y-axis ranges were selected appropriately, one would expect the points approximately in the middle to be most frequent, and the points at the edges to be least frequent. Also, why is there such a strong mirror-like diagonal pattern in the plot? Natural data rarely show such a distinct pattern unless the x and y variables are highly related to each other. Please check that the data plotted here are correct.
We have added to section 3.3 to address the pattern:

> *"The mirror pattern is likely the result of the EIS in part having a slight dependence on RH, as the RH can alter the height of the 700 mb level needed to calculate EIS. This does not impact results as this dependence is accounted for by environmental regimes."*

And

> *"The moistest, most unstable and the driest, stablest environmental regimes always have the largest number of observations. The moist, unstable regimes are likely comprised of trade cumulus or other pre-convective cloud types in unstable regions like the ITCZ. The dry, stable regimes are likely comprised of marine stratocumulus cloud decks off the coast of west coast of continents with large scale subsidence drying the free atmosphere above."*

p. 14, l. 3: "*Overall, the largest ln(AI) sensitivity is seen in stable, dry environments (Figure 6h)*." I don't see this shown in that figure.

We have added to section 3.4 "These environments are ~ 7K of stability and ~ 30% RH." to pinpoint the signal.

p. 16, l. 5: "*In the absence of constraints (top), λ exhibits larger variations in magnitude and sign than when cloud, environmental, or cloud and environmental constraints are in place (panels b and c and Figure 9).*" Was Fig. 8 supposed to be referred to here? I don't see a panel b and c in Fig. 9, but these trends are not evident in Fig. 8....

We have added references to appropriate figures in 3.5
And also "The unconstrained map (Figure 8 a) varies from -.53 to .77 compared the most constrained map where the sensitivity of warm cloud CRE to aerosol varies only from - .11 to .46."

p. 18, l. 1: It would be useful to also mention earlier on (e.g., methods?) that there were 1.8 million observations in the study.

You are correct and we should mention this earlier. We have added to the methods "Even with these starting constraints on LWP and height, there were 1.8 million satellite observations fitting these parameters within the time period."

References

Ackerman, S., Platnick, S., Bhartia, P., Duncan, B., L'Ecuyer, T., Heidinger, A., Skofronick-Jackson, G., Loeb, N., Schmit, T., and Smith, N.: Satellites see the World's Atmosphere, Meteorological Monographs, 2018.

Karlsson, J., Svensson, G., Cardoso, S., Teixeira, J., and Paradise, S.: Subtropical cloud-regime transitions: Boundary layer depth and cloud-top height evolution in models and observations, Journal of Applied Meteorology and Climatology, 49, 1845–1858, 2010.

[..*]Quantifying Variations in Shortwave Aerosol-Warm Cloud-Radiation Interactions Using Local Meteorology and Cloud State Constraints

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Abstract. While many studies have tried to quantify the sign and the magnitude of the warm marine cloud response to aerosol loading, both remain uncertain owing to the multitude of factors that modulate microphysical and thermodynamic processes within the cloud. Constraining aerosol-cloud interactions using the local meteorology and cloud liquid water may offer a way to account for covarying influences, potentially increasing our confidence in observational estimates of warm cloud indirect effects. Four years of collocated satellite observations from the NASA A-Train constellation, combined with reanalysis from MERRA-2, are used to partition marine warm clouds into regimes based on stability, the free atmospheric relative humidity, and liquid water path. Organizing the sizable number of satellite observations into regimes is shown to minimize the covariance between the environment or liquid water path and the indirect effect. Controlling for local meteorology and cloud state mitigates artificial signals and reveals substantial variance in both the sign and magnitude of the cloud radiative response, including regions where clouds become systematically darker with increased aerosol concentration in dry, unstable environments. [..[2]]A darkening effect is evident even under the most stringent of constraints[..[3]]. These results suggest it is not meaningful to report a single global sensitivity of cloud radiative effect to aerosol. To the contrary, we find the sensitivity can range from -.46 to .11 [..[4]]$\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$ regionally.

1 Introduction

Warm clouds play an important role in Earth's radiative balance[..[5]]. Cooling the atmosphere and [..[6]]covering 25% of the Earth's surface on average and reflecting incoming shortwave radiation[..[7]], any changes to their radiative properties should be well quantified and understood (Hahn and Warren, 2007). These clouds are most prevalent off the western coasts of continents as marine stratocumulus, near the tropics as trade cumulus, and in the storm track regions as stratus (Ackerman et al., 2018). Perturbations in aerosol, whether from natural sources like sea spray or anthropogenic activities

*removed: Understanding Aerosol-Cloud-Radiation

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like [..[8]]biomass burning, lead to cloud-aerosol interactions that alter cloud radiative properties through two main effects, the albedo and the cloud lifetime effects. First [..[9]]termed by Twomey in 1977[..[10]], the albedo effect, or the first indirect effect as it's also known, suggests that clouds will become brighter as a result of aerosol loading. For a fixed liquid water path, increased aerosol within a cloud increases the number of cloud condensation nuclei (CCN), forcing the mean drop size to decrease[..[11]], resulting in a brighter, more reflective cloud. The second indirect effect, or the cloud lifetime effect, proposed by Alrecht (1989) builds on this idea, noting that a decrease in mean drop size due to aerosol-cloud interactions may also delay the onset of collision coalescence, suppressing precipitation and, in turn, allowing the cloud to survive longer, grow larger, and ultimately reflect more shortwave radiation. [..[12]]Early estimates of the indirect effect estimated including the cloud lifetime effect may increase [..[13]]it by 1.25[..[14]]x (Penner et al., 2001). Work since then has concentrated on decreasing the range of uncertainty rather than separating the effects in observation based studies, as without explicit constraints in place on the cloud water, the two effects are intrinsically related through the liquid water content of the cloud (Mülmenstädt and Feingold, 2018).

However, observing the indirect effect is not as straight forward as looking out your window trying to spot brighter clouds. The magnitude and sign of the indirect effect is extremely sensitive to the method used to quantify it. As a result, the Intergovernmental Panel on Climate Change (IPCC) has low confidence in the current estimate of the global aerosol indirect effect (AIE) (Boucher et al., 2013). An accurate assessment of the total indirect effect will reduce error in climate sensitivity and further our understanding of the role of clouds in future climates (Bony and Dufresne, 2005).

Historically, methods of estimating the AIE employ a single linear regression of either the cloud's radiative effect or droplet radius against a proxy for aerosol concentration [..[15]](Platnick and Twomey, 1994; Lohmann and Feichter, 2005; Christensen et al., 2016). This method ignores all possible covariances between the cloud, aerosol, and any processes that may effect both and assumes one linear regression captures all effects, disregarding the [..[16]]role played by the local environment [..[17]]as a strong modulator of warm cloud properties and responses (Stevens and Feingold, 2009). Constraining the [..[18]]local meteorology, or the characteristics of the environment around the cloud, as well as cloud type can significantly alter the magnitude of the AIE compared to single, unconstrained global linear regression [..[19]](Gryspeerdt et al., 2014). Regional analyses[..[20]], such as treating the marine stratocumulus cloud decks off the west coasts as a homogeneous sample,

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instead capture assorted responses and magnitudes [..[21]]as they fail to extricate covariance with [..[22]]local meteorology (Bender et al., 2016). Observationally-based estimates simply cannot “turn off” the effects of entrainment or other environmental effects like a model, therefore [..[23]]observation based approaches must prescribe a way to diminish the effect of these influences on cloud radiative effects, even at a regional scale.

Modeling provides one pathway for estimating the global AIE that explicitly accounts for local meteorological conditions, however low clouds are one of the largest sources of error in current global climate models (GCM) (Williams and Webb, 2009). In particular, GCMs tend to overestimate liquid water path (LWP) in low clouds, which leads to an overestimation of the albedo (Nam et al., 2012). The artificially elevated LWP impacts the sensitivity to aerosol by assessing it under unrealistic conditions[..[24]]. Further, entrainment and precipitation are artificially dampened as a result of incorrect cloud parameterizations [..[25]]in GCMs (Tsushima et al., 2016; Lee et al., 2009). Many cloud-aerosol processes are explicitly resolved in large eddy simulation (LES) models, but these are limited to small scales. LES can prescribe exact environments, but again these are limited to idealized meteorologies, only realistic to small regions on Earth. The microphysical processes of aerosol activation, nucleation, and eventual raindrop formation [..[26]]can only be parameterized in current GCMs and will remain so for the foreseeable future. The resolution is too coarse to emulate all scales of aerosol-cloud interactions hence the dependence on parameterizations and large uncertainty in model-derived estimates (Wood et al., 2016). [..[27]]A solution to this problem is a combination of global climate modeling guided by observation-based analysis and coordinated LES modeling to understand and quantify the AIE (Stephens, 2005).

Observation-based methods must [..[28]]avoid the pitfalls of historical evaluations and define a clear methodology to limit covariance with local environmental conditions or buffering by the cloud. Buffering is when the cloud state and/or environment work to reduce the impact of aerosols on the cloud Stevens and Feingold (2009). Cloud characteristics, such as LWP, and the local meteorology, like stability, can compound uncertainty in evaluating the AIE because [..[29]]they influence both radiative properties and susceptibility to aerosol (Lee et al., 2009; Feingold and Kreidenweis, 2002). The AIE is specifically defined as the cloud response to aerosol and the resulting effects on the radiative properties[..[30]]. Any quantification of the AIE must avoid including the effects of the local environment on the cloud radiative properties. When the local meteorology was accounted for, Gryspeerdt et al. (2016) found the sensitivity of cloud fraction to aerosol loading was reduced by 80%. Quantifying the AIE therefore requires separating and constraining all processes that moderate cloud radiative properties from those specifically due to aerosol-cloud radiation interactions (Stevens and Feingold, 2009). Organizing clouds into con-

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[25]removed: (Tsushima et al., 2016; Lee et al., 2009). Further, the
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strained, bounded spaces based on the external and internal covarying [..[31]]conditions can improve aerosol-cloud-radiation impact estimates (Ghan et al., 2016).

This study examines the sensitivity of the shortwave radiative forcing of warm clouds to aerosol [..[32]]by employing a methodology that attempts to adequately constrain external influences while maintaining sufficiently robust statistics. Our methodology takes advantage of the vast sampling provided by satellites to systematically hold environmental conditions and cloud state approximately constant. We quantify the warm cloud sensitivity to aerosol for clouds of similar properties within similar environments. While most satellite studies of aerosol-cloud interactions are by necessity correlative, the more covarying factors that are controlled (at the individual cloud level), the more closely we can approximate a causal relationship. Although we cannot confirm causation due to the temporal resolution of the observations, some studies have begun utilizing the high temporal resolution of geostationary satellites to augment A-Train observations and fix this ongoing problem (Sauter and L'Ecuyer, 2017). In our study, a set of environmental conditions and cloud state parameters is referred to as a regime. This idea of stratifying observations into regimes has been successfully implemented before to analyze cloud processes [..[33]](Williams and Webb, 2009; Chen et al., 2014; Gryspeerdt and Stier, 2012; Oreopoulos et al., 2016).

The environmental and cloud state regimes adapted here are designed to homogenize the clouds and processes occurring, reducing covariance [..[34]]the cloud radiative response to aerosol and other influences. Observationally-based, regime-dependent cloud processes have been discerned most often over large regional scales, however, divergent signals can be lost depending on the size of the region analyzed (Grandey and Stier, 2010). Even on small, local scales, variance in the meteorology alters the strength of the observed effects (Liu et al., 2016). A [..[35]]study using satellite observations with regime constraints, for example, found a definite relationship between the warm cloud AIE varies and atmospheric stability on a global scale (Chen et al., 2014).

One important meteorological influence is the stability of the boundary layer. LES of warm clouds have further shown that environmental instability can alter the effects of aerosol loading on warm clouds (Lee et al., 2012). The need to incorporate stability into AIE estimates has also been noted in prior observational studies (Sorooshian et al., 2009; L'Ecuyer et al., 2009; Su et al., 2010). Warm clouds in stable environments may show an increasing LWP with respect to aerosol loading while unstable environments may exhibit a decrease in LWP (Chen et al., 2014). Su et al. (2010) found the stability and rate of subsidence work to modulate aerosol-cloud-radiation interactions in warm clouds.

The effects of large scale subsidence and entrainment can be captured by the relative humidity (RH) in the free atmosphere[..[36]], known to exert a powerful influence on warm cloud characteristics (Wood and Bretherton, 2004). Entrainment of free atmospheric air [..[37]]furthers the decoupling process [..[38]]by increasing the temperature and humidity gradients at

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the cloud top (Lewellen and Lewellen, 2002). Including RH in aerosol sensitivity studies accounts for [..[39]]some decoupling influence. Models affirm the effects of entrainment on the cloud layer depend [..[40]]in part on RH, as LES have shown RH moderates cloud feedbacks in low warm cloud simulations (Van der Dussen et al., 2015) . Ackerman et al. (2004) and Bretherton et al. (2007) further demonstrated using an LES model that entrainment of dry air from the free atmosphere alters the distribution of liquid water within a cloud, which could modify [..[41]]the warm cloud response to aerosol.

In his original work, Twomey postulated that cloud albedo ought to increase with aerosol provided LWP is held fixed[..[42]], after all, albedo is dependent on the [..[43]]optical depth and effective radius. The LWP has been shown to clearly control the second AIE via its influence on precipitation suppression [..[44]](L'Ecuyer et al., 2009; Sorooshian et al., 2009). Field campaign observations have [..[45]]noted this relationship as well. For example, the Atmospheric Radiation Measurement Mobile Facility Azores campaign fond the cloud radiative response depended largely on the LWP (Liu et al., 2016). LWP is intrinsically tied to the magnitude of the AIE . Failing to distinguish [..[46]]clouds by LWP will lead to large covariance and/or buffering in the system by the LWP.

For these reasons, we adopt the boundary layer stability and relative humidity of the free atmosphere [..[47]]in conjunction with LWP to segment observations into regimes at the [..[48]]individual satellite pixel scale. To illustrate the impact of these specific buffering factors, we sequentially [..[49]]increase constraints on the regression of the warm cloud radiative effect against aerosol, what we refer to as the sensitivity or λ. First, the sensitivity is constrained by only LWP to demonstrate the importance of accounting for cloud state alone when estimating aerosol response. Next, environmental regimes of stability and relative humidity are used segment warm clouds and, within each regime, the sensitivity of the cloud radiative effect to aerosol is assessed. These [..[50]]environmentally regimented observations are then further separated into LWP regimes to control for cloud state and environment simultaneously. Finally, the warm cloud sensitivity with all regime constraints is derived on a regional basis to account for local influences not captured by the global regime partitions.

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2 Methods

2.1 Data

The effect of aerosol on marine warm cloud shortwave radiative properties is diagnosed from observations collected by the NASA A-Train constellation from 2007 to 2010. The A-Train is a series of synchronized satellites which allow for collocated observations from a variety of instruments (L'Ecuyer and Jiang, 2011). Environmental information is provided by collocated reanalysis data from the Modern-Era Retrospective analysis for Research and Applications Version 2 (MERRA-2). Collocated observations from multiple instruments, combined with high resolution reanalysis at the pixel scale, allows an [..[51]]extensive vie of the roles the environment and cloud state [..[52]]play in modulating the warm cloud sensitivity to aerosol concentration.

2.2 Cloud

The Cloud Profiling Radar (CPR) on CloudSat and the [..[53]]Cloud-Aerosol Lidar and Infrared Pathfinder Satellite (CALIPSO) are used to restrict analysis to [..[54]]single-layer, marine warm clouds between 60° N and 60° S. All data is interpolated down to CloudSat's $\sim$ 1km footprint. The CloudSat 2B-CldClass-Lidar product that classifies cloudy pixels based on their vertical structure from merged radar and lidar observations is leveraged to filter out ice phase and multilayered cloud systems (Sassen et al., 2008; Austin et al., 2009). All observations are restricted to below the freezing level of CloudSat which is determined using an ECWMF-AUX collocated reanalysis dataset and set where ECWMF determines the 0° isotherm. The Advanced Microwave Scanning Radiometer - Earth Observing System (AMSR-E) liquid water path (LWP) aboard the Aqua satellite is then used to limit observations to scenes where the LWP is above [..[55]].02 $\frac{\mathrm{kg}}{\mathrm{m}^2}$ and below .4 $\frac{\mathrm{kg}}{\mathrm{m}^2}$ (Wentz and Meissner, 2007). Very thin clouds below [..[56]].02 $\frac{\mathrm{kg}}{\mathrm{m}^2}$ are likely thin veil clouds with low albedos that are not the focus of this analysis (Wood et al., 2018). An [..[57]]along-satellite track cloud fraction is determined by finding the average number of warm cloud pixels that satisfy these criteria (seen by CloudSat or CALIPSO, below the CloudSat determined freezing level, and LWP [..[58]]between .02 and .4 $\frac{\mathrm{kg}}{\mathrm{m}^2}$) over each 12 km segment of the CloudSat track on a pixel by pixel basis, a scale that represents both the local scale length of the boundary layer and field-of-view used to define cloud radiative effects from Clouds and the Earth's Radiant Energy System (CERES) (Oke, 2002). Marine warm clouds fitting these parameters reside within the boundary layer. Even with these initial constraints on LWP and height, there were 1.8 million satellite observations fitting these parameters within the time period.

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[57] removed: along satellite
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The [..[59]]warm cloud shortwave radiative effect is found by combining this along track warm cloud fraction with top of atmosphere (TOA) radiative fluxes from CERES. CERES has a total (.4 - 200 μm) and shortwave channel (0.4 - 4.5 μm) that allow outgoing shortwave and longwave fluxes at the top of the atmosphere to be estimated using appropriate bi-directional reflectance models. [..[60]]All-sky radiances from CERES are not restricted to any type of scene and include the raw radiances observed by CERES. The shortwave warm cloud radiative effect (CRE) is then defined in terms of the all sky and inferred clear sky forcings from CERES and warm cloud fraction from CloudSat. The clear sky flux ($F^{\uparrow}_{\text{Clear Sky}}$) is a regional, monthly mean estimate of cloud free outgoing shortwave radiation. Writing the all-sky net SW radiation at the top of the atmosphere as:

[..[61]]

$$(F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{All Sky}} = (F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{Clear Sky}} \times (1 - CF) + (F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{Cloudy}} \times CF \quad (1)$$

It is easy to show that for shortwave radiances:

[..[62]]

$$F^{\uparrow}_{\text{All Sky}} - F^{\uparrow}_{\text{Clear Sky}} \times (1 - CF) = CRE \quad (2)$$

where the warm $CRE_{SW} = CF \times F^{\uparrow}_{\text{Cloudy}}$

The [..[63]]instantaneous CRE for each warm cloud observation is used in conjunction with aerosol information and corresponding instantaneous cloud state and meteorological state constraints to derive the sensitivity of the cloud radiative effect to aerosol loading.

2.3 Aerosol

Aerosol index (AI) is used to characterize the concentration of aerosol in the atmosphere. AI is the product of the Angstrom exponent [..[64]](found using AOD at 550 and 870 nm) and AOD at 550 nm, both of which are derived from the Moderate-Resolution Imaging Spectroradiometer (MODIS) aboard the Aqua satellite. The Angstrom exponent, a measure of the turbidity of the atmosphere, is derived from multiple estimates of aerosol optical depth (AOD) [..[65]](Ångström, 1929; Remer et al., 2005). The MODIS Angstrom exponent provides information about the size of the observed aerosol as well as concentration (Levy et al., 2010). MODIS AI is derived from the auxiliary dataset (MOD06-1km-AUX) developed from the overlap of the CloudSat CPR footprint and the MODIS cloud mask at pixel level.Although AI is not a direct measurement of CCN in the air,

[59] removed: cloud

[60] removed: The shortwave

[61] removed: $(F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{All Sky}} = (F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{Clear Sky}} \times (1 - CF) + (F^{\downarrow}_{SW} - F^{\uparrow}_{SW})_{\text{Cloudy}} \times CF$

[62] removed: $F^{\uparrow}_{\text{All Sky}} - F^{\uparrow}_{\text{Clear Sky}} \times (1 - CF) = CRE$

[63] removed: clear sky flux ($F^{\uparrow}_{\text{Clear Sky}}$) is a regional, monthly mean estimate of cloud free outgoing shortwave radiation. The

[64] removed: and

[65] removed: at 550, 865, and 2100 nm

it has a higher correlation with CCN compared to the AOD and is therefore more suitable for aerosol-cloud interaction studies (Stier, 2016; Dagan et al., 2017). While AOD and the Angstrom exponent from MODIS are not available in cloud scenes, the collocated dataset interpolates these between clear sky scenes in order to infer a cloudy AI. For lower cloud fraction scenes, this interpolation is more accurate, however it is possible that in higher cloud fraction scenes, the accuracy of AI is reduced. This is a source of uncertainty within our results. Binning by relative humidity when evaluating the sensitivity should reduce some bias from aerosol swelling in humid environments.

2.4 Regimes

2.4.1 Environmental Regimes

MERRA-2 reanalyses collocated with each CloudSat footprint is used to define local thermodynamic conditions that distinguish environmental regimes. The environmental regimes employed here provide a crude representation of the local meteorology acting to inhibit or invigorate the cloud response. While these states, defined from percentile bins of the estimated inversion strength (EIS) and relative humidity at 700 mb (RH), do not capture the complete range of environmental factors that influence warm cloud development, they have been shown to provide fairly robust bulk classification for sorting satellite observations into meteorological regimes (Sorooshian et al., 2009; L'Ecuyer et al., 2009; Chen et al., 2014). Here, EIS is calculated using MERRA-2 temperature and relative humidity profiles and indicates the stability of the boundary layer. EIS incorporates effects of water vapor on the lower tropospheric static stability and is better correlated for all cloud types with cloud fraction.

From Wood and Bretheron (2006):

$$\mathrm{EIS} = \mathrm{LTS} - \Gamma_{\mathrm{m}}^{850}(\mathrm{z}_{700} - \mathrm{LCL}) \tag{3}$$

where Γ_m^{850} is the moist-adiabatic potential temperature gradient and LTS is the lower-tropospheric stability.

The relative humidity at 700 mb is used as a measure of the effect of entraining free tropospheric air (Karlsson et al., 2010). As the height of the 700 mb isobar is included in the equation for EIS, there is some covariability between EIS and RH. When referring to the effects of entrainment, it means the effects of RH. All observations within the 5% - 95% percentiles of both EIS and RH are partitioned into regimes of percentile limits. The bin limits depend on the number of bins implemented, which is varied in the results to establish the degree to which the environment must be constrained to accurately characterize sensitivity. For example, with 100 environmental regimes, the observations will be binned from by 10 percentile limits of both EIS and RH. Within each row of RH of the environmental regimes, there are the same number observations as within each column of EIS, however, within each individual environmental regime of both EIS and RH, the number of observations is dependent on the distribution of both EIS and RH.

2.4.2 Cloud States

Cloud states are defined by the LWP. Although there are other definitions of cloud regimes and cloud states used in other studies (e.g. Oreopoulos et al. (2017)), throughout ours cloud state or cloud morphology refers to the set of observations binned by liquid water path. While the stability and entrainment directly affect the LWP, we consider the LWP separately from the local meteorology as it represents the cloud thermodynamics more than the local environmental conditions. AMSR-E liquid water path, derived from the 19, 23, and 37 GHz channels, is used to separate observations into cloud state regimes (Wentz and Meissner, 2007). AMSR-E LWP is most accurate for low, marine warm clouds (Greenwald et al., 2007; Juárez et al., 2009). 99% of observations fell below a LWP of .4 $\frac{\mathrm{kg}}{\mathrm{m}^2}$ and analysis was restricted to observations with LWP below this limit. Since CRE is proportional to the optical depth of a cloud, which is directly related to the LWP, the sensitivity has a strong covariance with LWP (Stephens, 1978; Lee et al., 2009; Wood, 2012). Holding LWP effectively constant is therefore essential to estimating the AIE (Lohmann and Lesins, 2002). The number of LWP bins decreases from global to regional analysis due to sampling; on a global scale, seven LWP regimes are used, while on a regional scale, only four LWP regimes are used. Limits are placed to separate out the signals of low LWP clouds vs. high LWP clouds, as low clouds may be affected by evaporation-entrainment feedbacks while high LWP clouds may be affected by precipitation (Jiang et al., 2006; L'Ecuyer et al., 2009). While the environmental regimes are established on a percentile basis, cloud state regimes are set by having an increased number of bins for the lowest LWP clouds and a bin limit always set at .15 $\frac{kg}{m^2}$ to delineate clouds which are extremely unlikely to precipitate ($<$ .15 $\frac{kg}{m^2}$) and clouds more likely to precipitate ($>$ .15 $\frac{kg}{m^2}$) (L'Ecuyer et al., 2009). When environmental regimes are combined with cloud state constraints, the environmental regime limits remain constant throughout all cloud state regimes. The difference in the sensitivity of the warm cloud radiative effect to aerosol in one environmental regime versus another environmental regime at a constant LWP can therefore be more accurately attributed to aerosol.

2.5 Sensitivity

The [..[66]]warm cloud radiative sensitivity to aerosol, or λ, is defined [..[67]]as the linear regression of the shortwave CRE against ln(AI). While other studies have called similar metrics a susceptibility, we use the term sensitivity. The natural log of AI is used to better represent the effects of the smallest particles, which are more likely to act as CCN within a cloud (Köhler, 1936). The sensitivity is evaluated within environmental and cloud state regime frameworks on both global and regional scales. [..[68]]The observations are binned by 15 percentile bins of ln(AI). The AI bins are defined by the set of observations being regressed. The sensitivity is only calculated if there are 100 observations within the regime and the linear regression Pearson correlation coefficient is greater than .4. Throughout the study, although environmental and cloud state impacts are constrained through regimes, it cannot be stated with certainty that the observed changes in CRE are due to aerosol, only correlated with aerosol.

[66]removed: cloud

[67]removed: here

[68]removed: Variation in λ between regimes may signal buffering by the environment or cloud state.

The unconstrained sensitivity, or the sensitivity of [..[69]]the warm cloud shortwave radiative effect to ln(AI) [..[70]]without limits on region, LWP, stability, or RH, is computed as:

$$\lambda_0 = -\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \tag{4}$$

The partial derivative in this equation [..[71]]implies influencing factors other than aerosols should be held fixed. Here this is accomplished by evaluating the sensitivity with increasing constraints on the partial differential through regimes.

To hold the cloud state fixed, the sensitivity is found for distinct [..[72]]seven LWP regimes (k) and summed to yield a mean sensitivity:

$$\lambda_{LWP} = \sum [..^{73}]^{\mathrm{N_{LWP}}}{}_{\mathrm{k=1}} \left(-\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \right)_k \mathrm{N}_k \tag{5}$$

Where $\mathrm{N_k}$ is the [..[74]]number of observations of cloud state k. In our results, we evaluate the efficacy of increasing and decreasing the number of cloud states.

Similarly, the sensitivity within environmental regimes, defined by the estimated inversion strength and relative humidity of the free atmosphere, can be computed, weighted, and summed to account for meteorological covariability with ten regimes of each EIS (i) and RH (j), where $\mathrm{N}_{i,j}$ is the number of observations within each environmental regime:

$$\lambda_{ENV} = [..^{75}] \sum_{\mathrm{j=1}}^{\mathrm{N_{RH}}} \sum [..^{76}]^{\mathrm{N_{EIS}}}{}_{\mathrm{i=1}} \left(-\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \right)_{i,j} \mathrm{N}_{i,j} \tag{6}$$

By extension, both cloud and environmental conditions can be controlled via:

$$\lambda_{BOTH} = [..^{77}] \sum_{\mathrm{k=1}}^{\mathrm{N_{LWP}}} \sum_{\mathrm{j=1}}^{\mathrm{N_{RH}}} \sum [..^{78}]^{\mathrm{N_{EIS}}}{}_{\mathrm{i=1}} \left(-\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \right)_{i,j,k} \mathrm{N}_{i,j,k} \tag{7}$$

Where $\mathrm{N}_{i,j,k}$ is the number of observations within each environmental regime when constrained further by each of the cloud state regimes k.

Finally, it is recognized that these bulk constraints do not fully capture all of the local factors that influence aerosol-cloud interactions. AI alone does not fully constrain the effect of aerosol composition which varies regionally. Thus, to control for

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[69] removed: cloud

[70] removed: in the absence of constraints

[71] removed: immediately

[72] removed: LWP regimes

[74] removed: fraction of clouds that fall into LWP

these unaccounted for local effects, the sensitivity is further constrained by finding Eqn (7) on a 15° by 15° scale [..[79]]with four cloud state regimes (k), five regimes of stability (i), and five regimes of RH (j) for each of the 152 regions (l).

[..[80]]

$$\lambda_{\mathrm{ALL}} = \sum_{l=1}^{N_{\mathrm{Reg}}} \sum_{k=1}^{N_{\mathrm{LWP}}} \sum_{j=1}^{N_{\mathrm{RH}}} \sum_{i=1}^{N_{\mathrm{EIS}}} \left(- \frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})} \right)_{i,j,k,l} N_{i,j,k,l} \tag{8}$$

2.6 [..[81]]

[..[82]]

2.6 [..[83]]

[..[84]]

Figures/unconstrained.png

Figure 1. The sensitivity of CRE to aerosol ([..[85]]λ_0 from equation (4)) found globally without constraints on the environment, cloud state, or region. The red lines represent the standard deviation within each bin of ln(AI) and the blue dots represent the mean SW CRE for each bin.

3 Results

3.1 Unconstrained Sensitivity

The global sensitivity of warm cloud SW forcing to aerosol without any constraints [..[86]]described by Equation (4) [..[87]]is -12.81 [..[88]]$\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$ (Figure 1). This seems to capture the warm cloud AIE, after all the shortwave CRE increases with

[79]removed: .

[81]removed: Environmental Regimes

[82]removed: MERRA-2 reanalyses collocated with each CloudSat footprint is used to define local thermodynamic conditions that distinguish environmental regimes. The environmental regimes employed here provide a crude representation of the effects of the local meteorology acting on the cloud to inhibit or invigorate aerosol-cloud-radiation interactions. While these states, defined from percentile bins of the estimated inversion strength (EIS)and relative humidity at 700 mb (RH), do not capture the complete range of environmental factors that influence warm cloud development, they have been shown to provide fairly robust bulk classification for sorting satellite observations into meteorological regimes (Sorooshian et al., 2009; L'Ecuyer et al., 2009; Chen et al., 2014). Here, EIS is calculated using MERRA-2 temperature and relative humidity profiles. EIS incorporates effects of water vapor on the lower tropospheric static stability and is better correlated for all cloud types with cloud fraction (Wood and Bretherton, 2006). The relative humidity at 700 mb is used as a measure of the effect of entraining free tropospheric air. All observations within the 5% - 95% percentiles of both EIS and RH are partitioned into regimes . Environmental regime

aerosol loading as expected. However, this unconstrained estimate ignores the roles of buffering and covariance. The indicated variation of SW CRE within each ln(AI) [..[89]]bin alludes to variation in the overall effect not captured by a single linear regression. Although the R^2 is high, without constraints [..[90]]the increase in shortwave CRE cannot be attributed to only aerosol. Furthermore, from this estimate, no information is made known on how the sensitivity varies regionally, how cloud processes affect the AIE, or whether particular cloud states may be influenced more strongly by aerosol than others.

3.2 Sensitivity to Cloud State

The original description of the albedo effect by Twomey (1977) specified holding the LWP of the cloud constant. Following Twomey's original hypothesis, when warm clouds are separated [..[91]]by LWP into cloud states, it is clear that cloud morphology plays a role in modulating the magnitude of the sensitivity (Figure 2). The total weighted, summed sensitivity is -13.12 [..[92]]$\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$ for seven cloud states. From Figure 2, the lowest cloud states are less sensitive to aerosol, with a steep increase at $\sim$.8 $\frac{kg}{m^2}$. The sensitivity increases with LWP, peaking for [..[93]]LWPs between .1 and .15 $\frac{kg}{m^2}$. Beyond .15 $\frac{kg}{m^2}$, the trend reverses and the sensitivity decreases with LWP, consistent with the fact that thicker clouds are already bright and less susceptible to aerosol-induced changes (Fan et al., 2016). The non-linear relationship along with the known covariance between LWP and the AIE make it a vital component of the regime framework proposed here (Feingold, 2003). Constraints on LWP limit these influences (Feingold, 2003).

The key to implementing appropriately stringent regime constraints is to determine the minimum number of cloud [..[94]]states required to adequately capture [..[95]]LWP modulation of the total sensitivity. [..[96]]We will be using seven cloud states throughout our global analysis as it appears to capture the impact cloud state exerts on the sensitivity while permitting ample sampling for further division of observations throughout environmental regimes. The number of cloud states are

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limits are defined such that there are the same number observations within each percentile of either EIS or RH . The regime bounds depend on the resolution used, which is varied to establish the degree to which environmental factors must be constrained to accurately characterize sensitivity.

[83]removed: Cloud Regimes

[84]removed: AMSR-E liquid water path, derived from the 19, 23, 37 GHz channels, is used to separate observations into cloud regimes (Wentz and Meissner, 2007). AMSR-E LWP is most accurate for low, marine warm clouds (Greenwald et al., 2007; Juárez et al., 2009). 99% of observations fell below a LWP of 400 $\frac{\mathrm{g}}{\mathrm{m}^2}$ and analysis was restricted to observations with LWP below this limit. Since CRE is proportional to the optical depth of a cloud, which is directly related to the LWP, the sensitivity has a strong covariance with LWP (Stephens, 1978; Lee et al., 2009; Wood, 2012). Holding LWP effectively constant is therefore essential to estimating the AIE (Lohmann and Lesins, 2002). The lowest LWP regimes have the least chance of precipitating; the sensitivities of these regimes are unlikely to be affected by secondary aerosol-cloud-precipitation effects.

[86]removed: ,

[87]removed: ,

[88]removed: $\frac{Wm^{-2}}{ln(AI)}$

[89]removed: (red bars)

[90]removed: ,

[91]removed: into LWP regimes

[92]removed: $\frac{Wm^{-2}}{ln(AI)}$. Low LWP clouds

[93]removed: cloud

[94]removed: regimes

[95]removed: this

[96]removed: Observations were divided into

steadily increased from 3 [..[97]]to 7 [..[98]]to 11 [..[99]]to 23 [..[100]]partitions to follow a progressive increase in the number of bin limits from 4 to 8 to 12 to 24 limits, respectively. Overall, [..[101]]λ_{LWP} exhibits a similar trend regardless of [..[102]]partitioning. The peak sensitivity for all [..[103]]cloud states is around .1 $\frac{kg}{m^2}$[..[104]]. The curve of the sensitivity [..[105]]and the behavior [..[106]]of thicker clouds is not well captured using only [..[107]]3 LWP bins. The use of [..[108]]7 cloud states, on the other hand, [..[109]]reproduces the behavior of thicker clouds and guarantees a large number of samples within each cloud [..[110]]state appropriate for a linear regression, especially when later partitioning by additional influences.

3.3 Sensitivity within Environmental Regimes

Even when separated into cloud states, aerosol impacts on warm clouds can be strongly modulated by the local environment. To account for the local meteorology, warm clouds are separated into 100 environmental regimes defined according to the local stability and free tropospheric humidity at the time they were observed (Figure 3). This approach is similar to that employed by Chen et al. (2014). Within each EIS and RH regime, CERES shortwave CRE is linearly regressed against ln(AI). [..[111]]The processes and resulting response are modified by the local meteorology, indicated by the change in sensitivity for different environmental regimes. Unstable environments exhibit almost no variation in sensitivity, varying by only $\sim$1 $\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$, while stable regimes can vary by >10 ($\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$). The moisture content of free atmosphere [..[112]]influences the sensitivities in stable regimes more than unstable regimes with a clear divide at EIS = 1 K. [..[113]]The highest sensitivity is observed in stable regimes (EIS $>$ 5.0K) with a moderately dry free atmosphere (Figure 3). The most sensitive warm clouds reside in environments with a moderately dry relative humidity of around 27% for an extended range of stabilities from 5 to 10 K. Warming effects (positive sensitives) [..[114]]are observed in [..[115]]unstable, dry environments. A warming, or reverse

[97] removed: ,

[98] removed: ,

[99] removed: , and

[100] removed: cloud regimes based on LWP

[101] removed: λ

[102] removed: resolution (the number of regimes used)

[103] removed: partitions

[104] removed: , but using 11 or 23 regimes demonstrates the peak is actually closer to .15 $\frac{kg}{m^2}$. The amplitude

[105] removed: variation with LWP

[106] removed: at larger LWP is is

[107] removed: 4

[108] removed: 8 cloud regimes

[109] removed: generally reproduces the shape of the higher resolution bins but

[110] removed: regime

[111] removed: As the regime changes, the processes behind the cloud's response and resulting radiative sensitivity also change . The highest sensitivity is observed in stable regimes (EIS $>$ 5.0)with a moderately dry free atmosphere. In general, less stable regimes exhibit lower sensitivities for all RH. In the least stable and driest regimes, the sign of λ reverses, indicating that clouds become darker with increased aerosol loading, counter to the conventional view that polluted clouds become brighter and cool more effectively.

[112] removed: appears to influence

[113] removed: Above 1 K, λ increases with increasing RH, while in less stable environments, RH plays only a secondary role in modulating the sensitivity

[114] removed: as opposed to cooling effects,

[115] removed: low stability, low humidity

Twomey, effect has been noted to occur [..[116]]by others investigating the AIE (Chen et al., 2012, 2014). Consistent with these results, Christensen and Stephens (2011) found that up to 1/3 of ship-tracks, occurring in primarily [..[117]]unstable regions, are darker than their surroundings owing to their thermodynamic feedbacks. [..[118]]The weighted global sensitivity calculated using Equation (6) is [..[119]]-11. $\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$ when the influence of the environment is accounted for (Figure 3).

[..[120]]The number of partitions must be narrow enough to separate the various degrees of buffering by the local meteorology and yet allow an ample number of observations per environmental regime when calculating the constrained sensitivities. To determine an optimal resolution for this dataset, the distribution of observations and sensitivity are separated into 5, 10, and 15 EIS and RH partitions representing 25, 100, and 225 environmental states respectively (Figures 4, 5). The distribution of observations among environmental regimes varies smoothly with resolution (Figure 5). The minimum number of samples decreases from [..[121]]35,532 to 2,707 to 757 when the resolution increases from 25 regimes to 100 regimes to 225 regimes, respectively. The [..[122]]mirror pattern is likely the result of the EIS in part having a slight dependence on RH, as the RH can alter the height of the 700 mb level needed to calculate EIS. This does not impact results as this dependence is accounted for by environmental regimes. The moistest, most unstable and the [..[123]]driest, stablest environmental regimes always have the largest number of observations. Moist, unstable regimes are likely comprised of trade cumulus or other pre-convective cloud types in regions like the ITCZ. Dry, stable regimes are likely comprised of marine stratocumulus cloud decks off the west coast of continents.

The total sensitivity decreases as the resolution increases, from -11.29 to -11.04 to -10.99 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$ (Figure 4). The 5 by 5 framework degrades the smoothness in [..[124]]λ_{ENV} with respect to the different environmental states. The difference between the 10 by 10 and 15 by 15 estimates of sensitivity indicate that an increase in resolution after 10 partitions will lead to very little change in the overall sensitivity. However, an increased resolution decreases the number of clouds in all environmental regimes, which will be vital when the environmental regimes are further distributed among cloud states. The use of 100 regimes in analysis is appropriate to ensure proper distribution among all cloud states.

3.4 Accounting for Cloud and Environmental States

The preceding sections clearly demonstrate the importance of controlling for meteorological and cloud state dependencies when evaluating the sensitivity of cloud radiative effects to aerosol[..[125]], however it is time to revise our framework to include both sets of constraints. Here we define three-dimensional regimes that hold LWP approximately constant while also

[116] removed: in some regimes

[117] removed: low stability

[118] removed: Accounting for this strong environmental modulation of sensitivity, the

[119] removed: -11.04 $\frac{Wm^{-2}}{ln(AI)}$

[120] removed: Again, for the partial derivative in Eqn (6) to be applied correctly, the

[121] removed: 35532 to 2707

[122] removed: driest,

[123] removed: moistest

[124] removed: λ

[125] removed: . To account for the covarying impacts of cloud state, the environment, and aerosols on cloud radiative effects, we must simultaneously control for each of these factors

constraining the local meteorology (Figure 6). The sensitives estimated for each of the 700 resulting regimes are shown in Figure 6. The lowest LWP cloud [..[126]]states show a comparatively damped maximum sensitivity than the thicker cloud [..[127]]states. Higher LWP clouds exhibit an increasing maximum [..[128]]λ_{BOTH}. The variation in magnitude between cloud [..[129]]states within the same environmental regimes confirms that LWP exerts a strong control in modulating the magnitude of the response and must be held constant when estimating the AIE. Mixing different cloud states in Figure 3 likely conflates differing signals, inaccurately representing the sensitivity in the most populous environmental regimes.

Again, the constrained sensitivities show distinct evidence of [..[130]]a darkening effect where thin clouds in the driest, most unstable environments exhibit a warming, or darkening, response to aerosol loading. Within the environmental regimes that exhibit a darkening effect, the magnitude is strongly modulated by LWP, suggesting both the expected (cooling) and opposite (warming) responses depend on LWP, RH, and EIS. As LWP increases, a warming [..[131]]λ_{BOTH} favors increasingly moist, stable environments. [..[132]]

The summed and weighted sensitivity with constraints on both LWP and meteorology is -10.6 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$. Overall, the largest sensitivity is seen in stable, moderately dry environments (Figure 6h). [..[133]]These environments are $\sim$ 7K of stability and $\sim$ 30% RH independent of LWP. Their large sensitivity is due in part to their prominence, as most marine stratocumulus cloud decks occur in stable environments with a dry free troposphere[..[134]]. The weakest sensitivity occurs in unstable, dry regimes and stable, moist regimes. While these environmental conditions and cloud states are less common, [..[135]]discerning global warming signal with stringent constraints is significant. [..[136]]

These results also suggest that AIE is overestimated in approaches that do not hold the LWP approximately constant. When summed and weighted by frequency of occurrence, over almost all environmental regimes, constraining LWP damps [..[137]]the sensitivity (Figure 6). The difference between the [..[138]]LWP constrained and only environmentally constrained sensitivities reveals the strong dependence of cloud response on stability, RH, and LWP. In very few unstable environments, LWP constraints act to amplify the response. This effect is only observed in the the most moist and unstable or dry, stable states that have a high density of observations. LWP constrains in these regimes pulls out otherwise obstructed or buffered signals.

[126]removed: regimes
[127]removed: regimes
[128]removed: λ
[129]removed: regimes
[130]removed: an inverse Twomey
[131]removed: λ
[132]removed: The maximum sensitivity is always observed in stable environments, but thin clouds, or low LWP cloud regimes, are more responsive to dry or moist free atmospheres. In thinner clouds, especially, aerosol loading will increase the number of CCN in the cloud leading to rapid invigoration (Christensen and Stephens, 2011). This suggests that RH may play a more pronounced role in modulating aerosol effects in thinner clouds.
[133]removed: This
[134]removed: from an almost continual overlying high pressure system. The smallest
[135]removed: the fact that on a global scale with constraints on cloud state, some regimes show no sensitivity or a reverse Twomey effect
[136]removed: Even with the most stringent constraints, on a global scale the reverse Twomey effect is discerned.
[137]removed: λ
[138]removed: cloud regime

To assess the effect of the resolution used to define environmental states when LWP constraints are added Figure 6h is replicated using 25, 100, and 225 environmental states (Figure 7). Sensitivity estimates are less varied [..[139]](relative to Figure 3) when both the local meteorology and LWP are constrained , indicating that holding LWP fixed is essential regardless of [..[140]]the number of partitions of EIS and RH. The inclusion of [..[141]]LWP, however, places increasingly restrictive demands on sampling volumes since each environmental regime must be sufficiently populated enough to allow robust sensitivities to be derived within a majority of cloud [..[142]]state partitions.

3.5 Sensitivity on Regional Scales

None of the results presented thus far have considered [..[143]]regional scale variability. To account for local processes and systematic differences in aerosol (e.g. composition, size, source) not captured by the bulk, global metrics above, the cloud state and environmental regime framework is applied to [..[144]]15° grid boxes [..[145]]from 60°S to 60°N. Regional variations in cloud sensitivity with [..[146]]a varying number of constraints on local meteorology and cloud state are shown in Figures 8 and 9. In the absence of constraints (Figure 8 top), [..[147]]the sensitivity exhibits larger variations in magnitude and sign than when cloud, environmental, or cloud and environmental constraints are in place (panels b and c and Figure 9). The unconstrained map (Figure 8 a) varies from -.53 to .77 $\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$ compared the most constrained map where the sensitivity of warm cloud CRE to aerosol varies only from - .11 to .46 $\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$. In fact, without controlling for covarying influences of stability, entrainment, and cloud morphology, vast regions of predominantly trade cumulus clouds exhibit [..[148]]a darkening that reduce the globally integrated warm cloud AIE.

With constraints on only cloud state, the sensitivity shows greater variation in magnitude and sign than any other case (8 b). The tropics show an extreme darkening signal, much greater than the unconstrained case. The darkening likely occurs in the lowest, thinnest cloud state regimes and may be due to evaporation. The maximum cooling sensitivity occurs in the southern oceans at a much larger magnitude than the unconstrained case. These signals are likely inflated since covarying meteorological factors are not fully constrained. While limiting the effects of cloud morphology on buffering and covariance is necessary, it is not sufficient for accurately resolving global AIE.

When constrained by local meteorological conditions alone (Figure 8 c), [..[149]]the sensitivity is damped in all regions. The southern ocean no longer dominates the global AIE, instead the maximum effect is seen in the north Atlantic. The warming

[139]removed: between the three resolutions when LWP is constrained
[140]removed: environmental regime resolution
[141]removed: cloud regimes
[142]removed: regimes
[143]removed: variability on the local scale
[144]removed: all
[145]removed: globally
[146]removed: varying degrees of constraint
[147]removed: λ
[148]removed: an inverse AIE
[149]removed: λ

sensitivities, or darkening, that were prevalent in the equatorial region are significantly decreased, replaced by large regions of no [..[150]]

[..[151]]sensitivity. Clouds can be distributed among different LWP regimes, with differing sensitivities, that cumulatively cancel each other out even in similar environmental conditions. [..[152]]The environmental framework only controls for meteorological covariability, but cloud state plays a large role in modulating the sign and magnitude of effect. [..[153]]

The inclusion of cloud state through LWP into the regime framework is vital to adhere to the original theories of Twomey (1977) and Albrecht (1989). Both assumed the LWP to be held constant, however this cannot be true of observation based estimates of the AIE unless the LWP is explicitly limited to be approximately constant. As seen in Figure 8b, limits on LWP alone are not stringent enough to elucidate the true AIE and tend to artificially enhance sensitivities. The buffering effects of the environment and local modulating factors must also be accounted for.

[..[155]]Including both cloud and environmental regimes limits the co-variance between aerosol, stability, cloud state, and [..[156]]entrainment on cloud radiative properties (Figure 9). This likely captures the true regional variation in the response of CRE to aerosol more accurately than any of the other regional estimates. [..[157]]The areas of strongest and weakest sensitivities exhibit coherent patterns that tends to align with distinct cloud and aerosol types. The largest [..[158]]sensitivities are observed in the southern subtropical oceans[..[159]]. Warm clouds off the coast of California exhibit a larger sensitivity with minimal constraints, i.e. with only cloud state or environmental constraints. The equatorial region shows a slight [..[160]]warming to no effect. This is likely the region contributing to the [..[161]]darkening seen in the global regime framework for unstable, dry regions (Figure 6 h). The resulting global weighted mean [..[162]]sensitivity derived from Eqn (8) is likely [..[163]]representative of the complete spectrum of global [..[164]]shortwave warm cloud responses to aerosol.

[..[165]]

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[150]removed: λ, likely the result of the counteracting effect of increased entrainment and reduced particle size (Small et al., 2009).

[151]removed: However, clouds may

[152]removed: As demonstrated above, the

[153]removed: The use of only environmental regimes within a region can be appropriate when the LWP distribution is narrow but cloud morphology becomes extremely important in regions with a diverse population and broad distribution.

[155]removed: The inclusion of

[156]removed: the free atmosphere's effects

[157]removed: When both cloud and environmental constraints are applied (Figure 9), the effect is no longer universally dampened, as was the case with only environmental regimes or enhanced as when controlling only for cloud state (Figures 8).

[158]removed: warm cloud

[159]removed: , especially in the southeast Pacific and southern Atlantic. The California coast has a larger signal than when only regionally and environmentally constrained

[160]removed: inverse

[161]removed: inverse Twomey effect

[162]removed: AIE

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[164]removed: warm cloud sensitivity to aerosolthan any estimates provided here or other global analyses that fail to account for local effects

[165]removed: The inclusion of cloud state through LWP into the regime framework is vital to adhere to the original theories of Twomey (1977) and Albrecht (1989). Both assumed the LWP to be held constant, however this cannot be true of observation based estimates of the AIE unless the LWP is explicitly limited to be approximately constant. As seen in Figure 8b, limits on LWP alone are not stringent enough to elucidate the true AIE and tend to artificially enhance sensitivities. The buffering effects of the environment and local modulating factors must also be accounted for.

Table 1. [..[166]]Warm cloud shortwave radiative sensitivity to aerosol estimates with varying degrees of constraints in [..[167]] $\frac{\mathrm{W\,m^{-2}}}{\ln(\mathrm{AI})}$.

No Constraints	(λ)	-12.81
Cloud State Constraints	(λ_{LWP})	-13.12
Environmental Constraints	(λ_{ENV})	-11.0
Cloud & Environmental Constraints	(λ_{BOTH})	-10.6
Cloud & Environmental Constraints Regionally	(λ_{ALL})	-10.13

4 Discussion

The sample regressions show in Figures 1, 2, and 3 [..[168]]illustrate the ability of constraints to reduce the variance of the observations. These constraints translate into a range of global [..[169]]sensitivity estimates. As constraints are applied, the sensitivity decreases from -12.81 to -10.6 to -10.13 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$. The decrease in total sensitivity reveals the need to constrain LWP. Holding only cloud state constant can exacerbate the signal due to mixed meteorologies, but the first order dependence of CRE on LWP requires it to be held constant. When these are applied regionally, local signals are preserved allowing the closest to truth estimate of -10.13 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$. This estimate is only possible through the power of sampling provided by 1.8 million satellite observations partitioned among 700 regimes, or 15,200 when further partitioned on a regional basis to represent local scale processes.

In theory, partial derivatives, such as $\frac{\partial \mathrm{CRE}}{\partial \ln(\mathrm{AI})}$, [..[170]]assumes other variables are [..[171]]held constant. The folly in treating warm clouds as only a function of aerosol is [..[172]]evident in Figure 8, where regionally the sensitivity of the warm cloud CRE to aerosol changes with the constraints in place, even "homogeneous" marine stratocumulus cloud deck regions. Vast areas of darkening effects [..[173]]are substantially moderated when the local meteorology and LWP are explicitly considered (Chen et al., 2012). These regional reversals of sensitivity to aerosols demonstrate regime-specific responses [..[174]]on a regional basis. LWP in particular may play a large role in determining if a cloud brightens or darkens as a result of aerosol loading.

Partitioning by regime identifies environments and cloud states that buffer, amplify, or diminish cooling[..[175]]. Buffering can involve any number of [..[176]]meteorological processes that lead to an altered response [..[177]](Turner et al., 2007). For

[168] removed: clearly illustrate the role regimes play in constraining covarying environmental and cloud state effects on aerosol-cloud interactions. Table 1 shows that these variations

[169] removed: AIE

[170] removed: assume

[171] removed: treated as

[172] removed: evidenced by Figures 8and 9 where regionally λ changes with increasing constraints , even in the persistent and homogenous marine stratocumuls cloud deck in the southeast Pacific

[173] removed: , that would have dramatic implications for global AIE,

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[175] removed: due to the AIE

[176] removed: processes discussed

[177] removed: because the sensitivity is also a function of meteorological conditions

example, the local meteorology, especially RH, can work to inhibit or invigorate the cloud's response to aerosol (Lu and Seinfeld, 2005; Ackerman et al., 2004). Instilling limits on RH should decrease any co-variance between the lifetime effect and RH that could arise due to [..[178]]entrainment's role in cloud [..[179]]breakup (Kubar et al., 2015). [..[180]]Entrainment of drier air will force evaporation, decreasing particle size, while entrainment of moister air could have no effect or a reverse effect, increasing the number of CCN within the cloud.

Unstable regimes may act as a buffer to cloud brightening, evident when global observations are partitioned by EIS and RH (Figure [..[181]]

[..[182]]6h). Unstable regimes contain pre-convective clouds (Nishant and Sherwood, 2017). Shallow cumuli, a common pre-convective cloud type found in the equatorial trade regions, are not likely to undergo the same reaction to aerosol loading as stable warm clouds like marine stratocumulus. Unstable conditions lead to strong vertical mixing and a reduced aerosol sensitivity, as activation favors strong vertical mixing in a stable environment (Cheng et al., 2017). Instability may alter the evaporation-entrainment feedback of the cloud, resulting in little to no brightening of the cloud and a severely reduced sensitivity, the result of forced evaporation reducing particle size. A reduced particle size would affect the lifetime of the cloud as well as the cloud albedo, reducing the sensitivity of the warm cloud radiative effect to aerosol loading as seen in our results for some unstable, dry regions (Jiang et al., 2006). The most unstable regimes in both [..[183]]Figures (4) and (6h) display the smallest sensitivities, which may be due to in-cloud turbulence decreasing the activation efficiency of the aerosol. [..[184]]

Without controls on the local meteorology, signals like those seen off the coast of South America, a large negative effect dominating the tropical region, may be due in part to the instability of the region and not truly reflect cloud sensitivity to aerosol loading (Figure 8). In the equatorial Atlantic off the coast of Africa, the strong decrease in CRE with respect to aerosol may not be the result of aerosol loading but that of surface winds decreasing cloud cover (Tubul et al., 2015). Surface winds were not included in analysis because the dependence of the warm cloud radiative response to aerosols depends most on LWP, RH, and stability, with only some regions showing a dependence on surface winds in our initial analysis. In the tropics, the warming sensitivity may be meteorologically-driven by increased frequency of trade cumuli and pre-convective clouds as stability decreases. These positive, unconstrained sensitivities are damped with environmental regime constraints (Figure 8b and 8c), however, darkening regions still appear in the fully constrained map (Figure 9), demonstrating that [..[185]]a substantial population of warm clouds display a true, aerosol driven [..[186]]darkening effect.

[178]removed: RH

[179]removed: cover and

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[181]removed: 7).

[182]removed: Unstable

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[184]removed: Unstable regimes contain pre-convective clouds (Nishant and Sherwood, 2017). Shallow cumuli, a common pre-convective cloud type found in the equatorial trade regions, are not likely to undergo the same reaction to aerosol loading as stable warm clouds like marine stratocumulus. Environmental effects minimized by λ_{REG} culminate in various sensitivities seen in the bottom of Figure 8.

[185]removed: although some signal in the unconstrained map is due to other influences

[186]removed: reverse Twomey

The role of cloud state constraints is to hold [..[187]]LWP approximately constant. The sensitivity to aerosol depends strongly on LWP, consistent with Wood (2012) and Ackerman et al. (2004). This relationship between LWP and aerosol-cloud-radiation interactions must be parameterized in models in order to constrain covarying effects and models must accurately simulate LWP in order to faithfully represent the cloud response (Quaas et al., 2009; Wang et al., 2011). Model parameterizations have improved the representation of warm cloud moisture fluxes, which strongly control low cloud variance, but confidence in any AIE estimates depend on cloud parameterizations continuing to improve (Guo et al., 2014).

The environmental and cloud state regimes work to limit the co-varying effects on sensitivity estimates. On both global and regional scales, the environmental constraints reveal regime-specific responses (Figures 3, 8) that allow the separation of conditions that lead to a buffered response that is especially evident in the tropical regions which undergo a sign change when meteorological constrains are in place (Figure 8) (Mülmenstädt and Feingold, 2018).

[..[188]]In the equatorial regions, controlling for the local meteorology (Figure 8c) reduces both the sensitivity and reverse Twomey effect compared to both the unconstrained (Figure 8a) and cloud state constrained (Figure 8b) estimates. In regions that exhibit strong cloud darkening effects, a deepening boundary layer, with decreasing stability, decouple warm clouds like marine stratocumulus from the surface, fostering cloud break up, and in turn, decreasing the cloud fraction and associated CRE of the scene. The negative [..[189]]sensitivities seen in the unconstrained top panel of Figure 8 [..[190]]are likely a result of this process, which happens simultaneously with a reduced stability, and epitomize how a single linear regression of warm cloud CRE against ln(AI) can capture meteorological effects when unconstrained (Wyant et al., 1997).

Although not explicitly [..[191]]controlled for, partitioning by [..[192]]LWP should also somewhat [..[193]]limit the effects of precipitation[..[194]]. Clouds with less than [..[195]].15 $\frac{kg}{m^2}$ rarely precipitate, therefore enforcing a LWP limit at [..[196]].15 $\frac{kg}{m^2}$ delineates possibly precipitating from non-precipitating clouds (L'Ecuyer et al., 2009). If precipitation does modulate aerosol-cloud interactions, [..[197]]the influence would only be observed in the highest LWP cloud state regimes. This is not to say precipitation is not important to aerosol-cloud interactions. In principle the regime framework presented here [..[198]]must be adapted to subset scenes according to the presence of precipitation, but that is not the focus of our study.

[187] removed: cloud state approximately constantby constraining LWP. The distribution of warm clouds favors thin clouds, which have been shown to be more abundant and therefore more important to aerosol-cloud-radiation interactions (Hirsch et al., 2017). The results demonstrate that the

[188] removed: The environmental and cloud regimes work to limit the co-varying effects on sensitivity estimates. This is evident when the different regime frameworks are compared (Figure 8).

[189] removed: λ

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5 Conclusions

Explicitly sorting satellite data by liquid water path, stability, and entrainment places increasingly stronger constraints on the partial derivative of CRE against ln(AI). This is shown to limit covariance between aerosol-cloud-radiation interactions and the environment and cloud state. In the absence of such constraints, buffering or modulation of the response by local meteorology obfuscates estimates of the AIE (Stevens, 2007). By filtering abundant satellite observations according to the stability and relative humidity of the free atmosphere and cloud liquid water path, the local meteorology and cloud morphology are held approximately constant minimizing the chance of misinterpreting covarying of meteorology and cloud morphology as aerosol effects when regressing CRE against AI (Gryspeerdt et al., 2014). These environmental drivers are known to influence cloud extent and radiative effect, and with constraints through the use of regimes, we can better attribute changes in the CRE to aerosol (Turner et al., 2007). Our results suggest that without constraints, the global mean AIE can be over-estimated by as much as 40% and regional variations can be artificially enhanced by as much as a factor of 2.

With environmental and cloud [..[199]]state constraints in place on a regional basis (Figure 9), strong, regionally specific cloud responses [..[200]]are identified and confidently attributed to aerosols. Clouds in the southern subtropical oceans, such as marine stratocumulus, exhibit the largest sensitivity to aerosol[..[201]]. Trade cumuli in the equatorial region show a much smaller, almost negligible signal comparatively. In the northern oceans, warm cloud decks from mid-latitude cyclones through the north Atlantic interact with North American and European emissions, leading to a cooling effect.

Interestingly even after cloud state and meteorology are controlled, the analysis still reveals coherent regions of aerosol forced cloud darkening effect (Figures 6h, 9). This aggregate dimming, or reverse Twomey, effect occurs in 15% of the regions studied and appears to be a robust characteristic of low LWP clouds in unstable, dry environments. This is similar to other observation based studies which found the same dimming effect in [..[202]]$\sim$20% of warm clouds (Chen et al., 2012). Our study suggests such clouds are sufficiently abundant to consistently yield a net [..[203]]warming sensitivity over a substantial, coherent, region of the globe. Models must be able to recreate warm cloud responses, including the [..[204]]a dimming effect, if they are to accurately simulate global aerosol indirect effects.

Both on a regional and global scale, constraints reduce co-variance of sensitivity estimates (Gryspeerdt and Stier, 2012). With constraints, the sensitivity can range from .46 to -.11 $\frac{Wm^{-2}}{\ln(\mathrm{AI})}$ on a regional scale (Figure 9), while without constraints the range increases from .77 to -.52 $\frac{Wm^{-2}}{\ln(\mathrm{AI})}$ (Figure 8a), signaling covarying influences and buffering by the cloud distort the signal [..[205]]on even a regional scale. Future regime classifications should prescribe precipitation limits to further separate the effects of aerosol-cloud-precipitation interactions, which are especially important to the cloud lifetime effect, where precipitation suppression leads to a larger cloud extent and lifetime.

[199]removed: regime

[200]removed: can be identified that can confidently be

[201]removed: reflecting the interactions between the marine stratocumulus decks in the southeast Pacific and south Atlantic, which interact with aerosol

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[203]removed: inverse

[204]removed: reverse Twomey

[205]removed: even on

Data availability. All satellite observations and MERRA-2 reanalysis used in this study are available for download through the NASA's Data Portal at http://doi.org/10.17616/R3106C.

Competing interests. The authors declare that they have no conflict of interest.

6 Author Contributions

Alyson Douglas evaluated all data and wrote a majority of the paper. Tristan L'Ecuyer constructed the original premise, guided the evaluation, and helped with paper edits.

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Figures/lwp_vs_if.png

(a)

Figures/LWP_bin_examples/second_lwp_bin.png

(b)

Figures/LWP_bin_examples/fifth_lwp_bin.png

(c)

Figure 2. Values of the sensitivity of CRE to aerosol ([..[a]]λ_{LWP} from equation (5)) for different resolutions of cloud state regimes. [..[b]]The weighted, summed λ_{LWP} is -13.12 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$ with 8 partitions. Plots of warm cloud shortwave CRE against ln(AI) [..[c]]are shown below for [..[d]](b) thin (.04 to .06 [..[e]]$\frac{\mathrm{kg}}{\mathrm{m^2}}$) and (c) thick (.1 to .15 $\frac{\mathrm{kg}}{\mathrm{m^2}}$) cloud states. The red lines represent the standard deviation within each ln(AI) bin and the blue dots represent the mean SW CRE for each ln(AI) bin in plots (b) and (c)[..[f]].

[a] removed: λ)

[b] removed: Weighted

[c] removed: against CRE

[d] removed: LWPs between

[e] removed: (b

[f] removed: $\frac{\mathrm{kg}}{\mathrm{m^2}}$

Figures/resolution_env_only/10_ret_02_4.png

(a)

Figures/env_examples/zero_zero.png

(b)

Figures/env_examples/seven_six.png

(c)

Figure 3. The sensitivity of CRE to aerosol ([..[a]]λ_{ENV}) from equation (6) evaluated with constraints on the environment[..[b]]. When weighted and summed following equation (6), λ_{ENV} is -11. $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$[..[c]]. Plots of the individual regimes from an unstable (∼1K), dry environment (< 10% RH) (b) and stable (∼6K), moist environment (>30% RH) (c) where the red lines represent the standard deviation of the SW CRE within each ln(AI) bin and the blue dots represent the mean SW CRE for each ln(AI) bin.

[a]removed: λ
[b]removed: (
[c]removed:)

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(a) (b) (c)

Figure 4. [..[a]]The sensitivity of the warm cloud CRE to aerosol (λ_{ENV}) found [..[b]]using equation 6 for environmental frameworks of a) 25 (-11.29 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), b) 100 ([..[c]]-11. $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$) and c) 225 (-10.99 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$) regimes of EIS and RH.

[a]removed: Sensitivity
[b]removed: within
[c]removed: -11.04

Figures/diff_res_freq/freq_5.png Figures/diff_res_freq/freq_10.png Figures/diff_res_freq/freq_15.png

(a) (b) (c)

Figure 5. Frequency of clouds [..[a]]partitioned into of a) [..[b]]25, b) [..[c]]100, and c) [..[d]]225 environmental regimes of [..[e]]EIS and RH.

[a]removed: within environmental frameworks
[b]removed: 5
[c]removed: 10
[d]removed: 15
[e]removed: both

Figures/LWP_Separated/First.png | Figures/LWP_Separated/Second.png

(a) (b)

Figures/LWP_Separated/Third.png | Figures/LWP_Separated/Fourth.png

(c) (d)

Figures/LWP_Separated/Fifth.png | Figures/LWP_Separated/Sixth.png

(e) (f)

Figures/LWP_Separated/Seventh.png | Figures/ret_sums/10_ret_summed.png

(g) (h)

Figure 6. The sensitivity of CRE to aerosol ([..[a]]λ_{BOTH}) found with constraints on stability, RH and cloud state limits of a) .02 to .04 $\frac{kg}{m^2}$ (-3.7 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), b) .04 to .06 $\frac{kg}{m^2}$ (-2.2 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), c) .06 to .08 $\frac{kg}{m^2}$ (-1.4 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), d) .08 to .1 $\frac{kg}{m^2}$ (-1. $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), e) .1 to .15 $\frac{kg}{m^2}$ (-1.5 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), f) .15 to .2 $\frac{kg}{m^2}$ (-.5 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$), and g) .2 to .4 $\frac{kg}{m^2}$ (-.4 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$)[..[b]]. Panel (h) is the summed, weighted sensitivity λ_{BOTH} within each environmental regime. The weighted, summed sensitivity is -10.6 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$ (sum of panel (h)). Note the colorbar for panel (h) is adjusted due to weighting.

[a]removed: λ

[b]removed: $\frac{kg}{m^3}$

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(a)

Figures/ret_sums/10_ret_summed.png

(b)

Figures/ret_sums/15_ret_summed.png

(c)

Figure 7. The sensitivities of CRE to aerosol from equation (7) within environmental regime resolutions of a) 5 by 5 (-10.8 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$), b) 10 by 10 (-10.6 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$), and c) 15 by 15 (-10.6 $\frac{\mathrm{Wm^{-2}}}{\ln(\mathrm{AI})}$) summed over all cloud [..[a]]states. Unlike all previous sensitivity estimates, these are weighted by occurrence.

[a]removed: regime

Figures/maps/REGRESS.png

(a)

Figures/maps/CLOUD_REGIMES.png

(b)

Figures/maps/ENV.png

(c)

Figure 8. The sensitivity of CRE to aerosol [..[a]]evaluated regionally with (a) no regimes constraints, (b) only cloud state constraints, and (c) only environmental constraints for each 15° by 15° region. Total sensitivities are (a) -11.8, (b) -28.5, and (c) -13.8 when weighted by occurrence. $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$.

[a]removed: (λ) found

Figures/maps/ALL.png

Figure 9. The sensitivity of CRE to aerosol ([..[154]]λ_{ALL}) found on a regional basis with cloud state and environmental regime constraints. The total regime weighted, global warm cloud sensitivity to aerosol perturbations is -10.13 $\frac{\mathrm{Wm}^{-2}}{\ln(\mathrm{AI})}$.