Response to reviewer comments Aerosol-mid-latitude cyclone indirect effects in observations and high-resolution simulations

We thank the reviewers and editor for their consideration of our manuscript. Responses to specific comments are shown below. Responses are in bold font.

Editor comments:

A sample of important issues raised by the reviewers that must be addressed. (I'm not implying that others don't need to be addressed.):

1) The paper should lead with the observations. The models have indirect effects 'baked in' even at the 'convection permitting' resolution so the hypothesis posed at the top of pg 12 rings a little hollow.
Please more clearly delineate observations and modeling, as requested by the reviewers.
The paper is now organized so that the modelling and observational results are presented in parallel. We have tried to more clearly delineate observations and models. We have also tried to make it clearer that, while the first indirect effect is baked into the simulations, adjustments are not. For instance, examination of the response of clouds to aerosol as modeled by CASIM in stratocumulus(Grosvenor et al., 2017) and mixed-phase convection(Miltenberger et al., 2017) display a diverse array of adjustments to aerosol. We have added text in several parts of the paper discussing how the observational analysis cannot prove causation and how the simple modelling experiments should only be used to hypothesize a causal link.

2) The regression analysis presents problems. First, one cannot draw conclusions about causality because of co-variability. Granted, the wording around the regression analysis doesn't suggest causality but the closing statements in the abstract and conclusions do. Second, it is very difficult to judge whether the regression fit of the proposed power law is good enough for quantitative conclusions to be drawn (only ~ 65% of variance in CLWP is captured). Fig. 11 is not helpful. Please carefully reconsider how you present the regression analysis.
You are right, this was not clear the way it was written in the conclusions. We have tried to make it clearer that our closing statements are based on the combined evidence of our idealized model and our analysis of the observed variability in the Earth system. The first paragraph of the conclusions now reads:

"Analysis of observed covariability between meteorology (as characterized by warm conveyor belt (WCB) moisture flux), warm cloud microphysics (as characterized by cloud droplet number concentration (CDNC)), and cyclone cloud properties is consistent with increasing CDNC leading to an increase in cyclone cloud liquid water path, fractional coverage, and ultimately albedo. While suggestive, empirical analysis of the observational record cannot prove causality. We support this analysis by performing a set of simulations where CDNC is set at high and low values. The response of CLWP to changes in CDNC in these simulations elucidates the mechanism by which this covariability may be explained and provides support for causality flowing from enhanced CDNC to enhanced CLWP."

We have also tried to expand on what 65% of the variance means. We believe that Fig. 11 of the previous revision is useful in that it illustrates the fit in two dimensions and clarifies that there is some dependence on both predictors, but we agree that by itself it is only useful to orient the reader. We have provided 95% confidence intervals on the coefficients of the fit in Table 2. All the coefficients are significant at 95% confidence.

3) The elimination of the Holhuraun volcano study, which is inconvenient for the main (relatively strong) conclusions, presents a problem, as noted by Reviewer 1. In the current version that study is only peripherally referred to. The fact that the re-working of that analysis suggests weaker evidence for aerosol-cloud interactions might require separate publication, but now that you are in possession of that knowledge it cannot simply be brushed aside. You would do better to discuss it.

Thank you for your understanding in this regard and allowing us to clarify why we removed this case study. We responded to reviewer 1 showing our analysis of this case study. There does appear to be an effect, but we are concerned that we need to do more analysis using NAME (or another dispersion model) to better understand the uncertainty as to where the plume from Holuhraun travelled.

We agree with reviewer 1 that the paper is more complete with the inclusion of this case study. We have added a section on the eruption and we note that our results do not necessarily contradict the lack of response seen by (Malavelle et al., 2017). Examination of the difference in means between the climatological CLWP and CLWP during the eruption was significant in several cases, but shows sensitivity to cyclones very far from the eruption. We suspect this is related to uncertainty in the propagation and rain-out of the plume at these distances as simulated by the dispersion model. We want to run more simulations exploring the uncertainty regarding the eruption. This would necessitate that simulations with different emissions fluxes, heights, and precipitation removal efficiencies be explored. This is not feasible in the context of the current paper, which is why we removed this case from the previous version with the intent of following up on this case in the future when we could understand the uncertainty space surrounding the plume modelling. We have described these uncertainties in the text and stated the steps we will take in the next paper to clarify these uncertainties. We have also added analysis showing that CLWP was unusually high in cyclones predicted to have interacted with the volcanic plume. We hope that this gives the reader sufficient information to interpret our global analysis. All this material is in sections 2.3.2 and 3.7.

4) Regarding the modeling, Please provide more clarity on key components of the microphysics. The dependence of Nd on aerosol and updraft is discussed at length but what autoconversion scheme is used? This is probably a more important issue than activation. Some autoconversion schemes have particularly strong dependence on drop concentration while others do not and this has significant impact on aerosol-cloud interactions.

Shipway and Hill (2012) describes CASIM, which utilizes Khairoutdinov and Kogan (2000) to describe autoconversion. This was briefly referenced in the methods section so we have added text to clarify this:

“The CASIM microphysics scheme is described in Shipway and Hill (2012). The warm rain processes in CASIM is compared to other microphysics schemes in Hill et al. (2015). The cloud physics parameterization used in CASIM is described in Khairoutdinov and Kogan (2000).“

We have also added text to discuss how our results might be different with a different microphysics scheme. For warm rain there is modelling evidence that CASIM shows good agreement with other microphysics schemes (bin and bulk)(Hill et al., 2015) and so qualitatively, we would expect similar results, if all else is equal. Obviously, the simulations presented here include ice processes, which may change the d(rain rate)/dCDNC < 0 relationship (Koren et al., 2005) and as such may be scheme dependent. The influence of ice on aerosol cloud interactions is very important and very uncertain, as are the ice phase parametrizations(Tan et al., 2016). This is an open area of research and we have added some text about this uncertainty.

We also accept that dynamic feedbacks associated with microphysics are very important(Xue and Feingold, 2006;Xue et al., 2008;Hill et al., 2009) and a different microphysics scheme may lead to a different dynamical evolution. However, given the forcing in a cyclone region and the resolution in even the high-resolution simulations, we believe the using another multi-moment bulk scheme would lead to qualitatively similar answers as those presented here, assuming the ice phase processes are similar. We have added text discussing this as well.

This is discussed in the end of section 2.3.1:

“Finally, in the simulations presented in this paper we explore the response of the clouds in the UM treated by the CASIM cloud microphysics to changes in CDNC. A different cloud microphysics scheme would potentially yield a different adjustment to aerosol, but our results are unlikely to be qualitatively dependent on the simplistic activation scheme chosen here. We also acknowledge that the adjustment of cloud to aerosol in these idealized simulations will be a function of the CASIM microphysics scheme. Examination of CASIM in relation to other multi-moment schemes suggests that if the adjustment works through the warm rain process another multi-moment scheme would produce a qualitatively similar result (Hill et al., 2015). It is important to note that the simulations presented in this work include ice processes, which may affect the susceptibility of rain rate to changes in CDNC (Koren et al., 2005;Rosenfeld and Woodley, 2000). These effects may be highly dependent on the choice of microphysics scheme. Further, the representation of these effects in models is very uncertain and could substantially affect the predictions of our simulations. Another important mechanism not considered in our simulations is the dynamical feedback on aerosol-cloud interactions(Hill et al., 2009;Xue and Feingold, 2006;Xue et al., 2008). The dynamical evolution in response to changes in CDNC could change dramatically using a different microphysics scheme. We believe that our simulations are not strongly sensitive to the representation of these effects in CASIM given the resolution of the simulations and the forcing within a cyclonic system.

Overall, we present these simulations as an exploration of how clouds within cyclones respond to changes in CDNC through the warm rain process. These simulations are used to contextualize the observations and evaluate whether we may reproduce observational variability utilizing this idealized set of simulations. ”

5) Many of the supplementary figures indeed belong in the main text. Please reconsider the distribution.

We have moved the figures as suggested by reviewers. Several of the figures are just sensitivity tests replicating an existing figure and we have left those in the supplementary material.

Some specific comments of my own:

1) In the opening lines of the abstract there is a conflation of 21st century climate change and aerosol effects on midlatitude cyclones. Climate change does not equate to increases in N_d (drop concentration).

That was sloppy- we meant to say: 'infer climate sensitivity, which allows us to predict 21st century climate change(Andreae et al., 2005).'- we have amended this to read:
"Establishing how much aerosol emitted during the 20th century has enhanced the liquid water amount and thus the albedo of midlatitude storm systems is a key step in constraining the climate sensitivity inferred from the observational record."

2) Pg 5, lines 1 and 14: N_d is not retrieved at cloud top. It is based on cloud top effective radius and (path integrated) cloud optical depth
That is right, this has been amended. Thanks.

3) Pg 6: perhaps cloud property retrievals are not subject to thresholding but you still have to define cloud fraction so that all sky albedo can be calculated. How is cloud fraction defined? Is it consistent amongst analysis and model output?
The all-sky albedo is calculated by the SYN1DEG algorithm using the algorithms described in Doelling et al. (2013);(Doelling et al., 2016). This does use information about the cloud mask to create angular distribution models to convert radiances to fluxes(Loeb et al., 2003) and to account for diurnal variations between different satellite overpasses and geostationary data. It does not use the cloud fraction explicitly in the calculation of all-sky albedo beyond creation of the angular distribution model and using the geostationary observations to fill in the diurnal cycle. We have added text clarifying this.

We do utilize cloud mask from the CERES SYN1DEG data set, which is retrieved following Minnis et al. (2011). This data product is defined differently than the model, which is max-random overlap. Because of this we do not offer a comparison between model cloud fraction and CERES SYN1DEG cloud fraction.

Thank you for this comment. We have expanded the methods section to clarify these points:
"The 3-hourly data is averaged to create a daily-mean albedo and CF. CF, clear-sky albedo and all-sky albedo are provided in the CERES SYN1DEG data set edition 4(Wielicki et al., 1996;Doelling et al., 2013;Doelling et al., 2016). CF is calculated from MODIS and

geostationary satellites based on the Minnis et al. (2011) cloud mask. It is used in the calculation of the albedo retrieved by CERES as described in Doelling et al. (2016) to create an angular distribution model and to interpret geostationary observations of albedo in relation to the observations from CERES. It should be noted that without utilizing a satellite simulator(Bodas-Salcedo et al., 2011) we cannot directly compare cloud fraction to the aquaplanet simulations presented in this work."

4) Pg 7, "microphysics and aerosol explicitly interact" is a bit strong. The statement is true but a bit misleading given the resolution.
The sentence has been amended to explain that the interaction is explicit at the resolution of the model. Our intention was to convey that this was an improvement on parameterized convection:
"Without convection being parameterized microphysics and aerosol explicitly interact at the model resolution"

5) Pg 9, line 6: "disentangling". It seems like the meteorology is fixed and the aerosol is changed but this isn't what happens in nature so are you really disentangling anything of value? (see e.g., Feingold et al., PNAS 2016).
Good point- disentangle has been removed.

6) Pg. 10, line 15, what does "lifetime effect" mean in the context of a midlatitude cyclone. This terminology is vague at best. (See editorial comment below.)
Lifetime has been changed to adjustment throughout the paper.

7) Pg. 10, Fig. 2: how many simulations were performed. The observations average many initial conditions and aerosol-meteorology co-variability. Do the models all use the same initial conditions?
This was not clearly stated in the paper. We have added the clarification: *"Simulations were then run for 15 days. A single simulation was run at each resolution and aerosol concentration, giving a total of four simulations of 15 days each."*

8) Since aerosol concentration is fixed in the modeling, aren't you biasing your aerosol effects to be much stronger than would be the case if aerosol were washed out by the rain? If CASIM is indeed two-moment, it should remove aerosol and allow N_d to fluctuate accordingly.

To allow CDNC to fluctuate we would need to make some decisions about aerosol sources, which would increase the complexity of our simulations. In the context of our simple model we set CDNC and rain is inhibited by this. In the absence of dynamical feedbacks or ice processes we would expect a further enhancement in LWP and less precipitation cleaning out aerosol. This would mean that in the canonical representation of the Albrecht effect we would be somewhat under-representing our response. However, in our conceptual model we propose that rain rate is controlled by the large-scale environment and so we should not expect this fixed aerosol to affect the strength of the response as the LWP increases to push the rain rate back to be in agreement with the large-scale environment. This is good to discuss and we have added text to the aquaplanet section to clarify this assumption:
"It is also important to note that the fixing of CDNC at a constant value means that precipitation

does not affect CDNC via the removal of aerosol and thus CCN. The simulations presented here are intended to examine the adjustment in cyclone clouds to changes in CDNC as opposed to a change in aerosol fluxes. If aerosol were allowed to respond to precipitation we may speculate as to how this might affect the behavior of the cloud adjustment simulated by CASIM. As described in the following sections, the rain rate on a daily, cyclone-wide scale is determined by the large-scale environment. Subsequently, we may hypothesize that the feedback between aerosol, CDNC, and the rain rate is relatively weak, but we note that this assumption of fixed CDNC artificially removes this interaction pathway with the intent of understanding the adjustment in cloud properties to CDNC."

9) Pg. 14, lines 8-15: Wouldn't the open cells be raining and have low N and lower domain-averaged liquid water path?
We argue that their relatively low liquid water path and low CDNC should result in a higher susceptibility than in the frontal zones where aerosol susceptibility should be negligible. They are also raining, which is a necessary condition of having a precipitation-mediated cloud adjustment. This was not clarified in the text and we have added discussion to amend this as well as referencing the literature discussing the importance of precipitation to the open-closed cell transition:
"Numerous studies have linked the dominance of open or closed mesoscale cellular convection to precipitation, and aerosol modulation of precipitation(Stevens et al., 2005;Feingold et al., 2015;Koren and Feingold, 2011;Rosenfeld et al., 2006;Mechem et al., 2012;Goren and Rosenfeld, 2012;Wang and Feingold, 2009a, b). Because of the tenuous nature of this cloud regime, and because they are typically precipitating it is reasonable to suspect that they will be more susceptible to aerosol-driven changes in their macrophysics than either thick frontal clouds or non-precipitating clouds. It is not the intention of our investigation to examine the complex dynamics of mesoscale cellular convection, but we have chosen our observational data sets so that they do not exclude this cloud regime, and the localization of differences in CLWP between high and low $CDNC_{SW}$ *cyclone populations is suggestive given the existing literature regarding both the radiative importance of these clouds(McCoy et al., 2017c) and their relation to precipitation and aerosol(Koren and Feingold, 2011). Overall, this behavior motivates future work examining this region in higher resolution and higher complexity models that can resolve these features."*
Thank you for suggesting this.

Some editorial comments:

1) What do you mean by cloud-aerosol lifetime effect? This concept is particularly poorly defined. It doesn't reflect on the lifetime of the midlatitude cyclone. I assume you mean a reduction in precipitation efficiency. Regardless, please be precise.
Lifetime has been changed to adjustments to follow the more process-agnostic description used in the IPCC report. Thanks.

2) Acronyms like CDNC, while widely used, are bad practice since they can much more easily be represented by symbols (N, perhaps with subscript d) as you already do for aerosol concentration. The excessive use of acronyms will make papers in our field unreadable if we are

not more careful. CLWP could be replace by L_c, SZA by \theta, CF by f_c, etc. While I don't insist on these changes, please consider that they really help with clarity, particularly for those entering the field. (See pg. 15, what is CDNCSW??)

That was a typo on page 15- it was supposed to be CDNC_{SW}. It is true that reading papers from outside the field can often be difficult. However, use of symbols can also be confusing as they have less information for people to infer their meaning. The vague use of λ in the feedback and climate sensitivity community is one good example of this. In the interest of clarity we have added a table (table 1) of acronyms that can be consulted by the reader.

3) CCN, again is widely used, but has no quantitative meaning unless defined at a specific supersaturation. You might as well use the same symbol as for accumulation mode aerosol number concentration if indeed they are used interchangeably.

That is a good point, we have changed CCN to N_{acc} when referring to the simulations since that is all that is being perturbed.

4) The manuscript has some lapses in grammar (e.g., Pg 3 lines 20-22)and could benefit from some liberal use of commas in some places.

This has been amended.

5) Many figures and their fonts are much too small to be readable in print form.

Figure sizes and font sizes have been increased.

Report #1

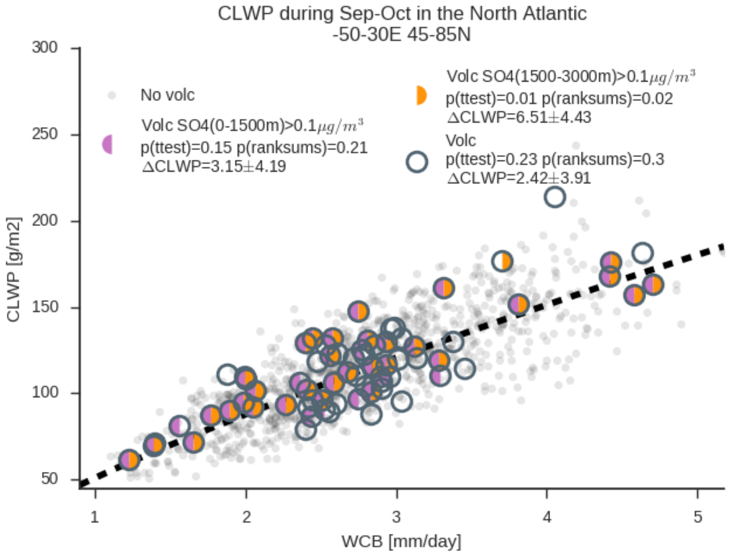
The weak evidence of the Honuluraun case from the first submission has been removed completely. The rebuttal states that the revised analysis gives even poorer evidence for an aerosol effect and the case will be revisited in another article. Such poor evidence is contradicting the claim of the article "aerosol-cloud indirect effects have substantially altered clouds in extratropical cyclones" (see abstract and conclusions). I consider this bad practise that we should not pursue in science.

We understand that the reviewer is concerned that by removing the Holuhraun case from the manuscript we are suppressing contradictory evidence. Overall, we find that the Holuhraun case study does not contradict the results presented in the body of the paper. If it did, we would agree with you that this is bad practice to remove it. Although we replicate the following discussion in the paper, we also want to present our analysis for the reviewer.

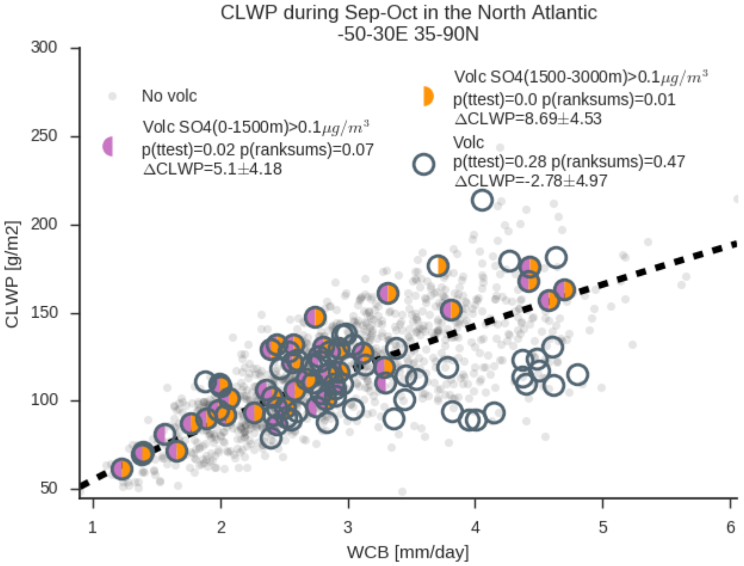
We examined September and October cyclones in the North Atlantic (50°W-30°E) during the eruption and in non-eruption years. The NAME dispersion model was used to flag cyclones as being significantly affected by SO4 from Holuhraun.

We want to see if Sept-Oct 2014 had unusually high CLWP given the synoptic environment. If we examine the climatological dependence of September-October CLWP on WCB (excluding 2014) for 50°W-30°E and within 20° latitude of Iceland we get the fit shown using the dashed line. A 20° latitude range is consistent with a 48-hour transit time from the fissure(McCoy and Hartmann, 2015). The cyclones during the eruption, and flagged during the eruption to have interacted with the plume are shown using open

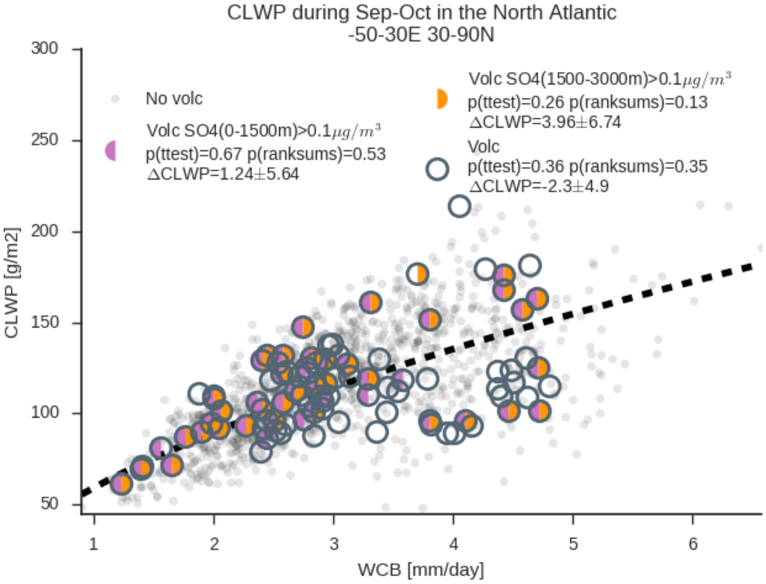
circles and half-circles, respectively. An average volcanic SO4 predicted by NAME of 0.1µg/m^3 in the southwest quadrant of the cyclones was used to flag whether the cyclones had interacted with the plume. The anomalies in CLWP relative to the climatology were calculated using the dashed-line fit. A t-test and rank-sums test were used to evaluate whether the mean anomaly during 2014 was unusual. We found that cyclones flagged by NAME with emissions at 1500-3000m were significantly different than the climatology at 95% confidence (see legend).



However, we also found that this was dependent on the region being considered. If we consider all cyclones whose centers were within 30° latitude (35°-90°N) find that the result is less sensitive to the emissions height.



But if we expand to latitudes from 30-90°N there are five cyclones near 30° that have a very high WCB, but a low CLWP. The negative anomaly from these cyclones results in the mean anomaly not being significant at 95% confidence.



We feel that this sensitivity of the population mean anomaly during September and October of 2014 to cyclones in excess of 30° latitude from Iceland is somewhat unrealistic. We hypothesize that expanding the dispersion modelling to consider more emissions heights, and fluxes, was well as altering the dispersion model's assumptions about its efficiency in removing aerosol via precipitation will allow us to better understand the robustness of our results. This excessively complicated the analysis in the paper and was not really feasible in the timeline of review.

We would like to reiterate: our removal of the Holuhraun case study was not due to the discovery of conflicting evidence- we just felt that we could do a better job understanding uncertainty in this case in a future paper using the tools we had developed in this paper. Based on the reviewers' request we have placed all of our tentative analysis of this case in the text and have outlined what simulations need to be done to constrain this case. We hope that this information will allow readers to evaluate the robustness of our results for themselves.

We have added this material in section 3.7 and described the plume modelling in section 2.3.2.

Moreover, the aerosol effect on cyclone albedo that the current study suggests should not be called substantial. Substantial differences in the LWP are in my view rather associated with meteorological variability as otherwise the analysis would not need to be classified by WCBs (e.g. Fig. 2). Now that the methods are described in more detail, there seem to be additional problems with the data analysis that I also briefly comment on in the following.

Substantial and dominant are not the same thing. We stated that meteorology dominates cyclone variability. It is hard to see how this could not be the case (see page 18 line 15 of the previous submission). We have tried to add a few more comments into the text making it clear that the variability is dominated by meteorology.

The used all-sky albedo includes cloudy and clear pixels. Hence the analysis does not only include aerosol-cloud interaction, but also aerosol-radiation interaction that cause increased all-sky albedo. There is therefore no basis to separate the effect of aerosol on cloud albedo from the effect of aerosol on the clear-sky reflectivity. This might be primarily a problem in the cool sector of the cyclone with broken clouds that seem to show the greatest effect (P. 14 14-18). Using the term "Aerosol-cyclone indirect effect" is therefore misleading as it suggests a separation between aerosol-radiation and aerosol-cloud interaction within cyclones.

Thank you for suggesting this. We used the all-sky albedo because we felt that it was less sensitive to thresholding than the in-cloud albedo. It is fair to note that the difference in albedo between the high and low CDNC cyclone populations could be contributed to by aerosol direct effects. We have added text to this effect to the manuscript. We have also provided analysis of the clear-sky albedo difference between cyclone populations. We find that the clear-sky difference was on the order of 0.005±0.001. The difference in all-sky albedo was nearly an order of magnitude larger (0.032-0.018). The effect of this clear-sky difference in albedo averaged in cloudy and clear regions should be relatively small compared differences in albedo due to changes in cloud properties given that midlatitude cyclones tend to have cloud fractions in excess of 70%. Thank you for suggesting this calculation. The following text has been added to the discussion:

" In this analysis we have used all-sky albedo from CERES to examine the response of cyclone albedo to changes in CDNC. This variable was chosen because it does not impose a criterion for what it considers to be a cloud when calculating the albedo, as a cloudy-sky albedo would. If we were to use in-cloud albedo this would necessitate the albedo perturbation being restricted to only confidently cloudy pixels (see Marchand et al. (2010)). For example, this could exclude situations where mesoscale cellular convection was occurring as these regions would not necessarily be considered cloudy. As pointed out by previous studies (McCoy et al., 2017c), these clouds may have a significant impact on all-sky albedo. However, use of all-sky albedo may potentially conflate aerosol direct effects and indirect effects.

First we provide an estimate of how much cloud fraction differences may contribute to the difference in albedo. Because cloud fraction and albedo have a fairly linear relation in the midlatitudes on a monthly time scale (Bender et al., 2011;Bender et al., 2017) we provide a calculation of the change in albedo related to changes in cloud cover. The observed midlatitude slope of the relation between albedo and fractional cloud cover of 0.4 from Bender et al. (2017) implies a change in albedo between the top and bottom third $CDNC_{SW}$ *populations of 0.005-0.007±0.002 (Fig. 10). As shown in Fig. 10, the difference in mean cloud cover between the populations is significant at 95% confidence, but it does not appear to contribute to the majority of the effect on albedo."*

Fig. 1:
CERES albedo shows a dependence on the SZA not only for angles greater than 30 degree. Indeed there the increase in albedo with SZA is strongest, but also for smaller SZA, we can see a dependence, e.g., the same albedo of 0.18 could be associated with a cloud fraction of roughly 80-90%. This might be a problem since the aerosol effects on cloud cover has a magnitude consistent with this uncertainty (Fig. 7). Maybe there is a better way for accounting for the SZA problem than just cutting off the worst part.

Because this is a 3D radiative transfer problem there is no easy solution to removing SZA dependence. As shown in the supplementary material, the SZA effect tends to conflict with the signal from aerosol-cloud interactions so the limiting of SZA is not creating the signal attributed to aerosol-cloud interaction.

Specific comments:

P. 6 9-11: "potentially allowing examination of broken cloud cover"
The satellite data of the study is daily averaged that is fairly coarse since extra-tropical cyclones are mobile such that the mean smoothes the quantities under investigation, e.g., CDNC and cloud cover.
This was unclear. What was meant was that if we had used only in-cloud retrievals of albedo we would throw out all the data from broken cloud because these do not necessarily meet the criterion required to perform a cloud retrieval (for example see Marchand et al. (2010)). By using all-sky albedo this allows us to consider contributions from sub-pixel clouds and clouds that are not surrounded by cloudy pixels. This has been expanded on in the text to make it clear that we are only considering their contribution to their albedo, we aren't picking them out.

P. 8 19- 28: "should be thought of in the context of a 'fixed-CDNC' set of experiments"
Given the way CDNC is calculated in the simulations, CDNC is by no means fixed, but rather artificially constrained.
The wording has been changed. Thanks.

P. 16 8-9: "To calculate the difference in terms of a radiative flux the difference in albedo is multiplied by the annual mean climatological downwelling SW"
I like the idea, but think the estimate is rather crude. Maybe rephrase it and rather speak of typical fluxes one would expect from certain values of downwelling SW fluxes. One such value could than be the climatological mean of a certain geographical position, but I nevertheless perceive such arbitrary estimates as rather uncertain and would not rely on them. That being said, it is misleading to have these values as a key result in the conclusions without further explanation or uncertainty estimate (P. 19 1-4).
This is a really useful insight - thanks. The way we calculated it was meant to be somewhat agnostic as to when and where the cyclones occur. The more relevant quantity in this regard is the albedo, but we felt that the albedo is a less intuitive quantity for a broad audience. We have changed the conclusions and abstract to state the albedo, as opposed to estimated change in SW and added discussion of this calculation to make it clear that this

assumes that the cyclones are randomly distributed between 30-80° throughout the year. We note that, for example, the seasonal cycle of DMS emissions in the Southern Ocean means that cyclone CDNC is highest during the period of strongest downwelling SW, so our calculation underestimates the change in SW because it ignores this source of covariability between insolation and CDNC. We now discuss that this is a quick calculation to contextualize the albedo difference and provide the difference in albedo first as you suggest. Thanks.

" To calculate the difference in terms of a radiative flux the difference in albedo is multiplied by the annual mean climatological downwelling SW associated with the CERES EBAF-TOA data set between 30°-80°. It is important to note that this assumes that the cyclones being affected are randomly distributed in latitude and during the year. This may be somewhat reasonable for anthropogenic pollution, but not for biogenic aerosol sources. Specifically, planktonic sulfur sources have a substantial seasonal cycle leading to their contribution in albedo occurring during the period of maximum insolation (McCoy et al., 2015). The difference in reflected SW provided here is only intended to act as a rough guide to contextualize the change in albedo."

P.19 5-8:

And how does the estimate compare to these other studies?

The page and line number correspond to the acknowledgements. We infer based on the content in the previous comment that this references the difference in SW. It is difficult to offer a direct comparison to other studies as they do not offer a comparable estimate of forcing from CDNC changes in the midlatitudes. We have tried to expand on comparisons between the present study and previous studies.

Report #2

The authors made major changes to this manuscript, focusing it and dropping some of the analysis (i.e Volcanic simulations) as requested by the reviewers. This is a much better presentation. I am not convinced that the authors have fully responded to the reviewers and concerns about the type of model used for this study, but this paper is getting there.

I tried to look at the whole manuscript again, since the focus changed. And it STILL needs some substantial revisions.

The paper is better. I think the abstract still feels disjointed. The paper does not know whether it is a modeling study or an observation study. The problem is that the 'observations' rest on a reanalysis model, and the key 'microphysical' parameter is just a passive tracer of human emissions in that model. Again, it feels like something slapped together from a few projects. But it's one less project now (only 2), so that's fine.

We appreciate the reviewer thoroughly re-evaluating our manuscript. As discussed extensively in the previous round of revisions the observations do not rely on MERRA2 reanalysis. The results are the same when using MODIS CDNC observations. That is to say, we could remove MERRA2 from the paper and the conclusions would be the same. We have provided supplementary analysis using MERRA2 proxy CDNC it because we are

concerned that there may be some residual remote-sensing biases in MODIS CDNC. The only reason we have shown results from MERRA2 is that it is not going to have retrieval error. Ultimately, the *two* estimates of CDNC (reanalysis and observations) result in the same conclusions. We have tried to further clarify that there are two CDNC estimates being used in this paper and to clarify that we are doing that because, independently they may suffer from modelling error (for MERRA2) or from retrieval bias (MODIS), but if they produce the same results then these results are at least not a function of either of these errors.

I think it needs better organization of the results and separation between observations and models. Significantly, the manuscript should decide the order of presenting results. E.g.: section 3.1 is called:

3.1 Observed relations between meteorology, mid-latitude cyclones, and aerosol

But then immediately:

3.1.1 Aquaplanet simulated mid-latitude cyclones and their response to changes in CCN

Put this in a more logical order please. It should perhaps flow from observations to model simulations. The figures could be kept in a similar way, or separated, but I don't think the present treatment works.

We have reorganized. Thank you. We found it very difficult to organize it as purely observations and then purely model, so we have organized things to go in parallel so we compare modeling and observations for each step of the analysis. We have expanded the number of sections in the discussion to make it easier to follow the analysis.

Also: with the narrower focus, there should not be a need for supplementary material, and this should be brought forward.

We feel that it is good to use the SM to examine the result of various sensitivity studies that are effectively just the same figures as the main text, but repeated with slightly different parameters. These could equally just be 'the calculation is not sensitive to X (not shown)', but it seems better to just show it in the SM.

A few broad notes:

1. I think what I fundamentally have a problem with is applying sophisticated tools like a high resolution model in coarse ways (aquaplanet, specified and simplified aerosols and microphysics). And then saying that is what climate change will look like. I think this is valid for the WCB analysis and that partitioning is fine and interesting, but I think it is difficult to then compare that to the real world.
We are only using the model to understand how to interpret the observations. In this case the aquaplanet is used to justify the WCB partitioning, and the idea that the CLWP should

adjust at a given WCB in response to changes in CDNC. Based on your suggestion we have tried to clarify that our closing statements are based on the combined evidence of observations (which cannot show causality) and simplified modelling (which does not fully capture all the processes in the real world). We describe the pros and cons of using a convection-permitting model for this type of simulation and feel that the use of a high-resolution model allows us to do a better job resolving relevant processes than a traditional GCM.

2. Perhaps it needs to be stated that most of what is going on in mid-latitude cyclones is resolved at the scale of the model. That's not really stated, and would probably be scientifically defensible. But it does NOT look that way from the model results: convection matters, because the answers change.
We have discussed how the simulations at resolutions from 1-16km compare as presented in Field et al. (2017). In the discussion comparing the cyclones in the simulations and in the observations we show several dissimilarities. The model grid will not be saturated some of the time when there should be a cloud. Because we don't have a cloud scheme we will never have a cloud in these cases.

From Figure 2 and Figure 6: the MERRA2 and LR results look similar, except the Low and High CDNC have opposite effects. The high resolution aquaplanet does look like observations, but this is fundamentally different than low resolution.
That's right. Based on this analysis MERRA2 isn't building in the CDNC effect. It is true that both the LR and MERRA2 cyclones have a much lower sensitivity to WCB moisture flux. However, we do find that even the low-resolution UM simulation has a qualitatively similar response to increased CDNC at a fixed rain rate/WCB as the high-resolution model does.

3. Trying to pull parts of a reanalysis (like using sulfate as a proxy for CDNC) is dangerous. Not only are uncertainties in the data swept under the rug, but the reanalysis is also then being pulled in different directions. I know this has been published before and 'it works', but I think that really brushes away a lot of processes.
4. The WCB part is useful, except to the extent that ice and ice indirect effects is not really treated. But I think trying to make inferences then about the real atmosphere from the coarse analysis of the MERRA data is very dangerous.

Please see comment above regarding the presentation of MERRA2-inferred CDNC and MODIS-observed CDNC in parallel.

5. The analysis takes too 'high level' a view and does not dive deep enough into what is going on in the model. What is going on to make the LR and HR simulations respond differently? You imply it is convection. But you could show that in the model, by figuring out where convection is in the cyclones, and relating that to the changes in Figure 6.

This is beyond the scope of the present analysis. Ultimately, if we just used the LR model or the HR model it would not affect our interpretation of the observations. Based on your

comment we have rewritten the statement that differences between LR and HR are related to convection. There are several substantial differences between the LR and HR simulations- not just convection. Thanks.

Some further detailed notes:

P5, L15, "MERRA2 sulfate mass is a good predictor of CDNC as observed by MODIS". You say no cross talk: but if aerosols and clouds are convolved in MODIS, then there is cross talk because MODIS aerosol is assimilated to get SO4?

This is discussed at length in our papers (McCoy et al., 2017a;McCoy et al., 2017b), thus we did not replicate the discussion here and just referenced the papers. Several pieces of evidence indicate that both MERRA2 SO4 and MODIS CDNC are good predictors of the true CDNC. We find this by comparing in-situ results to MODIS CDNC. They compare well, and this has been shown by other authors (Bretherton et al., 2010;Bennartz and Rausch, 2017;Painemal and Zuidema, 2011) in addition to our analysis in (McCoy et al., 2017a). We also find that long-term trends in SO4, which are consistent with long-term trends in observed SO2 emissions, are skillful at predicting long-term trends in CDNC (McCoy et al., 2017a). The MERRA2 aerosol retrieval utilizes AOD to nudge the reanalysis, but this is adjusted for swelling (Bosilovich et al., 2015;McCarty et al., 2016;Chin et al., 2002). There may be residual signal that the correction (Chin et al., 2002) does not account for, but again, this is why we reproduce everything with MODIS observations.

Figure 2: This is a mess, because the contour intervals are all different. If you make them the same, I think you are going to end up with some wildly different results. I think that makes the analysis and comparisons very difficult.

We note in the text that the model and observations give different looking cyclones. This is expected. We didn't utilize a cloud scheme because that would require additional tuning. We apologize for the colorbars. The color intervals and ticks should be the same now. Previously we had only set the color range so the ticks were all the same, but python clipped the colorbars in a weird way.

Figure 3 also seems hard to compare: Aquaplanet, MAC-LWP and MERRA2 seem very different.

As noted, we expect this. This is discussed in the beginning of section 3.1.2 from the previous round of revisions. We have made sure that both the ticks and range on all the color bars are the same now.

P12, L17: the problem is that sulfate mass concentration in MERRA may not interact appropriately with clouds (e.g., incorporation in cloud drops, scavenging, etc) if there are no indirect effects.

Please see comment above regarding the presentation of MERRA2-inferred CDNC and MODIS-observed CDNC in parallel.

P15, L10: The albedo discussion here is not convincing. The model (Figure 9) looks quite different than the other analysis in Figure 8.

We discuss that the albedo difference in the simulations is not very similar to the albedo difference in cyclone populations. We have expanded the discussion of the difference in albedo between the cyclone populations in Section 3.5.

P15, L20: Im confused by this paragraph, and concerned at the method. The 2nd sentence on line 23 is missing a critical clause. Also: here is another place where you have mixed up observations and simulations in the flow. Perhaps this should be with observations.

There appears to be some discrepancy with the version on ACPD and the version you are referencing. Line 23 only has one sentence. We have tried to clarify the separation between observations and simulations in the discussion.

P16, L20: I'm not convinced a regression model works here based on the previous analysis and the different behavior with MERRA.
Please elaborate on what aspects of the analysis you feel are not suited for a regression model. The coefficients are all significant at 95% confidence. Heuristically, if we look at CLWP as a function of the predictors it seems to depend on both pretty clearly (Fig 11). We are happy to do any other tests of the fitness of the model that you suggest. Also, see comments above regarding use of both MERRA2 and MODIS separately.

P18, L7: This first conclusion: you come up with the same results of many models that do not match observations (e.g. Mallevele et al 2017). Why should these results with bulk microphysics be any more valid? The processes in your high resolution simulations are the same for stratiform cloud microphysics as in many global models. Why would these simulations not have the same issues?
Based on the comments of reviewer 1 we have now shown how our results compare to (Malavelle et al., 2017). We have rewritten the conclusions substantially to more clearly express that our result is not strongly dependent on model resolution.

P18, L18: Please comment on the limits of the regression model.
We have added confidence intervals to the coefficients in the regression model (see table 2).

Report #3
This revised version of the manuscript explores the impact of aerosols on liquid water path in extratropical cyclones. This new version is more detailed and more convincing that the previous paper, but there are still issues, mostly to do with the presentation of the results and the structure of the paper. The comments below list the various parts were rewriting or modifications are needed to make the paper clearer and more fluid.
Based on the sensitivity of the results to the source of cloud droplet number concentration, the conclusions should be somewhat expanded and in some places toned down.

Otherwise, the manuscript is acceptable for publication after minor but highly recommended revisions.

We thank the reviewer for their time reading our revised submission. While the CDNC source does seem to have a minor quantitative effect on the results, we found that the change in CDNC (MERRA2 or MODIS) data set did not qualitatively impact the results. One thing that should be noted in the manuscript, and which might explain some of the perceived discrepancy between MERRA2 and MODIS is that because of remote-sensing constraints the MODIS observations are only available when the sun is at low solar zenith angle. This means that MODIS doesn't have observations over the Southern Ocean in winter, which is one of the lowest CCN places on Earth. MERRA2 does have the ability to predict SO4 in high latitude winter so it has a large number of low CCN cyclones. The Southern Ocean in winter in MERRA2 is just based on a climatology of DMS emissions over the remote Southern Ocean, but the mechanism explaining this behavior (no light for phytoplankton in winter and thus only sea spray) is well understood and agrees with numerous other lines of evidence (McCoy et al., 2015;Ayers and Gras, 1991;Boers et al., 1994). However, this low CDNC data that MERRA can infer doesn't disagree with the results from MODIS and only really seems to extend them to very low CDNC.

Detailed comments:

1. The document needs some cleaning and tightening as the current presentation still includes remnants of the previous version. Section 2.3.1 is the only subsection in section 2.3. Similarly 3.1 is the only subsection of this level in the results section 3.

Thank you, we have reordered our sections.

2. In the conclusions, please discuss where the additional variability in cloud liquid water path might come from, as you indicate that 60% (and not 100%) or this variability is caused by the moisture flux and aerosol impact on cloud droplet number concentration.

We have added additional discussion to this section speculating where the additional variability may originate from.

"While we should not expect to explain all of the variability in CLWP no matter how many predictors we use, it is likely that the explained variability in our regression model could be improved by (1) a more skillful metric for moisture flux into the cyclone, (2) a more accurate observation of $CDNC_{SW}$*, or (3) additional information regarding ice and mixed-phase cloud properties. In regards to point (1): we have chosen to predict moisture flux in this way so that we may observe it utilizing microwave radiometers. In regards to points (2) and (3): we note that both of these retrievals are difficult and are likely to improve as the remote sensing community examines them in more depth. Overall, explaining the majority of extratropical cyclone liquid water path variability utilizing two predictors is a useful contribution to our understanding of the midlatitudes."*

Thanks.

3. You might want to relate your results to a recent paper by Naud et al. (2017), who explored the co-variations between aerosol optical depth and cloud cover in extratropical cyclones. In particular they examine cold and warm fronts separately, which might help explain better your assertion that the cold frontal region is where the albedo change with aerosols is concurrent with the change in liquid water path and cloud fraction. Also, while cited, the Grandey et al (2013)

study who explore the impact of extratropical cyclone strength on the cloud-aerosol relationship could be further discussed in light of the results of this paper in the conclusions. Both of these studies to some extent contradict the statement in the introduction that "aerosol-cloud indirect effects have not been observed in extratropical cyclones" (L25, p2). Although neither study can establish a causal relationship, the fact remains that observations in extratropical cyclones have already been used to explore aerosol-cloud relationships.
Thank you, we were not aware of the recent Naud et al. paper, and it is very relevant. We have expanded the discussion of both papers and removed our statement in the introduction in light of this discussion.

4. In the introduction please indicate where these cyclones are. In 2.1 it is specified that they are over the oceans but a latitude range should be specified. I suppose that these are all in the northern hemisphere? Are they found in both Atlantic and Pacific oceans? Are the Mediterranean cyclones also included? At least it is specified for the simulations but at the bottom of page 12, it sounds as if the observed cyclones are sampled in both hemispheres. If indeed this is the case, then "southwest quadrant" is misleading.
This is discussed in the methodology. The cyclones are from 30°-90° over oceans for both hemispheres. Cyclones must have a minimum ocean coverage to be considered, and this removes Mediterranean cyclones which have too much land within the composite (although removing these cyclones didn't change our results qualitatively). As discussed on line 25 page 12, all cyclones are flipped so that north, or up, is poleward. We have added a figure illustrating this. While we recognize that it is potentially confusing to use the subtext SW, we think bottom-left is also confusing. The idea is to identify the WCB region, but sub WCB would be confusing, as would sub CF for cold-front, since no front rotation has been used. We have indicated the averaging region on the schematic in figure 1 that we have added.

5. Section 2.2.3: please add a couple of sentences to explain how the MERRA-2 sulfate mass is used to obtain CDNC.
Done, thank you.

6. Section 3.1.1: the section on the aquaplanet simulations is very short, and about half of this section is in fact about the relation between the moisture flux and precipitation, which is more general than just about model simulations. The main result here seems to be that regardless of resolution and aerosol concentration the relationship between moisture flux and precipitation rate is unchanged. No mention of a relationship between CDNC and liquid water path is made. Both figures associated with this section are in the supplemental material. It would be preferable to have these figures in the paper since they do illustrate the discussion.
The sections have been altered. Now the first section is on how WCB predicts CLWP. Thanks.

This would be especially useful because in fact this discussion is confusing: on the one hand, it gives the impression that the rain rate vs moisture flux relationship does not change with the aerosol concentration (or resolution). But Fig. S3 suggest that it does. To bring this matter to rest though, the figure should be clarified by either drawing a linear regression per model configuration or constraining the data points so they would have the same WCB value per

configuration. Another confusing matter is this: if WCB is kept fixed and LWP changes with aerosols, then surely precipitation should as well, no?
Linear regression for all the models has been added along with a plot of total precipitation rate. Thanks.

This is a good point about precipitation. We have tried to add some discussion to clarify this. At a short time scale we might expect precipitation to change within the cyclone in response to a transient change in CDNC, however the precipitation falling out of the cyclone will have to eventually have to match the moisture flux into the cyclone in order for mass to be conserved. To do this the LWP must adjust to a higher value so that rain rate increases. Text has been added at several points in the manuscript to clarify this.

7. Section 3.1.2: again the title of this section is misleading, it says "observed' cyclone properties and yet the very first sentence is about comparing observations with simulations. It seems that pages 9-11 are in fact part of a single section on the simulations while a new section should be when the work using the observations alone starts (second paragraph p12)
The sections have been reorganized, thanks.

8. Are the three WCB regimes of Figure 2 defined based on the observations? Two questions arise: are the three population very different in the number of members and would sampling issues affect the results? Are the distribution of WCB per region for observations and the different model configurations very different? Why not use the same color scale for all composites in Figure 2?
The placement of these regimes was defined in Fig 4a. We chose these regimes because the cyclone structure was very different in these three regimes so visually it seemed more interesting to the reader. We noted the number of members in each regime in the subplots. You are correct that terciles are a more natural binning. We changed the bins accordingly.

Apologies about the colorbars on figure 2. We set the color limits, but not the contour limits so the colorbars have the same range, but they only showed some of the ticks. We have corrected this now.

9. The discussion on why the southwest quadrant is a good place to sample for CDNC could be improved. First this quadrant is dominated by low-level clouds and so MODIS derived CDNC is probably better sampled there. It is not clear however that it would be representative of the entire cyclone, and in particular the warm sector and warm frontal zone that are dominated by high-level clouds and thus CDNC information is missing. Second the warm conveyor belt which is ingesting moisture into the cyclone tends to originate from the south east and is not always found in the southwest quadrant. So it is not clear that the "southwest quadrant is likely to be the source of moisture and aerosol for the cyclone". Figure S6 is quite important for this discussion and yet once again it is in the supplemental material. The whole discussion on how to best partition the cyclone population based on CDNC needs to be improved, it is quite confusing still. For example, it is unclear what Figure 4 is really telling us.
Figure 4 is just there to help visualize the number and distribution of cyclones and where the different $\mathrm{CDNC_{SW}}$ and WCB populations are demarcated. It's not a key figure, but it seemed helpful for readers to orient themselves.

We have moved figure S6 into the main text.

We acknowledge that the conveyor belt does not always originate in the south-west (poleward oriented), but this appears to be the case in the majority of cases and in the literature (Eckhardt et al., 2004;Naud et al., 2012). We have shown the sensitivity of our analysis to this assumption in the SM. This is one possible explanation of why our regression model doesn't have a higher explained variance, and we have included this discussion in the MS. We can also see that SO4 is higher in this quadrant, indicating that the aerosol ingestion predicted by MERRA2 is in this quadrant. As you suggest, moving this to the main text would be useful.

10. How significant is the separation in Figures 3a and 3b? based on this figure and the tests presented in Figs S7, S8, and S9, it seems that the separation is best for the southwest quadrant possibly because this is where low-level clouds dominate and ice contamination in the satellite observations is less. It seems that the results really target this specific quadrant and that little is known of the clouds that are found in other parts of the cyclones.
The populations have overlapping standard deviations as one might expect (shaded area), but the means of the populations are different at 95% confidence based on the standard error in the mean (solid lines) and a normal distribution. We note that this result is not sensitive to whether the satellite CDNC is being used or if the inferred reanalysis CDNC (which doesn't have ice cloud contamination) is used. This is now stated more clearly. Thank you.

11. Discussion of Figure 7: here it might be worth comparing with the results of Naud et al 2017 (fig 10). Also, why not define the three WCB regimes based on terciles of the entire cyclone population? This would alleviate the small number of member issue for the 5 mm/day category?
Our results seem very consistent with the results in (Naud et al., 2017). Thank you for bringing this paper to our attention. We have added comparison with this paper. We have repeated the analysis using equal terciles.

12. Page 16, discussion on albedo effects: it is quite worrying that the difference between the MODIS and MERRA2 constrained albedo variations with aerosol are this important for low values of WCB which constitute the largest number of cyclones. This should probably be said somewhere. Unrelated: the whole discussion on albedo could probably be presented in its own subsection.
The albedo discussion has been moved to its own section. Thank you for this suggestion. Use of terciles does make the albedo difference quite a bit more pronounced when MODIS CDNC is used- it is still only really present in the top tercile of WCB moisture flux, which is interesting. However, we show that for WCB moisture flux in excess of 3 mm/day the two populations clearly diverge in respect to the cyclone-mean albedo. We speculate that this may reflect difficulties in the representation of cyclone dynamics for low moisture flux cyclones by MERRA2. MODIS is not affected by this issue.

13. I do not see how Figure 11 is showing a "stronger increase in CLWP for a given increase in CDNC_SW in more pristine storms". Either there is an error in the sentence or the caption of Figure 11 needs rewriting.
We just meant that the white lines bend more at low $\mathrm{CDNC_{SW}}$. We have tried to clarify this in the text.

14. Conclusions: last sentence. Given the observations at your disposal and the disagreements between MODIS and MERRA-2 constrained relations, I would tone down this last sentence, as I am not convinced that this is a demonstration, but rather the observation of co-variations in accordance with the expectation of the sort of effect aerosols should have (as demonstrated with the simulations though).
There is relatively minor disagreement between the relations predicted by MODIS and MERRA2 (as in Figure 3). However, we have added discussion to clarify our reasoning and make it clear that we are making a statement based on empirical analysis of observations. We find that similar results can be replicated based on an idealized model and have a theory that we think explains the mechanism, but ultimately we can't show causality as in a lab study.

Typos:
Line 21, p 3: replaced "is" with "area" between "composite" and "located"
Line 17, p9: write what "CMIP" stands for.
Line 16, p14: "extent" is misleading as this is a term often used for vertical extent. "fraction" or "cover" might be more appropriate.
Line 5, p15: add "s" to "support"
Line 21-24: this sentence is too long and is missing a verb towards the end.
Line 13, p16: replace "can be" by "can display" for example
Thank you for your careful reading of our manuscript and your comments. These typos have been fixed.

Andreae, M. O., Jones, C. D., and Cox, P. M.: Strong present-day aerosol cooling implies a hot future, Nature, 435, 1187, 2005.
Ayers, G. P., and Gras, J. L.: Seasonal relationship between cloud condensation nuclei and aerosol methanesulphonate in marine air, Nature, 353, 834-835, 1991.
Bennartz, R., and Rausch, J.: Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations, Atmos. Chem. Phys. Discuss., 2017, 1-32, 10.5194/acp-2016-1130, 2017.
Boers, R., Ayers, G. P., and Gras, J. L.: COHERENCE BETWEEN SEASONAL-VARIATION IN SATELLITE-DERIVED CLOUD OPTICAL DEPTH AND BOUNDARY-LAYER CCN CONCENTRATIONS AT A MIDLATITUDE SOUTHERN-HEMISPHERE STATION, Tellus Series B-Chemical and Physical Meteorology, 46, 123-131, 10.1034/j.1600-0889.1994.t01-1-00004.x, 1994.
Bosilovich, M., Akella, S., Coy, L., Cullather, R., Draper, C., and Gelaro, R.: MERRA-2. Initial evaluation of the climate, Tech. Rep. Ser., Global Modeling and Data Assimilation, RD Koster, ed., NASA/TM-2015-104606, 2015.
Bretherton, C. S., Wood, R., George, R. C., Leon, D., Allen, G., and Zheng, X.: Southeast Pacific stratocumulus clouds, precipitation and boundary layer structure sampled along 20° S

during VOCALS-REx, Atmos. Chem. Phys., 10, 10639-10654, 10.5194/acp-10-10639-2010, 2010.
Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B. N., Duncan, B. N., Martin, R. V., Logan, J. A., Higurashi, A., and Nakajima, T.: Tropospheric Aerosol Optical Thickness from the GOCART Model and Comparisons with Satellite and Sun Photometer Measurements, Journal of the Atmospheric Sciences, 59, 461-483, 10.1175/1520-0469(2002)059<0461:TAOTFT>2.0.CO;2, 2002.
Doelling, D. R., Loeb, N. G., Keyes, D. F., Nordeen, M. L., Morstad, D., Nguyen, C., Wielicki, B. A., Young, D. F., and Sun, M.: Geostationary Enhanced Temporal Interpolation for CERES Flux Products, Journal of Atmospheric and Oceanic Technology, 30, 1072-1090, 10.1175/jtech-d-12-00136.1, 2013.
Doelling, D. R., Haney, C. O., Scarino, B. R., Gopalan, A., and Bhatt, R.: Improvements to the Geostationary Visible Imager Ray-Matching Calibration Algorithm for CERES Edition 4, Journal of Atmospheric and Oceanic Technology, 33, 2679-2698, 10.1175/jtech-d-16-0113.1, 2016.
Eckhardt, S., Stohl, A., Wernli, H., James, P., Forster, C., and Spichtinger, N.: A 15-Year Climatology of Warm Conveyor Belts, Journal of Climate, 17, 218-237, 10.1175/1520-0442(2004)017<0218:aycowc>2.0.co;2, 2004.
Field, P. R., Brožková, R., Chen, M., Dudhia, J., Lac, C., Hara, T., Honnert, R., Olson, J., Siebesma, P., de Roode, S., Tomassini, L., Hill, A., and McTaggart-Cowan, R.: Exploring the convective grey zone with regional simulations of a cold air outbreak, Quarterly Journal of the Royal Meteorological Society, 143, 2537-2555, 10.1002/qj.3105, 2017.
Grosvenor, D. P., Field, P. R., Hill, A. A., and Shipway, B. J.: The relative importance of macrophysical and cloud albedo changes for aerosol-induced radiative effects in closed-cell stratocumulus: insight from the modelling of a case study, Atmos. Chem. Phys., 17, 5155-5183, 10.5194/acp-17-5155-2017, 2017.
Hill, A. A., Feingold, G., and Jiang, H.: The Influence of Entrainment and Mixing Assumption on Aerosol–Cloud Interactions in Marine Stratocumulus, Journal of the Atmospheric Sciences, 66, 1450-1464, 10.1175/2008jas2909.1, 2009.
Hill, A. A., Shipway, B. J., and Boutle, I. A.: How sensitive are aerosol-precipitation interactions to the warm rain representation?, Journal of Advances in Modeling Earth Systems, 7, 987-1004, 10.1002/2014MS000422, 2015.
Khairoutdinov, M., and Kogan, Y.: A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, Monthly weather review, 128, 229-243, 2000.
Koren, I., Kaufman, Y. J., Rosenfeld, D., Remer, L. A., and Rudich, Y.: Aerosol invigoration and restructuring of Atlantic convective clouds, Geophysical Research Letters, 32, 2005.
Loeb, N. G., Manalo-Smith, N., Kato, S., Miller, W. F., Gupta, S. K., Minnis, P., and Wielicki, B. A.: Angular Distribution Models for Top-of-Atmosphere Radiative Flux Estimation from the Clouds and the Earth's Radiant Energy System Instrument on the Tropical Rainfall Measuring Mission Satellite. Part I: Methodology, Journal of Applied Meteorology, 42, 240-265, 10.1175/1520-0450(2003)042<0240:admfto>2.0.co;2, 2003.
Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S., Allan, R. P., Karset, I. H. H., Kristjánsson, J. E., Oreopoulos, L., Cho, N., Lee, D., Bellouin, N., Boucher, O., Grosvenor, D. P., Carslaw, K. S., Dhomse, S., Mann, G. W., Schmidt, A., Coe, H., Hartley, M. E., Dalvi, M., Hill, A. A., Johnson, B. T., Johnson, C. E., Knight, J. R., O'Connor, F. M., Partridge, D. G., Stier, P., Myhre, G., Platnick, S., Stephens, G. L., Takahashi, H., and

Thordarson, T.: Strong constraints on aerosol–cloud interactions from volcanic eruptions, Nature, 546, 485-491, 10.1038/nature22974 http://www.nature.com/nature/journal/v546/n7659/abs/nature22974.html#supplementary-information, 2017.
Marchand, R., Ackerman, T., Smyth, M., and Rossow, W. B.: A review of cloud top height and optical depth histograms from MISR, ISCCP, and MODIS, Journal of Geophysical Research-Atmospheres, 115, D16206
10.1029/2009jd013422, 2010.
McCarty, W., Coy, L., R, G., A, H., Merkova, D., EB, S., M, S., and K, W.: MERRA-2 Input Observations: Summary and Assessment, Technical Report Series on Global Modeling and Data Assimilation, 46, 2016.
McCoy, D. T., Burrows, S. M., Wood, R., Grosvenor, D. P., Elliott, S. M., Ma, P.-L., Rasch, P. J., and Hartmann, D. L.: Natural aerosols explain seasonal and spatial patterns of Southern Ocean cloud albedo, Science Advances, 1, 2015.
McCoy, D. T., and Hartmann, D. L.: Observations of a substantial cloud-aerosol indirect effect during the 2014–2015 Bárðarbunga-Veiðivötn fissure eruption in Iceland, Geophys. Res. Lett., n/a-n/a, 10.1002/2015GL067070, 2015.
McCoy, D. T., Bender, F. A. M., Grosvenor, D. P., Mohrmann, J. K., Hartmann, D. L., Wood, R., and Field, P. R.: Predicting decadal trends in cloud droplet number concentration using reanalysis and satellite data, Atmos. Chem. Phys. Discuss., 2017, 1-21, 10.5194/acp-2017-811, 2017a.
McCoy, D. T., Bender, F. A. M., Mohrmann, J. K., Hartmann, D. L., Wood, R., and Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA and AeroCom, Journal of Geophysical Research: Atmospheres, n/a-n/a, 10.1002/2016JD026141, 2017b.
Miltenberger, A. K., Field, P. R., Hill, A. A., Rosenberg, P., Shipway, B. J., Wilkinson, J. M., Scovell, R., and Blyth, A. M.: Aerosol-cloud interactions in mixed-phase convective clouds. Part 1: Aerosol perturbations, Atmos. Chem. Phys. Discuss., 2017, 1-45, 10.5194/acp-2017-788, 2017.
Minnis, P., Sun-Mack, S., Young, D. F., Heck, P. W., Garber, D. P., Chen, Y., Spangenberg, D. A., Arduini, R. F., Trepte, Q. Z., and Smith, W. L.: CERES edition-2 cloud property retrievals using TRMM VIRS and Terra and Aqua MODIS data—Part I: Algorithms, IEEE Transactions on Geoscience and Remote Sensing, 49, 4374-4400, 2011.
Naud, C. M., Posselt, D. J., and van den Heever, S. C.: Observational Analysis of Cloud and Precipitation in Midlatitude Cyclones: Northern versus Southern Hemisphere Warm Fronts, Journal of Climate, 25, 5135-5151, 10.1175/jcli-d-11-00569.1, 2012.
Naud, C. M., Posselt, D. J., and van den Heever, S. C.: Observed Covariations of Aerosol Optical Depth and Cloud Cover in Extratropical Cyclones, Journal of Geophysical Research: Atmospheres, 122, 10,338-310,356, 10.1002/2017JD027240, 2017.
Painemal, D., and Zuidema, P.: Assessment of MODIS cloud effective radius and optical thickness retrievals over the Southeast Pacific with VOCALS-REx in situ measurements, Journal of Geophysical Research-Atmospheres, 116, D24206
10.1029/2011jd016155, 2011.
Shipway, B. J., and Hill, A. A.: Diagnosis of systematic differences between multiple parametrizations of warm rain microphysics using a kinematic framework, Quarterly Journal of the Royal Meteorological Society, 138, 2196-2211, 10.1002/qj.1913, 2012.

Tan, I., Storelvmo, T., and Zelinka, M. D.: Observational constraints on mixed-phase clouds imply higher climate sensitivity, Science, 352, 224-227, 10.1126/science.aad5300, 2016.
Xue, H., and Feingold, G.: Large-Eddy Simulations of Trade Wind Cumuli: Investigation of Aerosol Indirect Effects, Journal of the Atmospheric Sciences, 63, 1605-1622, 10.1175/jas3706.1, 2006.
Xue, H., Feingold, G., and Stevens, B.: Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection, Journal of the Atmospheric Sciences, 65, 392-406, 2008.

Aerosol-mid-latitude cyclone indirect effects in observations and high-resolution simulations

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Abstract. Aerosol-cloud interactions are a major source of uncertainty in ~~predicting 21st century climate change~~inferring the climate sensitivity from the observational record of temperature. The adjustment of clouds to aerosol is a poorly constrained aspect of these aerosol-cloud interactions. Here, we examine the response of midlatitude cyclone cloud properties to a change in cloud droplet number concentration (CDNC). ~~Using~~ Idealized experiments in ~~high~~high-resolution, convection-permitting global aquaplanet simulations with constant CDNC ~~we~~are compared to thirteen years of remote-sensing observations. Observations and idealized aquaplanet simulations agree that increased warm conveyor belt (WCB) moisture flux into cyclones is consistent with higher cyclone liquid water path (CLWP). When CDNC is increased in the simulations, the CLWP consistent with a given WCB moisture flux increases. When the observational record is partitioned into high and low CDNC cyclone populations it is found that CLWP is higher in the high CDNC population at a fixed WCB moisture flux, in agreement with the idealized model. We propose that the large-scale environment controls cyclone rain rate via the moisture flux into the cyclone; enhanced CDNC decreases precipitation efficiency; then the CLWP adjusts to maintain the rain rate dictated by the large-scale environment. If cyclones in the top and bottom tercile of CDNC are contrasted it is found that they not only have higher CLWP, but also cloud cover, and albedo. The difference in cyclone albedo between the cyclones in the top and bottom third of CDNC is observed by CERES to be between 0.018 and 0.032, which is consistent with a 4.6-8.3 Wm^{-2} in-cyclone enhancement in upwelling shortwave when scaled by annual-mean insolation. ~~The region responsible for this~~

~~brightening is spatially concurrent with enhancement in CLWP and cloud fraction in the region linked to the location of the cold front. predict that increased cloud droplet number concentration (CDNC) in midlatitude cyclones will increase cyclone liquid water path (CLWP), and albedo. Insight into how to disentangle synoptic variability from cloud microphysical variability is gained by noting that variations in warm conveyor belt (WCB) moisture flux predict a large fraction of CLWP variability. In the framework of these predictions 13 years of observations from microwave radiometers as compiled in the Multisensor Advanced Climatology Liquid Water Path (MAC-LWP) dataset, along with observations from the Moderate Resolution Imaging Spectroradiometer (MODIS), Clouds and the Earth's Radiant Energy System (CERES), and reanalysis from The Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA2) are analyzed. In keeping with the results from the convection-permitting aquaplanet simulations, cyclones that have a higher CDNC are observed to have a higher CLWP at a given WCB moisture flux.~~ Based on a regression model to observed cyclone properties~~,~~ roughly 60% of the observed variability in CLWP can be explained by CDNC and WCB moisture flux. ~~and a standard deviation change in CDNC is estimated on average to have 20-30% of the impact of a standard deviation change in WCB moisture flux on CLWP. The difference in cyclone albedo between the cyclones in the top and bottom third of observed CDNC is observed by CERES to be consistent with a 4.6-8.3 Wm^{-2} enhancement in upwelling shortwave within cyclones. The region responsible for this brightening is spatially concurrent with enhancement in CLWP and cloud fraction in the region linked to the location of the cold front. This study indicates that the aerosol-cloud indirect effects have substantially altered clouds in extratropical cyclones in response to anthropogenic aerosol.~~

1. Introduction

The degree to which the aerosol indirect effects that result from anthropogenic aerosol emissions have acted to increase planetary albedo and mask greenhouse gas warming is highly uncertain (Andreae et al., 2005;Carslaw et al., 2013;Boucher et al., 2014;Forster, 2016). Establishing how much the aerosol emitted during the 20th century has enhanced the liquid water amount and thus the albedo of midlatitude storm systems is a key step in constraining ~~predictions of 21st century warming~~the climate sensitivity inferred from the observational record.

Extratropical cyclones play an important role in not only determining midlatitude albedo, but also the transport of moisture, heat, precipitation, and momentum (Hartmann, 2015;Catto et al., 2012;Hawcroft et al., 2012;Trenberth and Stepaniak, 2003;Schneider et al., 2006). Based on observational case-studies and modelling it is known that both the synoptic-scale atmospheric processes and much smaller scale cloud microphysical processes play a role in regulating the cyclone lifecycle (Naud et al., 2016;Grandey et al., 2013;Lu and Deng, 2016;Thompson and Eidhammer, 2014;Igel et al., 2013;Lu and Deng, 2015;Zhang et al., 2007;Naud et al., 2017).

In ~~general~~general, for warm rain processes, ~~enhanced~~ enhancement in aerosol that can act as cloud condensation nuclei (CCN) should enhance cloud droplet number concentration (CDNC, the first indirect, or Twomey effect) (Nakajima et al., 2001;Charlson et al., 1992;Twomey, 1977). This effect has the potential to suppress precipitation and lead to a greater retention of liquid water within the cloud (the second indirect, lifetime, cloud adjustment, or Albrecht effect) (Albrecht, 1989;Gryspeerdt et al., 2016;Sekiguchi et al., 2003). Empirical studies have established some evidence supporting the existence of these effects in liquid clouds (Gryspeerdt et al., 2016;Quaas et al., 2008;Nakajima et al., 2001;Sekiguchi et al., 2003;McCoy et al., 2017b;McCoy et al., 2015;Meskhidze and Nenes, 2006;Toll et al., 2017;Naud et al., 2017), although it has been argued that compensating physical processes may offset these microphysical perturbations(Stevens and Feingold, 2009;Malavelle et al., 2017;Igel et al., 2013). ~~Aerosol-cloud indirect effects have not been observed in extratropical cyclones~~Covariability between aerosol optical depth (AOD) and cloud cover in extratropical cyclones has been shown by previous studies(Naud et al., 2017;Grandey et al., 2013)- supporting the idea that cloud adjustments occur in ~~midlatitude~~this regime ~~cyclones~~. Here we use global, high-resolution simulations and remote-sensing observations to indicate that aerosol-cloud interactions produce an increase in the cloud liquid water content, cloud extent, and albedo of extratropical cyclones.

In section 2 we will discuss the observations, and idealized simulations of cloud responses to changes in CDNC used to ~~disentangle~~ examine the effects of aerosols and meteorology on cloud properties. We will also discuss the ~~dispersion~~ modelling of volcanic plumes from the 2014-2015 Holuhraun eruption used to provide preliminary comparison to recent results indicating an insensitivity of cloud water content to volcanic aerosol (Malavelle et al., 2017). In section 3 we present our analysis of our idealized aquaplanet simulation and we test the hypothesis arrived at in these simulations in the observational record. In section 4 we summarize our results~~ and discuss how they relate to predicting 21st century climate change~~. ~~We note that a~~A ~~table~~list of the acronyms used in this study is provided in Table 1~~Table 1~~.

2. Methods

2.1 Cyclone compositing

Many previous studies have demonstrated the usefulness of averaging around cyclone centers to examine midlatitude behavior-including aerosol variability (Field et al., 2011;Field and Wood, 2007;Naud et al., 2016;Catto, 2016;Naud et al., 2017;Grandey et al., 2013). A variety of different techniques for locating cyclone centers and compositing around elements of cyclones exist in the literature utilizing pressure fields (Jung et al., 2006;Löptien et al., 2008;Hoskins and Hodges, 2002;Field et al., 2008); geopotential height (Blender and Schubert, 2000); and vorticity (Sinclair, 1994;Hoskins and Hodges, 2002;Catto et al., 2010). In this study we utilize the methodology described in Field and Wood (2007). This algorithm locates cyclone centers based on sea level pressure (SLP) and then composites around each center. In this study we use the same constants relating to minima, slope and concavity of SLP contours as defined by Field and Wood (2007) to locate cyclone centers. As in Field and Wood (2007) SLP is resolved at 2.5°, and each composite is 4000 km across. When cyclone compositing is performed on observations only cyclone centers with 50% or more of the composite ~~is~~ area located over ocean are considered valid. All observations that are over land are removed from the composite. Cyclones centers are located in both hemispheres, but southern hemisphere cyclone composites are shown oriented so that they have a consistent orientation with northern hemisphere cyclones (Fig. 1~~Fig. 1~~).

2.2 Observations

2.2.1 SLP

The Modern-Era Retrospective Analysis for Research and Applications version 2(Bosilovich et al., 2015) (MERRA2) daily-mean sea level pressure (SLP) was used to locate cyclone centers in the observational record from 2003-2015 using the algorithm described above. ~~(Buchard et al., 2015;McCarty et al., 2016;Randles et al., 2016;Rienecker et al., 2011)~~

2.2.2 MAC-LWP

The Multi-Sensor Advanced Climatology framework used for developing monthly cloud water products (Elsaesser et al., 2017) is adapted for use here to create diurnal-cycle corrected and bias-corrected daily datasets for total liquid water path (LWP, where path is the mass in an atmospheric column), 10-meter wind speed, and water vapor path (WVP).

One possible caveat in our analysis is that there may be cross-talk among the microwave emissions. That is to say, signal that should be attributed to wind or WVP could be attributed to LWP(Elsaesser et al., 2017). However, retrievals of WVP and wind speed have been shown to be unbiased relative to in situ observations and widespread cross-talk issues are unlikely (Mears et al., 2001;Wentz, 2015;Trenberth et al., 2005;Meissner et al., 2001;Elsaesser et al., 2017).

Because microwave radiometers must make assumptions regarding the partitioning of precipitating and non-precipitating liquid this represents a systematic uncertainty in the microwave LWP data set. To bypass this source of uncertainty we utilize the total LWP data product provided by MAC-LWP. The total LWP observations from this data set represent the precipitating and non-precipitating liquid water averaged over both cloudy- and clear-sky. In this study we define the sum of precipitating and non-precipitating LWP within the cyclone as cyclone-LWP (CLWP). It should be noted that the MERRA2 reanalysis total precipitable liquid water (the TQL data in MERRA2) was compared to the microwave CLWP as a rough indicator of how MERRA2's cyclone properties covaried with its predicted sulfate.

2.2.3 CDNC

Cloud droplet number concentration (CDNC) is the key state variable that moderates the relationship between aerosol and cloud properties such as LWP and cloud fraction (Wood, 2012). In this study we use two different data sets to describe CDNC: (1) the CDNC retrieved ~~at cloud top~~ by the Moderate Resolution Imaging Spectroradiometer (MODIS) (King et al., 2003;Nakajima et al., 2001;Grosvenor and Wood,

2014) and (2) 910 hPa sulfate mass from the MERRA2 reanalysis. Data set (2) is used to assess the robustness of our analysis in regards to any remote-sensing retrieval errors in data set (1).

Retrievals of CDNC from the MODIS instrument were performed as described in Grosvenor and Wood (2014) and are the same data evaluated in McCoy et al. (2017a). In the present study, level-2 swath data (joint product) from MODIS collection 5.1 (King et al., 2003) is filtered by removing pixels with solar zenith angles greater than 65° to eliminate problematic retrievals at a pixel-level (Grosvenor and Wood (2014). The daily-mean CDNC at 1°x1° resolution is calculated from the filtered level 2 swath data and only low (cloud tops below 3.2 km), liquid clouds were used to calculate CDNC. Only 1°x1° regions where the cloud fraction exceeds 80% are considered valid (Bennartz et al., 2011) and the CDNC is calculated using the 3.7µm MODIS channel effective radius.

The second estimate of CDNC is provided by MERRA2 using sulfate (SO_4) mass. Previous studies have shown that MERRA2 sulfate mass is a good predictor of CDNC as observed by MODIS (McCoy et al., 2017a;McCoy et al., 2017b). The relationship used in the present study to calculate CDNC from 910hPa sulface mass is $\log_{10} CDNC = 0.41 \log_{10} SO4 + 2.1$, where CDNC is in units of cm^{-3} and SO4 is in units of $\mu g/m^3$. Since MERRA2 aerosol assimilation does not ingest MODIS cloud properties the CDNC from MODIS should not influence MERRA2 sulfate(Randles et al., 2016). One caveat to using MERRA2 sulfate as a proxy for CDNC when investigating ~~the~~ cloud-aerosol ~~lifetime~~ adjustments~~effect~~ is that MERRA2 does ingest microwave-observed rain rates up until 2009 and clear-sky microwave WVP into its reanalysis(McCarty et al., 2016). The possible influence of the assimilation of these cloud and meteorological properties into the MERRA2 reanalysis are evaluated in section 3. It should be noted that support for the usefulness of this data product has been provided by studying long-term trends related to volcanism and pollution controls. These have shown consistency between MODIS CDNC and sulfate mass from MERRA2 as well as observations of boundary-layer sulfur dioxide from the ozone monitoring instrument (OMI)(McCoy et al., 2017a).

These two datasets use independent approaches to estimate CDNC and will not be subject to the same errors in representing the true cloud microphysical state. Using estimate of CDNC from these two sources will yield insight into the observational uncertainty surrounding CDNC.

2.2.5 Albedo and cloud fraction from CERES

The analysis presented in this work focuses on changes in liquid water in cyclones. However, changes in the cloud fraction (CF) and the all-sky albedo are central in the evaluation of the forcing related to cloud adjustments to aerosol. ~~Finally, we~~We ~~examine changes in cyclone albedo cloud fraction (CF) and albedo.due to changes in cloud properties.~~ We utilize observed albedo and CF from the CERES 3-hourly observations, where the all-sky albedo is for clear and cloudy regions. The decision to use all-sky albedo has been made to parallel previous studies (Bender et al., 2017;Bender et al., 2016;Engstrom et al., 2015b;Engstrom et al., 2015a) and has the benefit of not being sensitive to ~~the~~ thresholding ~~in to only consider~~in the same way that cloud property retrievals are(Marchand et al., 2010). If we used an in-cloud albedo this would mean that only confidently cloudy pixels would be considered. ~~and potentially~~By using an all-sky albedo this allows~~ing~~ ~~examination~~ consideration of the contributions of broken and sub-pixel cloud cover to albedo.~~,~~ ~~which These~~Broken cloud ~~clouds~~ are~~is~~ a prominent feature in midlatitude cyclones and ~~has~~ have the ability to substantially influence ~~their~~ all-sky ~~brightness~~ albedo (McCoy et al., 2017c). One important caveat to this methodology is that we cannot partition the albedo change ~~is due~~into ~~to~~ the direct effect of aerosols, the first indirect effect, and adjustments. To offer an estimate of the change due to the direct effect we examine the CERES clear-sky albedo.~~(Naud et al., 2017)~~

The 3-hourly data ~~albedo~~ is averaged to create a daily-mean albedo and CF. ~~This~~ CF, clear-sky albedo, and all-sky albedo ~~data is~~are provided in the CERES SYN1DEG data set edition 4(Wielicki et al., 1996;Doelling et al., 2013;Doelling et al., 2016). ~~Cloud fraction (~~ CF is calculated~~)~~ from MODIS and geostationary satellites based on the ~~provided in the CERES SYN1DEG~~Minnis et al. (2011) ~~data set~~cloud mask. It is used in the calculation of the albedo retrieved by CERES as described in Doelling et al. (2016) to create an angular distribution model and to interpret geostationary observations of albedo in relation to the observations from CERES. ~~were used to examine total cloud cover and were averaged in the same way as albedo to create a daily mean cloud fraction.~~It should be noted that ~~(Marchand et al., 2010)~~without utilizing a satellite simulator(Bodas-Salcedo et al., 2011) we cannot directly compare ~~this~~cloud fraction ~~quantity~~ to the aquaplanet simulations presented in this work.

To calculate the shortwave (SW) forcing that is consistent with ~~changing~~ albedo differences we need to know the downwelling SW. ~~the average insolation in the midlatitudes was calculated.~~ Mean solar insolation (30°-80°) was calculated using the CERES EBAF-TOA edition 4 data set(Loeb et al., 2009). This quantity was used to estimate the change in reflected SW from the difference in albedo.

The dependence of albedo on solar zenith angle (SZA) is well-documented and needs to be either removed or treated in order to contrast variations in albedo generated by clouds across latitudes and seasons (Bender et al., 2017). The dependence of albedo on cloud fraction and SZA in 3-hourly CERES data is shown in Fig. 2~~Fig. 2~~. Above a SZA of 45° the albedo depends strongly on SZA. While this is a real effect of low sun angles, we are more interested in understanding the albedo of cyclones without the SZA effect. To mitigate this effect we remove observations where SZA exceeds 45° from the 3-hourly observations. To examine sensitivity to this cutoff we also utilize SZA cutoffs of 30° and 60°. The effect of these different cutoffs on the dependence of albedo on CF is shown in Fig. S2.

2.3 Models and Simulations

2.3.1 Aquaplanet

Two sets of simulations in the MetOffice Unified Model (UM) vn10.3 based on GA6 (Walters et al., 2017) were created to test the sensitivity of ~~the~~ cloud adjustment to changes in CDNC ~~cloud-aerosol indirect effect~~ to model resolution in an idealized aquaplanet setting. ~~These~~ The simulations were performed in~~models were~~ a GCM-surrogate ~~model~~ setting and a convection-permitting ~~model~~setting. The GCM-surrogate model provides a comparison to the resolution of a typical GCM and was run at 1.89°x1.25° horizontal resolution. It incorporated a parameterized convection scheme, but no cloud-scheme was implemented meaning that only convective and large-scale clouds were simulated. The convection-permitting model was run at 0.088°x0.059° and neither convection parametrization nor cloud scheme were used. It is accepted that using this resolution (roughly 6.8km in the midlatitudes) does put the convection-permitting simulation within the convective ‘Grey Zone’. The use of simulations at this resolution presents both benefits and drawbacks. Without convection being parameterized microphysics and aerosol explicitly interact at the model resolution allowing the cloud system to evolve in terms of changes to the rain and the anvils of the convection as well as cloud-to-cloud interactions mediated via cold pools and modifications to the thermodynamic and moisture profiles. However, while we are able to afford global aquaplanet runs at this resolution, it is not sufficiently finely resolved to completely resolve convection (as noted above) and this may lead to unknown errors in the simulations. We acknowledge these potential shortcomings.~~,~~ ~~but note that~~However, our results are able to probe process-~~–~~related interactions in a way that parametrized convection simulations are structurally incapable of. ~~In terms of the mean field response of explicit convection carried out at different grid resolutions~~ I~~i~~ntercomparison of simulations at scales ranging from 1-16km show minimal change to the mean statistics of simulated cloud fields (Field et al., 2017). ~~giving~~ This gives us some confidence that our results will not just be a product of the resolution of the simulations. Overall, we find in the following sections that both GCM-surrogate and convection permitting simulations increase CLWP as aerosol increases. The response of CLWP to aerosol in the convection-permitting simulation is more pronounced than the GCM-surrogate simulation, but does not contradict it.

Both convection-permitting and GCM-surrogate simulations were run with 70 vertical levels. The Cloud-AeroSol Interacting Microphysics (CASIM) two-moment microphysics scheme (Hill et al., 2015;Shipway and Hill, 2012;Grosvenor et al., 2017;Miltenberger et al., 2017) was used for all clouds in the convection-permitting simulation and for large-scale cloud cover in the GCM-surrogate simulation. The CASIM microphysics scheme is described in Shipway and Hill (2012). The warm rain processes in CASIM is compared to other microphysics schemes in Hill et al. (2015). The cloud physics parameterization used in CASIM is described in Khairoutdinov and Kogan (2000). Convective clouds in the GCM-surrogate simulation do not parameterize aerosol-cloud interactions. This is consistent with most~~Most~~ operational climate and global numerical weather prediction models ~~do not include aerosol-aware convection~~(Boucher et al., 2014)~~, as is the case in the UM operational climate model~~.

Sea surface temperature (SST) was held fixed in the simulations and the atmosphere was allowed to spin up for a week at low resolution and then for another week at high resolution. The SST profile used in the aquaplanet was derived from a 20-year climatology run from the UM in standard climate model configuration. The January SST was averaged with a north-south reflected version of itself and then zonally averaged to provide a symmetrical SST.

The aerosol profile in the control simulation was held constant at 100 cm^{-3} in the accumulation mode at the surface up until 5km and then exponentially decreased after 5km with an e-folding of 1 km. Aerosol-~~–~~cloud interactions were parameterized using a simple Twomey-type parameterization(Rogers and Yau, 1989) with $CDNC = 0.5N_{acc}w^{0.25}$ with N_{acc} being accumulation mode aerosol number concentration and w being updraft velocity limited such that at w=16m/s CDNC= N_{acc}~~N~~.~~,~~ ~~and~~ The vertical velocity was set to have~~w~~ ~~has~~ a minimum value of 0.1m/s. Aerosol was held constant throughout the simulation. The effects of enhanced aerosol on clouds was investigated by increasing aerosol at the surface to 2000 cm^{-3} in a channel between 30°N and 60°N (with an exponential decay after 5km with an e-folding of 1 km, as in the control simulation). Ice number was controlled using a simple temperature-dependent relationship (Cooper, 1986). Simulations were then run for 15 days. A single simulation was run at each resolution and aerosol concentration, giving a total of four simulations of 15 days each.

It is important to note that an increase in CDNC with increasing ~~CCN~~ N_{acc} is guaranteed by the activation parameterization used in these simulations. However, the intention of these simulations is to evaluate the response of macrophysical cloud properties to changes in CDNC and these aquaplanet simulations should be thought of in the context of an ~~‘fixed~~artificially constrained ~~-~~CDNC~~’~~ set of experiments as opposed to ‘fixed-CCN’ experiments. In addition to the large change in CDNC between the different sensitivity experiments a small amount of variability in CDNC is introduced by vertical velocities in excess of 0.1 m/s, as ~~noted~~ described above.

It is also important to note that the fixing of CDNC at a constant value means that precipitation does not affect CDNC via the removal of aerosol and thus CCN. The simulations presented here are intended to examine the adjustment in cyclone clouds to changes in CDNC as opposed to a change in aerosol fluxes. If aerosol were allowed to respond to precipitation we may speculate as to how this might affect the behavior of the cloud adjustment simulated by CASIM. As described in the following sections, the rain rate on a daily, cyclone-wide scale is determined by the large-scale environment. Subsequently, we may hypothesize that the feedback between aerosol, CDNC, and the rain rate is relatively weak, but we note that this assumption of fixed CDNC artificially removes this interaction pathway with the intent of understanding the adjustment in cloud properties to CDNC.

Finally, i~~I~~n the~~se~~ simulations presented in this paper we explore the response of the clouds in the UM treated by the CASIM cloud microphysics to changes in CDNC. A different cloud microphysics scheme would potentially yield a different ~~lifetime~~ adjustment to aerosol~~effect~~, but our results are unlikely to be qualitatively dependent on the simplistic activation scheme chosen here. We also acknowledge that the adjustment of cloud to aerosol in these idealized simulations will be a function of the CASIM microphysics scheme. Examination of CASIM in relation to other multi-moment schemes suggests that if the adjustment works through the warm rain process another multi-moment scheme would produce a qualitatively similar result (Hill et al., 2015). It is important to note that the simulations presented in this work include ice processes, which may affect the susceptibility of rain rate to changes in CDNC (Koren et al., 2005;Rosenfeld and Woodley, 2000). These effects may be highly dependent on the choice of microphysics scheme. Further, the representation of these effects in models is very uncertain and could substantially affect the predictions of our simulations. Another important mechanism not considered in our simulations is the dynamical feedback on aerosol-cloud interactions(Hill et al., 2009;Xue and Feingold, 2006;Xue et al., 2008). The dynamical evolution in response to changes in CDNC could change dramatically using a different microphysics scheme. We believe that our simulations are not strongly sensitive to the representation of these effects in CASIM given the resolution of the simulations and the forcing within a cyclonic system.

Overall, we present these simulations as an exploration of how clouds within cyclones respond to changes in CDNC through the warm rain process. These simulations are used to contextualize the observations and evaluate whether we may reproduce observational variability utilizing this idealized set of simulations.

2.3.2 ~~Plume~~Dispersion model simulations ~~from the eruption of~~of the 2014-2015 Holuhraun eruption in Iceland

The 2014-2015 eruption of Holuhraun in Iceland emitted a ~~massive~~large quantity of sulfur into the ~~lowermost~~ troposphere ~~the North Atlantic~~(Gettelman et al., 2015;Schmidt et al., 2015) and served as a ~~useful test~~case study of how clouds respond to changes in aerosol (McCoy and Hartmann, 2015;Malavelle et al., 2017). Because Holuhraun is in the midlatitudes it offers an ~~useful case study~~opportunity to examine how cyclone properties are altered by sulfate aerosol particles. The Numerical Atmospheric-dispersion Modelling Environment (NAME) is a Lagrangian dispersion model (Schmidt et al., 2015;Jones et al., 2007) ~~was~~that was used to ~~model~~simulate the ~~plume~~chemical conversion and dispersion of sulfur dioxide and ~~of~~ sulfate aerosol particles for the first two months of the ~~from~~ Holuhraun eruption.

Simulations were run ~~with~~using reanalysis meteorology for the eruptive period from the UM as described in Schmidt et al. (2015).

The output from the Holuhraun simulations ~~of Holuhraun's emissions during September and October 2014 were~~was used to determine which cyclones had interacted with the volcanic sulfate plume. The simulations were set-up using a time varying flux of SO_2 of 100 kt/d between 31 August 2014 and 13 September 2014 and 60 kt/d thereafter in line with observations and fluxes derived in a previous study (Schmidt et al., 2015). Emissions were distributed uniformly between 1500-3000m, consistent with ~~the~~observed ~~emission~~plume heights during September 2014(Schmidt et al., 2015). Sensitivity to emission height was tested ~~using~~by running a second simulation with emissions between 0-1500m. The near-surface sulfate mass was calculated by taking the mean over the bottom five model levels (100m-900m). This sulfate mass was ~~and then~~ used ~~. These simulations were used~~ to determine ~~whether~~which cyclones interacted with the sulfate plumes from the eruption ~~from Holuhraun had interacted with cyclones~~ during September and October of 2014.

3. Results and discussion

In this section we present observational analysis showing that mid-latitude cyclone liquid water content, cloud cover, and ultimately, albedo covary with changes in cloud droplet number concentration (CDNC). This work was motivated by a set of idealized convection-permitting experiments designed to examine how mid-latitude cyclone properties change in response to cloud microphysics. In section 3.1 we will discuss the characterization of cyclone systems in the midlatitudes and how we can stratify them in relation to the large-scale environment. In section 3.2 we will examine how meteorology determines cyclone properties and compare this dependence across our aquaplanet simulations, and observations. In section 3.3 the response of cyclones to a change in CDNC in the aquaplanet simulations will be contrasted with the covariability between CDNC and cyclone-mean properties in the observational record. In section 3.4 we will examine which parts of midlatitude cyclones differ between high and low CDNC populations and will contrast these observations with the change in cyclone structure in the aquaplanet simulations. In section 3.5 we show that the all-sky albedo in midlatitude cyclones differs between high and low CDNC populations. In section 3.6 we fit a regression model to explain cyclone liquid water path as a function of microphysics (CDNC) and meteorology (WCB moisture flux) and find we are able to explain the majority of extratropical cyclone variability by these two predictors. Finally, in section 3.7 we examine cyclones during the eruption of Holuraun utilizing dispersion modelling to examine the propagation of the volcanic plume.

3.1 ~~Observed relations between meteorology, mid-latitude cyclones, and aerosol~~Large-scale environmental controls on mid-latitude cyclones in relation to microphysical perturbations

~~3.1.1 Aquaplanet simulated mid-latitude cyclones and their response to changes in CCN~~

Compared to the meteorological drivers of cyclone formation, aerosol-cloud interactions are subtle and difficult to observe. ~~However, by using model simulations we can add or remove aerosol to disentangle aerosol-induced alterations to midlatitude storm cloud properties from the meteorology driving them. We created a suite of simulations in the MetOffice Unified Model (UM) that explores aerosol-cloud interactions as described in the methods section. Because the focus of this study is to understand maritime, midlatitude storms the model has no land surface (an aquaplanet) allowing an unbroken storm track providing more cyclones to be analyzed without the complications of landmasses on their evolution. A control simulation and enhanced aerosol simulation were run at each resolution to see how cyclones differed when aerosol was increased. In the control simulation aerosol concentration was set at a value of 100 cm^{-3} near the surface and in the enhanced aerosol simulation the accumulation mode aerosol concentration was set to 2000 cm^{-3} near the surface in the 30°N-60°N latitude band. Only liquid droplets are directly affected by the aerosol changes. For ice, number concentrations followed a simple temperature-dependent relationship, which is also not unusual of a GCM participating in the CMIP. Minimal impact is made on ice concentrations through variations to CCN (hence small changes to LW). We do not vary parametrizations that control the ice number when we vary CCN.~~

To understand the contributions of aerosol and meteorology to cyclones we need to characterize what constitutes a cyclone. Cyclone centers were identified using sea level pressure (SLP) in keeping with previous studies (Field and Wood, 2007) as described in the methods section. Cyclone centers were identified in both the northern and southern hemispheres between 30°-90° latitude over ocean. Cyclone compositing was performed to identify centers for both observed and simulated cyclones. Because microwave CLWP cannot be retrieved over land surfaces only cyclone centers with a substantial fraction of the cyclone over ocean were considered valid. A minimum ocean coverage of 50% within the 2000 km radius composite was required to include cyclone centers in our analysis. As noted in the methods section, southern hemisphere cyclones are flipped so that their orientation is consistent with cyclones in the northern hemisphere (Fig. 1~~Fig. 1~~). That is to say, the poleward half of the cyclone is shown in the top part of the composite and the equatorward half is shown in the bottom half of the composite.

Now that we have created a database of observed cyclones we need to stratify them by the large-scale environmental controls on their development in some way. Considerable research has been devoted to investigating the dependence of cyclone properties on meteorology using cyclone composites(Catto, 2016). One that has been found to be particularly useful is the so-called warm conveyor belt (WCB) metric (Field and Wood, 2007;Pfahl and Sprenger, 2016;Harrold, 1973). This relies on a simple model of cyclone development as described in Harrold (1973) and is calculated as the product of cyclone-mean wind speed and water vapor path multiplied by a constant describing the width of the warm conveyor belt as defined in Field and Wood (2007). It should be noted that cyclone-~~-~~mean here and in the rest of this article refers to an average taken within a 2000 km radius of the cyclone center. WCB moisture flux is a proxy for the moisture flux ingested by the cyclone and is a good predictor of the cyclone-mean rain rate in observations and models (Field and Wood, 2007;Field et al., 2011). These studies investigated observations and relatively low-resolution global climate model simulations.

We created a suite of simulations in the MetOffice Unified Model (UM) that is intended to explore aerosol-cloud interactions within mid-latitude cyclones, these simulations are described in the methods section in more detail. Because the focus of this study is to understand maritime, midlatitude storms the model has no land surface (an aquaplanet) allowing an unbroken storm track providing more cyclones to be analyzed without the complications of landmasses on their evolution. A control simulation and enhanced aerosol simulation were run at each resolution to see how cyclones differed when CDNC was increased. In the control simulation accumulation mode aerosol concentration was set at a value of 100 cm^{-3} near the surface and in the enhanced aerosol simulation the accumulation mode aerosol concentration was set to 2000 cm^{-3} near the surface in the 30°N-60°N latitude band. Only liquid droplets are directly affected by the aerosol changes. For ice, number concentrations followed a simple temperature-dependent relationship, which is also not unusual of a GCM participating in the climate model intercomparison project (CMIP). Minimal impact is made on ice concentrations through variations in N_{acc} (hence small changes to longwave radiation~~LW~~). We do not vary parametrizations that control the ice number when we vary N_{acc}. Examination of the results of our convection-permitting simulations show that t~~T~~he relationship between WCB flux and ~~rain~~ precipitation rate as well as rain rate is ~~appears to be relatively~~ relatively invariant as a function of model resolution ~~in the aquaplanet simulations analyzed in this study~~ and aerosol concentration (Fig. S3), although it is worth noting that the slope of the relationship between precipitation or rain rate and WCB moisture flux is somewhat shallower in the low--resolution model. Further, ~~This is also supported by analysis of other GCMs(Field et al., 2011).~~ u~~U~~se of this WCB metric is particularly useful in the context of our analysis because it can be measured by microwave radiometer allowing us to readily compare simulations and observations.

This consistency across models of varying spatial resolution and observations of real-world cyclones seems reasonable as it should be a consequence of mass conservation. ~~Essentially~~That is to say, once in equilibrium, the water mass flux that goes into the cyclone must be precipitated out. The perturbed aerosol environment reduces the efficiency of warm rain production for a given water path and therefore should lead to a higher equilibrium water path for a given mean rain rate or WCB flux. Reliable observations of ice cloud properties are not available(Jiang et al., 2012) so it is difficult to infer the importance of ice cloud in this mechanism. However, i~~I~~t is interesting to note that in the context of the aquaplanet simulations presented here the frozen water path in the cyclones did not change between control and enhanced aerosol experiments, indicating that this aerosol-cyclone indirect effect primarily acts through the warm rain process, at least within ~~these~~ our aquaplanet simulations (Fig. S4).

Casting our analysis as a function of WCB moisture flux means that we are investigating cyclone responses to changes in CDNC at a set precipitation rate. One possibility is that this framework will prove expedient to our analysis of cloud ~~lifetime effects~~adjustments to aerosol changes in cyclones given the divergence in precipitation responses in previous studies, ranging from intensification of precipitation (Zhang et al., 2007;Thompson and Eidhammer, 2014;Wang et al., 2014), to unchanged precipitation(Igel et al., 2013), or suppression of precipitation(Lu and Deng, 2016).

3.~~1.~~2 ~~Observed cyclone properties~~Comparison between observed and simulated cyclone properties and their dependence on meteorology

Before examining the response of cyclones to changes in cloud microphysics we first compare observed and simulated cyclones and their response to changes in synoptic meteorology as characterized by WCB moisture flux.

Comparison between MAC-LWP observations of cyclone-composited CLWP and aquaplanet simulations are shown in Fig. 3~~Fig. 3~~. To compare cyclone composites in similar meteorology conditions the cyclone composites are shown stratified ~~by~~ into terciles of WCB moisture flux. The terciles are determined by the observational record of WCB moisture flux and correspond to 0-2.21mm/day, 2.21-2.88 mm/day, and above 2.88 mm/day ~~into regimes of 1-3, 3-5, and 5-7 mm/day~~. The bounds on WCB from these terciles are ~~not recalculated for the~~used in the presentation of the aquaplanet simulations. ~~These regimes are selected to include weak, relatively typical and extremely large WCB moisture fluxes.~~

Overall, the simulations carried out at convection-permitting resolution and the observations show reasonable agreement in structure and some agreement in absolute value. Both the convection-permitting and GCM-surrogate simulations generally have a lower CLWP than the observations, but this is not surprising because no cloud-scheme is used in these simulations. That is to say, only supersaturations resolved at the model's resolution will produce cloud. Use of a cloud scheme would increase the CLWP and bring the simulations into better absolute agreement with observations. However, the cloud scheme would require a choice of critical relative humidity(Quaas, 2012;Grosvenor et al., 2017), which would complicate our analysis of these simulations across resolutions. The GCM-surrogate simulation has a much lower CLWP than either the convection-permitting simulations or the observations. This is also likely to be at least partially due to the lack of a cloud scheme meaning that only convection or times where the entire grid box is saturated will be cloudy. Cyclone-centric composites of MERRA2 total precipitable liquid water are shown in Fig. 3~~Fig. 3~~f and agree ~~qualitatively~~ somewhat with MAC-LWP observations, although the difference between different WCB moisture flux regimes is not as strong and the cyclones are significantly more diffuse. We will discuss the MERRA2 cyclone properties in the following sections in the evaluation of MERRA2 sulfate.

One consistent behavior observed across the aquaplanet simulations and observations in Fig. 3~~Fig. 3~~ is the enhancement in CLWP with increasing WCB moisture flux. By stratifying ~~the~~ cyclones by the WCB moisture flux we show that WCB moisture flux plays a significant role in determining the CLWP simulated in the UM at GCM-surrogate and convection-permitting resolutions and in observations. ~~by the WCB moisture flux we find~~ show ~~that WCB moisture flux plays a significant role in determining the CLWP (Fig. 4Fig. 3c).~~ As one might expect, a greater flux of moisture into the cyclone results in a larger total CLWP. Such a clear WCB-CLWP relationship provides a useful metric with which to stratify midlatitude cyclones. In this framework we can now ask: for a given WCB moisture flux, do variations in the aerosol that is active as CCN available to the cyclone and hence CDNC result in a different CLWP?

3.3 The response of the mean properties of mid-latitude cyclones to changes in cloud microphysics

As we saw in the last section, the WCB moisture flux into cyclones exerts a ~~significant~~substantial control on the amount of liquid within the cyclone and is a quantity that we may observe remotely. We ~~must~~ now ask the question: if we segregate cyclones into low CCN and high CCN populations will this behavior change?

Determining whether observed midlatitude cyclones have a higher or lower CCN available is ~~more~~ difficult~~ than comparing high and low CCN simulations in an idealized aquaplanet~~. One approach would be to use the observed cloud droplet number concentration (CDNC). This provides a good proxy for CCN (Wood, 2012), but as described in the methods section it is potentially problematic because retrieval errors relating to overlying ice cloud(Sourdeval et al., 2016), cloud heterogeneity (Grosvenor and Wood, 2014;Sourdeval et al., 2016), and low sun-angle(Grosvenor and Wood, 2014) may spuriously bias the measurements making it difficult to interpret any observed covariation between cyclone properties and CDNC. That is to say, retrieval error may be hypothesized to lead to any covariability that we discover in our analysis.

Ultimately the source of our concern in ~~regards to~~ basing our entire analysis on observed CDNC is that it may suffer from retrieval errors. To avoid these ambiguities we take a similar approach to previous studies (Boucher and Lohmann, 1995) and use both CDNC observed by MODIS and the sulfate mass concentration at the surface simulated by MERRA2 reanalysis(McCoy et al., 2017b;McCoy et al., 2015;McCoy et al., 2017a). ~~as a proxy for CCN (McCoy et al., 2017b;McCoy et al., 2015;McCoy et al., 2017a).~~ This use of the sulfate mass proxy as a ~~supporting data set~~proxy for MODIS observations of CDNC is advantageous because it is not susceptible to retrieval error and because MERRA2 does not have a parameterized cloud-aerosol indirect effec~~t, further simplifying its interpretation as a proxy of CCN.~~t. If we see a similar behavior when we use MERRA2 sulfate to stratify cyclones into low and high CDNC populations as we do when we stratify using observed CDNC then this covariability is not created by remote sensing retrieval biases.

Using the daily-~~-~~mean MERRA2 SO4 we calculate a CDNC proxy within cyclones following the relationship established in previous studies (McCoy et al., 2017b). This gives a CDNC proxy that is calculated using the MERRA2 ~~model~~reanalysis and, independently, an ~~top-down~~ observation of CDNC from MODIS. We examine whether both metrics for CDNC show similar behaviors when composited around cyclone centers.

By examining cyclone-centric composites of CDNC we see a~~A~~n enhancement in ~~proxy~~ CDNC observed by MODIS and inferred from MERRA2 ~~occurs~~ in the southwest quadrant (for a poleward-oriented composite; Fig. 1~~Fig. 1~~, and Fig. 4abc). This region ~~that~~ has been hypothesized by previous studies ~~is likely~~ to be the source of moisture and aerosol for the cyclone ~~(Fig. 4a,b,cFig. S6)~~ (Cooper et al., 2004;Naud et al., 2016;Joos et al., 2016). Based on these studies the southwest quadrant of the cyclone composited CDNC will be used to stratify cyclones by CCN and will be referred to as CDNC$_{SW}$. This region is shown in Fig. 1~~Fig. 1~~. ~~Note~~Again, we note that in this study all cyclone composites are oriented so that north is toward the pole and south is toward the equator so that northern and southern hemisphere cyclones are consistently oriented.

Because of the restrictions on what retrievals of CDNC are considered reliable (Grosvenor and Wood, 2014) large regions of the cyclone composite inhabited by ice cloud may be missing, in contrast no data is missing from MERRA2 sulfate because it is a reanalysis product. Examples of cyclone composited CDNC from MODIS, and MERRA2 are shown in Fig. 4~~Fig. 4~~ab~~Fig. S6ab~~. While MERRA2 infers enhancement in CDNC in the southwest quadrant, MODIS shows a higher CDNC in the north (or poleward, Fig. 1~~Fig. 1~~) part of the composite, which is likely due to retrieval bias at low sun angles and from heterogeneous cloud. Due to the vagaries of retrieving CDNC from space in the presence of broken or icy cloud, the cyclone--composited CDNC has quite different structures depending on whether it is observed by MODIS or whether MERRA2 SO4 is used as a proxy for CDNC.~~,~~ ~~However,but~~However, the inter-cyclone variability in both cyclone-mean CDNC and CDNC_{SW} observed by MODIS and inferred from MERRA2 are in agreement (Fig. 4~~Fig. 4Fig. S6~~d). Further, ~~and~~ when MERRA2 is sampled where MODIS can perform a retrieval (effectively removing SO4 data when overlying ice cloud is present), the pattern of CDNC within the mean cyclone composite is in better agreement (Fig. 4~~Fig. 4Fig. S6~~cd).

Using WCB moisture flux as a measure of the meteorological condition and CDNC_{SW} as a measure of CCN available to the cyclone we may evaluate the observational record and compare it to the aquaplanet simulations of ~~aerosol~~ CDNC enhancement. ~~When~~ W~~w~~e ~~compare~~examine the observational record of cyclone-mean CLWP by stratifying it into the top and bottom third of observed CDNC_{SW}. This is done separately using CDNC -inferred from MERRA2 and observed by MODIS (Fig. 5~~Fig. 5~~cd). There is a systematic separation in mean CLWP between high and low ~~CCN~~CDNC_{SW} cyclones ~~becomes apparent~~ (Fig. 6~~Fig. 6~~ab). The mean separation between cyclone populations is 12.7±0.7 g/m2 when MODIS is used to perform the partitioning, where the uncertainty is the 95% confidence interval assuming a normal distribution. When MERRA2 sulfate is used the difference is 15.3±0.58 g/m2. Is this behavior replicated by our idealized aquaplanet simulation?

We answer this question by comparing the low and high CDNC simulations and stratifying by WCB flux, as we did with the observations of cyclone properties. In this case we uniformly perturb the CDNC, as opposed to comparing populations within the observations. Simulations at convection-permitting resolution and low resolution are examined. When the simulations are stratified and compared in this way their behavior mimics the observations. That is to say, for a given WCB, higher N_{acc} translates to a higher CLWP (Fig. 6~~Fig. 6~~c). The difference between the control and CDNC-enhanced simulations is more pronounced in the convection-permitting model. This may be because in the GCM-surrogate simulation aerosol-cloud interactions are not represented for convection, while in the convection-permitting simulation aerosol-cloud interactions are treated in the same way for all cloud elements. It may also reflect the cloudier base state of convection-permitting simulation. However, it is possible that the aerosol-cloud indirect effect as simulated by traditional GCMs that do not include aerosol-aware convection is systematically too weak in the midlatitudes. This is because increased model CLWP results in enhanced reflection of shortwave radiation to space (Fig. S5), although thick ice clouds may mute the enhancement of reflected shortwave radiation.

Our hypothesis, based on our analysis of ~~our~~ idealized simulations and observed cyclones, is that *enhanced CCN should enhance CLWP in midlatitude storms for a given WCB moisture flux*. While this hypothesis is evocative, we should note a few potential caveats in our analysis.

~~One potential caveat to this approach is that the~~The first potential caveat is that it is possible that the $CDNC_{SW}$ inferred from MERRA2 sulfate has somehow been affected by the observations ingested into the MERRA2 reanalysis to create a spurious increase in sulfate in cyclones with larger CLWP, although as we have noted earlier the mechanism by which this could happen is not clear. ~~However~~To evaluate whether this can be the case we examine ~~, if the same procedure~~the total precipitable liquid predicted by MERRA2 ~~within~~composited around cyclone centers~~s~~. The total precipitable liquid is stratified by WCB moisture flux and then split into high and low $CDNC_{SW}$ populations utilizing MERRA2 predicted sulfate mass. L~~is applied to the total precipitable liquid in MERRA2 and if $CDNC_{SW}$ inferred from MERRA2 is used to stratify the MERRA2 cyclones into high and low $CDNC_{SW}$ then l~~ow $CDNC_{SW}$ cyclones have a higher CLWP than high $CDNC_{SW}$ cyclones at a fixed WCB moisture flux. That is to say, they have the opposite behavior of the observations from MAC-LWP (Fig. 6~~Fig. 6~~d). Based on this analysis we see that~~It appears that~~ MERRA2's reanalysis is not ingesting observations of cloud properties in such a way that it spuriously drives variations in the $CDNC_{SW}$ inferred from MERRA2 sulfate mass.

A second caveat to our analysis of cyclone properties as presented above is ~~It should be noted~~ that there is some sensitivity to ~~whether~~what region of the cyclone is used to characterize CDNC. If the cyclone-mean CDNC is used to stratify the cyclones ~~or~~instead ~~i~~ of $CDNC_{SW}$ ~~is used~~the separation between high and low CDNC populations changes slightly (Fig. S6~~7~~ab). ~~If the cyclone-mean CDNC is used to stratify cyclones , but t~~The mean CLWP for high CDNC cyclones is still ~~higher~~significantly higher than the CLWP for low CDNC cyclones at 95% confidence ~~if the cyclone-mean CDNC is used~~for moisture fluxes below 5 mm/day. The position of the warm conveyor belt is sometimes not in the SW quadrant. Additional sensitivity tests ~~of use of~~using the southern half of the composite and the south-east quadrant of CDNC to stratify cyclones are shown in Fig. S7~~8~~ and Fig. S8~~9~~. Only use of the south-eastern quadrant

(Fig. S8~~9~~) for stratification results in large portions of the high and low CDNC cyclone population being indistinguishable ~~within the standard error~~at 95% confidence. This is in agreement with previous studies of the moisture flux (Eckhardt et al., 2004;Naud et al., 2012)- the moisture flux is not ~~strongly~~exclusively ~~constrained to~~in the southwest (poleward oriented) quadrant, but is frequently in this region. Examination of the CDNC inferred from MERRA2 sulfate also supports the idea that aerosol is imported into the cyclone in the SW (with N being poleward, Fig. 1~~Fig. 1~~) quadrant (Fig. 4~~Fig. 4~~b). One possibility is that identification of frontal features (Naud et al., 2012) would better allow averaging around the element of the cyclone that carries ~~CCN~~aerosol into the cyclone. However, based on previous flow studies of aerosol within cyclones (Joos et al., 2016;Cooper et al., 2004;Naud et al., 2016) ~~without identification of frontal features~~ we believe that the CDNC$_{SW}$ offers a ~~better~~good overall representation of the importation of CCN into the cyclone and it will be used for the remainder of the analysis.

3.4 Differences in the structure of clouds within cyclones ~~in response~~as a function ~~to~~of microphysics

Having examined the difference in cyclone-mean properties between high and low CDNC$_{SW}$ populations we now examine differences in cyclone-centered cloud structure between these populations. The mean composite within each tercile of WCB moisture flux and in the -high and low CDNC$_{SW}$ populations are calculated and the difference between the composites is taken.

The difference in cloud properties between high and low CDNC$_{SW}$ cyclones share features between observations and modelling, primarily an increase in the MAC-LWP CLWP in the south-west sector of the cyclone (Fig. 7~~Fig. 7~~ and Fig. 8~~Fig. 8~~). ~~Naud et al. (2017)~~ This increase in MAC-LWP CLWP is particularly interesting as this is the region typically inhabited by open cellular convection trailing the cold front and a major source of error in simulated cyclone properties (Bodas-Salcedo et al., 2014;Naud et al., 2014;Bodas-Salcedo et al., 2012;McCoy et al., 2017d). Numerous studies have linked the dominance of open or closed mesoscale cellular convection to precipitation, and aerosol modulation of precipitation(Stevens et al., 2005;Feingold et al., 2015;Koren and Feingold, 2011;Rosenfeld et al., 2006;Mechem et al., 2012;Goren and Rosenfeld, 2012;Wang and Feingold, 2009a, b). Because of the tenuous nature of this cloud regime, and because they are typically precipitating it is reasonable to suspect that they will be more susceptible to aerosol-driven changes in their macrophysics than either thick frontal clouds or non-precipitating clouds. It is not the intention of our investigation to examine the complex dynamics of mesoscale cellular convection, but we have chosen our observational data sets so that they do not exclude this cloud regime, and the localization of differences in CLWP between high and low $CDNC_{SW}$ cyclone populations is suggestive given the existing literature regarding both the radiative importance of these clouds(McCoy et al., 2017c) and their relation to precipitation and aerosol(Koren and Feingold, 2011). Overall, this behavior motivates future work examining this region in higher resolution and higher complexity models that can resolve these features.

~~We answer this question by comparing the low and high CDNC simulations and stratifying by WCB flux. This shows that for a given WCB, higher CCN translates to a higher CLWP (Fig. 3c). The difference between the control and aerosol-enhanced simulations is more pronounced in the convection-permitting model. This may be because in the GCM-surrogate simulation aerosol-cloud interactions are not represented for convection, while in the convection-permitting simulation aerosol-cloud interactions are treated in the same way for all cloud elements. As a consequence, the aerosol-cloud indirect effect as simulated by traditional GCMs that do not include aerosol-aware convection may be systematically too weak in the midlatitudes. This is because increased model CLWP results in enhanced reflection of shortwave radiation to space (Fig. S5), although thick ice clouds may mute the enhancement of reflected shortwave radiation.~~

~~Our hypothesis, based on these idealized simulations, is that *enhanced CCN should enhance CLWP in midlatitude storms for a given WCB moisture flux*. In this context we now examine the observational record afforded to us by the microwave radiometer data in the MAC-LWP data set. In addition to observing CLWP, the microwave radiometer is also able to measure water vapor path and wind speed, allowing for observations of WCB moisture flux and comparison to the idealized aquaplanet simulations.~~

Examination of the differences in observed cloud ~~extent~~ coverage between high and low $CDNC_{SW}$ cyclones exhibit a similar pattern of differences to CLWP with enhanced cloud cover in the south-west quadrant of the composites in the second and third tercile of WCB moisture flux (Fig. 9~~Fig. 9~~). This feature is compellingly similar to the difference in cyclone properties between high and low aerosol optical depth cyclones as shown by Naud et al. (2017). It is interesting to note that the difference in CF between high and low CDNC$_{SW}$ cyclone populations are substantially more dependent on whether observed or inferred CDNC is being used to partition the cyclone populations. For cyclones whose WCB moisture flux is in excess of 3mm/day the low and high CDNC$_{SW}$ populations diverge in much the same way when using inferred or observed CDNC (Fig. 10~~Fig. 10~~). It is unclear why the MODIS and MERRA2 partitionings of the cyclone population do not agree for CF as well as they do for CLWP (Fig. 7~~Fig. 7~~). However, population mean CF is different between high and low CDNC$_{SW}$ populations at 95% confidence with a mean difference of 1.38%±0.49 to 1.9%±0.49, depending on which CDNC data set is used (note that the difference in cloud fraction is given in ~~absolute~~ percent cloud cover, not percentage difference).

3.5 Differences in albedo between high and low CDNC cyclones

~~It is also interesting to note that at high WCB moisture flux cyclones with high CDNC$_{SW}$ have a lower CLWP and CF in the southeast quadrant and higher CLWP and CF in the southwest quadrant. Given the relatively small number of midlatitude cyclones with WCB moisture flux greater than 5 mm/day (Fig. 4a) it is unclear if this effect is robust or a statistical artifact, but does appear regardless of which CDNC$_{SW}$ data set is used.~~

As shown above, systematic differences in cyclone coverage and liquid content seem to exist between low and high CDNC$_{SW}$ populations, but ~~to offer a more accurate prediction of 21st century climate change~~to better infer climate sensitivity using the temperature record the key variable to constrain is the change in reflected shortwave radiation due to aerosol indirect effects (Forster, 2016;Stevens, 2015;Andreae et al., 2005). The difference in cyclone-composited albedo observed by CERES between cyclones whose CDNC$_{SW}$ are in the top and bottom third of the population is shown in Fig. 11~~Fig. 11~~. When MODIS is used to partition the cyclone populations by CDNC$_{SW}$, the~~.~~ a~~A~~lbedo increases with increasing CDNC$_{SW}$ in the western side of the cyclone and is roughly consistent with the regions whose CLWP and CF increased (Fig. 7~~Fig. 7~~ and Fig. 9~~Fig. 9~~). ~~Low moisture-flux cyclones (WCB<3 mm/day)~~Cyclones in the lowest two terciles of WCB moisture flux show relatively little difference in albedo if MERRA2-inferred CDNC$_{SW}$ is used to stratify the observational record (and appear to decrease somewhat in the lowest tercile). ~~However, if MODIS observations are used to stratify the record the difference in albedo is much more pronounced across all WCB moisture flux categories.~~We may speculate that this reflects poor representation of the transport of aerosol into low moisture flux cyclones by MERRA2, but the reason for this disagreement is unclear.

We contrast this difference in all-sky albedo between cyclone populations in the real world with the difference in albedo simulated in the aquaplanet simulations at low and high resolution. ~~(McCoy et al., 2017b)(McCoy et al., 2017b) Overall, the consistency in differences in CLWP, cloud fraction, and albedo support the idea that the difference in albedo between low and high CDNC$_{SW}$ cyclones is at least partially due to changes in cloud macrophysics as opposed to purely changing cloud microphysics (CDNC) or aerosol direct effects.~~ Differences in simulated albedo between high and low ~~CCN~~ N$_{acc}$ simulations bear some general similarities to the observations (Fig. 12~~Fig. 12~~). ~~although a~~Albedo increases are much more uniform throughout the entire cyclone region. There is some localization to the southwestern portion of the composite, but the difference in composites is not clearly analogous beyond the sign of the difference. This lack of structural correspondence between simulated and observed all-sky albedo differences may reflect the extremely large difference in ~~CCN~~ N$_{acc}$ imposed on the simulations. This large difference in N$_{acc}$ may have ~~lead~~led~~ing~~ to a saturation of the ~~aerosol-cloud lifetime effect~~enhancement in CLWP by increasing CDNC. This may also reflect the lack of structure imposed on N$_{acc}$ by cyclone ~~structure~~dynamics in the simulations because N$_{acc}$ is not depleted by precipitation or advected by the large-scale flow, as it is in the observational record(Cooper et al., 2004).

~~**Marchand et al. (2010)(McCoy et al., 2017c)(McCoy et al., 2017b;Quaas et al., 2008;Bellouin et al., 2013;Gryspeerdt et al., 2017)(Bender et al., 2011;Bender et al., 2016;Engstrom et al., 2015b)**~~

Having inspected the differences in the structure of cloud properties and albedo between high and low CDNC$_{SW}$ populations we will now examine the difference in cyclone-mean albedo. To do this we first divide the high and low CDNC$_{SW}$ populations into 15 equal quantiles of WCB moisture flux. The mean albedo and the standard error in the mean ($SE=\sigma n$) (where sigma is standard deviation and n is the total number of observations) are calculated in each quantile (Fig. 13~~Fig. 13~~a). The 95% confidence interval is calculated assuming the distribution is normal. ~~The mean albedo is higher for the high CDNC$_{SW}$ population. When MODIS is used to retrieve CDNC the albedo is on average 0.03 higher in the high CDNC$_{SW}$ population. If MERRA2 inferred CDNC is used then the cyclone albedo is only higher for the high CDNC$_{SW}$ population when WCB moisture fluxes is greater than 2 mm/day.~~

To calculate the difference in mean albedo between high and low CDNC$_{SW}$ populations in each quantile the quantile-average WCB moisture flux needs to be examined. Because the mean WCB in each quantile may be slightly different for the high and low $\mathrm{CDNC}_{\mathrm{SW}}$ populations the mean and standard error for the high $\mathrm{CDNC}_{\mathrm{SW}}$ population is linearly interpolated so that the mean WCB moisture flux in each quantile ~~between~~ is the same for the low and high $\mathrm{CDNC}_{\mathrm{SW}}$ populations. For each quantile the standard error in the difference is propagated as $SE_{High-Low} = \sqrt{SE_{High}^2 + SE_{Low}^2}$. The average difference in albedo across quantiles is taken. The associated standard error in the averaged difference in albedo is calculated as $\sqrt{\sum \frac{SE_i^2}{15^2}}$. The difference and ~~standard error~~95% confidence interval in the difference between high and low $\mathrm{CDNC}_{\mathrm{SW}}$ populations as a function of WCB is shown in Fig. 13~~Fig. 13~~b.

The mean cyclone albedo is higher for the high $\mathrm{CDNC}_{\mathrm{SW}}$ population at 95% confidence. When MODIS is used to retrieve CDNC the albedo is on average 0.032±0.002 higher in the high $\mathrm{CDNC}_{\mathrm{SW}}$ population. If MERRA2 inferred CDNC is used then the cyclone albedo is only higher for the high $\mathrm{CDNC}_{\mathrm{SW}}$ population when WCB moisture fluxes is greater than 2 mm/day with an average difference of 0.018±0.002.

To calculate the difference in terms of a radiative flux the difference in albedo is multiplied by the annual mean climatological downwelling SW associated with the CERES EBAF-TOA data set between 30°-80°. It is important to note that this assumes that the cyclones being affected are randomly distributed in latitude and during the year. This may be somewhat reasonable for anthropogenic pollution, but not for biogenic aerosol sources. Specifically, planktonic sulfur sources have a substantial seasonal cycle leading to their contribution in albedo occurring during the period of maximum insolation (McCoy et al., 2015). The difference in reflected SW provided here is only intended to act as a rough guide to contextualize the change in albedo.

~~This~~ Multiplying the albedo by climatological insolation yields a difference in reflected SW between high and low $\mathrm{CDNC}_{\mathrm{SW}}$ populations of 8.30±0.31 Wm^{-2} if MODIS is used to stratify cyclones and 4.62±0.33 Wm^{-2} if MERRA2-inferred CDNC is used (Fig. 13~~Fig. 13~~b). This result does show some sensitivity to the maximum solar zenith angle considered acceptable for the 3-hourly CERES data. A maximum SZA of 30° yields values of 5.63~~58~~±0.9~~48~~ and 3.93~~4.23~~±1.18~~0.63~~ Wm^{-2} for MODIS and MERRA2-inferred CDNC, respectively (Fig. S9~~10~~)). A maximum SZA of 60° yields values of 6.28~~50~~±0.48~~27~~ Wm-2 and 2.15~~88~~±0.5~~28~~ Wm^{-2} (Fig. ~~S11~~S10). If all SZAs are included, ~~this reverses~~ the position of the low and high CDNC_{SW} cyclones are reversed (Fig. S11~~2~~). However, the inclusion of all SZAs observed by CERES includes albedos where the SZA effect dominates so low CF and low CLWP cyclones can ~~be~~ have a considerably higher albedo (Fig. S2~~1~~). Again, this effect is physical, but including the seasonal cycle and position of cyclones in our analysis via this effect makes it difficult to disentangle the very pronounced SZA effect from changes associated with changes in cloud properties. It is also worth noting that the difference in albedo estimated using MERRA2 SO4 to stratify cyclones is likely to have a larger sensitivity to the maximum SZA cutoff used because MODIS CDNC retrievals are not possible when the SZA exceeds 65° and so cyclones in winter are not considered in the analysis, while MERRA2 SO4 allows these cyclones to be examined.

~~(Bender et al., 2011;Bender et al., 2017)(Bender et al., 2017)(Fu and Liou, 1992)Naud et al. (2017)It is worth mentioning that~~ I~~i~~n this analysis we have used all-sky albedo from CERES to examine the response of cyclone albedo to changes in CDNC. This variable was chosen because it does not impose a criterion for what it considers to be a cloud when calculating the albedo, as a cloudy-sky ~~n in-cloud~~ albedo would. If we were to use in-cloud albedo this would necessitate the albedo perturbation being restricted to only confidently cloudy pixels (see Marchand et al. (2010)). For example, this ~~would~~could exclude situations where mesoscale cellular convection was occurring as these regions would not necessarily be considered cloudy. As pointed out by previous studies (McCoy et al., 2017c), these clouds may have a significant impact on all-sky albedo. ~~may well be the most sensitive to perturbations in CDNC.~~ However, use of all-sky albedo may potentially conflate aerosol ~~several non-macrophysical effects with changes in cloud liquid water content and extent~~direct effects and indirect effects.

First we provide an estimate of how much cloud fraction differences may contribute to the difference in albedo. Because cloud fraction and albedo have a fairly linear relation in the midlatitudes on a monthly time scale (Bender et al., 2011;Bender et al., 2017) we provide a calculation of the change in albedo related to changes in cloud cover. The observed midlatitude slope of the relation between albedo and fractional cloud cover of 0.4 from Bender et al. (2017) implies a change in albedo between the top and bottom third CDNC_{SW} populations of 0.005-0.007±0.002 (Fig. 10~~Fig. 10~~). As shown in Fig. 10~~Fig. 10~~, the difference in mean cloud cover between the populations is significant at 95% confidence, but it does not appear to contribute to the majority of the effect on albedo.

Changes in scattering from aerosol ~~optical depth (AOD)~~ could enhance clear-sky, and ultimately all-sky, albedo ~~all-sky~~ within cyclones. We examine the cyclone-composited ~~AOD from MERRA2~~clear-sky albedo observed by CERES~~, which is nudged to agree with MODIS when MODIS observations are available and allows the calculation of a daily mean~~. The albedo in cloud-free regions of the cyclone is 0.005 higher ~~The AOD is higher~~ in the high CDNC cyclone population if the observed CDNC from MODIS is used, but is unchanged if MERRA2-inferred CDNC is used to partition the cyclone population~~, as one would suspect, but the difference is less than 0.07 in the mean~~ (Fig. 14~~Fig. 14~~). This change in albedo implies a 1.38±0.16Wm^{-2} change in reflected SW in cloud-free regions if it is scaled by the annual-mean insolation. The change in all-sky albedo is nearly an order of magnitude larger (Fig. 13~~Fig. 13~~). Cyclone cloud cover is usually in excess of 70% (Fig. 10~~Fig. 10~~)- so this change in cloud-free albedo when averaged over cloudy and clear regions implies a relatively small contribution from the direct effect~~and this AOD difference should not translate to a significant difference in all-sky albedo~~.

Finally, changes in CDNC in cyclones should contribute ~~strongly~~ to this brightening, but based on the estimated midlatitude brightening due to changes in CDNC made in previous studies(McCoy et al., 2017b;Quaas et al., 2008;Bellouin et al., 2013;Gryspeerdt et al., 2017), it is unlikely that they contribute the entirety of the albedo difference. Overall, we provide this analysis of the difference in observed all-sky albedo to show that the high and low CDNC$_{SW}$ cyclone populations do not have the same brightness~~, despite having different cloud properties~~. This shows that in the midlatitude cyclone regime the adjustment in cloud macrophysical properties to a change in cloud microphysics does not act in such a way that it counteracts the forcing associated with the first indirect effect, as has been suggested to be the case in other regimes (Stevens and Feingold, 2009;Malavelle et al., 2017;Toll et al., 2017;Seifert et al., 2012;Seifert et al., 2015).~~(McCoy et al., 2017c)~~

3.~~1.3~~6 Regression model of CLWP

Given the pervasiveness of the relationships between CLWP, CDNC$_{SW}$, and WCB we create a simple regression model of CLWP to allow us to assess how much of the variance is explained by these parameters and the relative importance of 'meteorology' and 'aerosol'. The relationship between CLWP, WCB and CDNC$_{SW}$ shows differing behavior as a function of CDNC$_{SW}$ with a stronger increase in CLWP for a given increase in CDNC$_{SW}$ in more pristine (low CDNC$_{SW}$) storms ~~(Fig. 11, Fig. S13)~~ – that is to say, examination of the average CLWP as function of CDNC$_{SW}$ and WCB moisture fluxes shows that increasing CDNC$_{SW}$ at a fixed WCB moisture flux implies a larger increase in CLWP for CDNC$_{SW}$<100 cm^{-3} (Fig. 15~~Fig. 15~~, Fig. S1~~3~~2)~~, in keeping with Carslaw et al. (2013)~~. Using the observational record from 2003-2015 we train a regression model

$$CLWP = aWCB^{b}CDNC_{SW}^{c} - d \tag{1}$$

where WCB is in units of mm/day, CDNC is in cm^{-3}, and CLWP is in mm. Coefficients for the regression model trained using CDNC$_{SW}$ observed by MODIS and inferred from MERRA2 sulfate are shown in Table 2~~Table 2~~. The regression model explains 62%-67% of the variance in the observed CLWP. By using two predictors we are able to explain the majority of extratropical cyclone liquid water path variability.

It is interesting to consider how susceptible CLWP is to a perturbation in CDNC$_{SW}$ in the space of WCB and CDNC$_{SW}$. That is to say, what parts of the cyclone population would be more susceptible to changes in CDNC and which are effectively only sensitive to meteorology in the context of equation 1? We illustrate this by examining the response of equation 1 to typical perturbations in each predictor. In the context of this illustrative analysis a standard deviation is considered a typical perturbation. The standard deviation in WCB and CDNC$_{SW}$ are calculated across the data record. The coefficients for equation 1 shown in Table 2~~Table 2~~ are then used to calculate the change in CLWP for a standard deviation increase in WCB and CDNC$_{SW}$. This illustrates the relative importance of changes in aerosol (as exemplified by CDNC$_{SW}$) and changes in meteorological environment (as exemplified by WCB moisture flux) and is visualized in Fig. 16~~Fig. 16~~ for equation 1 trained using MODIS CDNC$_{SW}$.

Based on the simple visualization in Fig. 16~~Fig. 16~~ (and Fig. ~~S14~~ S13 if the CDNC$_{SW}$ inferred from MERRA2 is used to train the model) we can see that changes in CLWP for very pristine (CDNC$_{SW}$<60cm^{-3}), large moisture flux cyclones (WCB>4 mm/day) due to unit standard deviation perturbation in CDNC$_{SW}$ are estimated to be as large as 50% of those from a standard deviation perturbation in meteorology (WCB flux), while very polluted (CDNC$_{SW}$>120cm^{-3}), small moisture flux cyclones (WCB<2mm/day) are nearly insensitive to changes in CDNC$_{SW}$. This result is in keeping with

Carslaw et al. (2013), which demonstrated the importance of understanding low CCN regions to constrain the aerosol-cloud indirect effect. The sensitivity of our regression model to CDNC changes supports the importance of understanding CCN sources in remote, pristine regions. Averaged over the observational record, the mean relative contribution of aerosol changes to the variability in CLWP is 20% (30% if MERRA2-inferred CDNC$_{SW}$ is used) based on the observed distribution of cyclones in CDNC$_{SW}$ and WCB space. Evidently the dominant role is played by meteorology, but CDNC variability plays a non-~~neglgible~~negligible role.

3.7 Examination of the Holuhraun eruption case study ~~in relation to this study~~

Recent investigation by Malavelle et al. (2017) utilizing observations and climate model simulations showed that, despite the massive emission of sulfur dioxide by the Holuhraun fissure in Iceland during September and October of 2014 (Gettelman et al., 2015;Schmidt et al., 2015), and a detectable change in cloud microphysics (McCoy and Hartmann, 2015), cloud liquid water path and coverage did not deviate detectably from their climatological behavior. As described above, based on global observations of extratropical cyclones we infer that both cloud cover and liquid water path within cyclones adjust~~s~~ in response to changes in CDNC- a hypothesis which is consistent with the idealized modelling we have performed. Are our results consistent with the ~~extensive and careful~~ analysis presented in Malavelle et al. (2017)?

~~First, we~~We examine ~~how our key predictors of~~ cyclones ~~behavior, CDNC$_{SW}$ and WCB moisture flux, differed in~~in ~~September and October of 2014 in the~~the vicinity of Iceland (50°W-30°E and 45°N-85°N) and how September and October 2014 differed from the climatological behavior of cyclones in this region~~ from their climatological behavior~~. This region is consistent with previous modelling of trajectories originating at the Holuhraun fissure over the course of 48 hours (McCoy and Hartmann, 2015). ~~The climatological September-October WCB moisture flux and CDNC$_{SW}$ in is shown in Fig. S14, and Fig. S15. This is compared to the population mean behavior of cyclones during September and October of 2014 in the region of Iceland. This shows relatively small increases in CDNC$_{SW}$. However, not~~Not every cyclone in this region interacted with ~~sulfur~~the sulfate aerosol plumes from Holuhraun. To restrict the cyclone population to cyclones that might have been affected by the volcanic sulfate plume the NAME dispersion model was used to simulate ~~emissions~~the dispersion of both SO_2 and sulfate aerosol -from Holuhraun. The average near-surface volcanic ~~SO4~~sulfate aerosol mass predicted by NAME was calculated in the southwest quadrant of cyclones during September and October of 2014. Near-surface s~~S~~ulfate aerosol -mass concentrations in excess of 0.1 μg/m^3 were considered to indicate that a cyclone had interacted with the plume. ~~The WCB moisture flux and CDNC$_{SW}$ observed during 2014 is shown in Fig. S14 and S15 for emissions beginning at 0-1500m and 1500-3000m. In the context of the dependence of CLWP on CDNC$_{SW}$ and WCB moisture flux inferred from global observations, difference in CDNC$_{SW}$ during 2014 relative to the climatological CDNC$_{SW}$ did not infer a massive change in CLWP. This is consistent with the small change in in-cloud LWP observed by Malavelle et al. (2017).~~

Does our ability to examine cyclones during the eruptive period in relation to their WCB moisture flux reveal any additional information? If cyclones within the 50°W-30°E and 45°N-85°N study region are examined in this context it does appear that CLWP during the eruption might have been higher than the climatological mean. This is shown in Fig. 17~~Fig. 17~~. Cyclones during September and October for non-eruption years were used to train a power law fit to WCB moisture flux. The cyclones within the study region during September and October were split into different populations: non-eruption years;~~,~~ the eruption year;~~,~~ and the cyclones that dispersion modelling predicted to have interacted with volcanic sulfur. Two different emissions scenarios were considered in the dispersion model: emission heights set at 1500-3000m; and emissions set at 0-1500m. Anomalies relative to the climatological fit were calculated for each of the four cyclone populations. A t-test with and a non-parametric Wilcoxon rank-~~-~~sums test were used to calculate if the anomalies relative to WCB in the cyclone population~~s~~ differed significantly from the climatology for non-eruption years. Cyclones during September and October of 2014 were not unusual, but cyclones that the NAME dispersion model predicted to have interacted with the plume were ~~different~~anomalous at 95% confidence (Fig. 17~~Fig. 17~~). This was only the case when volcanic emissions were ~~at~~set at 1500-3000m height in the NAME dispersion model. The mean anomaly relative to climatology in this case was 6.51±4.43 g/m^2.

While evocative, these results appear to have some sensitivity to the region being considered. If cyclones within 30° latitude are considered, then the cyclones flagged by either emissions height scenario have a significantly different mean CLWP than the climatology (Fig. S14~~6~~). However, if cyclones spanning the entire latitude region 30°-90°N are considered the presence of several high WCB moisture flux, but relatively low CLWP, cyclones centered between ~~near~~ 30°N and 35°N that NAME predicts to have interacted with the plume lead to the population means no longer being different at 95% confidence (Fig. S15~~7~~).

We hypothesize that a more extensive investigation of the dispersion of sulfur from Holuhraun would allow a more conclusive identification of which cyclones really did interact with the plume. The number of possible free variables such as plume height; emissions flux from the fissure; and even the efficiency of aerosol rain-out in the dispersion model complicate this evaluation. A more complete evaluation of the Holuhraun case study in this framework is reserved for a future work.

Overall, w~~W~~e find agreement with the results presented in Malavelle et al. (2017) in that models who strongly adjust LWP in response to this eruption are likely to over-predict aerosol-cloud adjustments. However, we also find that cyclones predicted to have interacted with the volcanic plume had elevated CLWP relative to the climatological behavior of cyclones in that region. Direct comparison to Malavelle et al. (2017) is difficult because ~~we~~ the present study ~~(Malavelle et al., 2017)~~examines clouds within midlatitude cyclone systems while Malavelle et al. (2017) aggregates anti-cyclonic and cyclonic regions. It is possible that examination of more pristine, remote marine eruptions such as those shown in Gassó (2008) and examined in Toll et al. (2017) could ~~could~~ provide another useful constraint on aerosol-cloud adjustments as they would occur in a relatively low CDNC$_{SW}$ regime, which appears to be quite sensitive to perturbations in microphysics (Fig. 16~~Fig. 16~~).

~~However, by using model simulations we can add or remove aerosol to disentangle aerosol-induced alterations to midlatitude storm cloud properties from the meteorology driving them. We created a suite of simulations in the MetOffice Unified Model (UM) that explores aerosol-cloud interactions as described in the methods section. Because the focus of this study is to understand maritime, midlatitude storms the model has no land surface (an aquaplanet) allowing an unbroken storm track providing more cyclones to be analyzed without the complications of landmasses on their evolution. A control simulation and enhanced aerosol simulation were run at each resolution to see how cyclones differed when aerosol was increased. In the control simulation aerosol concentration was set at a value of 100 cm^{-3} near the surface and in the enhanced aerosol simulation the accumulation mode aerosol concentration was set to~~

~~2000 cm^{-3} near the surface in the 30°N-60°N latitude band. Only liquid droplets are directly affected by the aerosol changes. For ice, number concentrations followed a simple temperature-dependent relationship, which is also not unusual of a GCM participating in the CMIP. Minimal impact is made on ice concentrations through variations to CCN (hence small changes to LW). We do not vary parametrizations that control the ice number when we vary CCN.~~

4. Conclusions

Analysis of observed covariability between meteorology (as characterized by warm conveyor belt (WCB) moisture flux), warm cloud microphysics (as characterized by cloud droplet number concentration (CDNC)), and cyclone cloud properties is consistent with increasing CDNC leading to an increase in cyclone cloud liquid water path, fractional coverage, and ultimately albedo. While suggestive, empirical analysis of the observational record cannot prove causality. We support this analysis by performing a set of simulations where CDNC is set at high and low values. The response of CLWP to changes in CDNC in these simulations elucidates the mechanism by which this covariability may be explained and provides support for causality ~~between~~flowing from enhanced CDNC ~~and~~to enhanced CLWP. We hypothesize that rain rates are controlled by the large-scale environment as a consequence of mass conservation in the midlatitudes. When CDNC is increased a larger LWP is needed to give the same rain rate(Hill et al., 2015;Wood et al., 2009). The LWP adjusts to allow the rain rate to be equal to the moisture flux into the cyclone along the warm conveyor belt. This is hypothesized to lead to the observed covariance between CLWP and CDNC in the WCB region.

In summary, b~~B~~ased on the idealized simulations we have performed and our analysis of the observational records~~s~~ ~~shown here~~ we ~~have developed and tested the hypothesis~~propose that *enhanced CCN, leading to enhanced CDNC, –should enhance CLWP in midlatitude storms for a given WCB moisture flux*. It is possible that this effect is not constrained to midlatitude cyclones and we may speculate that clouds in other regimes whose rain rate is the same have a higher LWP with increasing aerosol.

The several elements of our study are consistent with previous modelling and observational studies. An aerosol indirect effect on the clouds in midlatitude storms has been predicted by simulations of the North Pacific (Wang et al., 2014;Joos et al., 2016), and observed in the intensification of the North Pacific storm track(Zhang et al., 2007). Naud et al. (2017) and Grandey et al. (2013) ~~examined~~ diagnosed covariability between cloud cover and aerosol optical depth in extratropical cyclones. ~~and~~ D~~d~~espite using a completely different set of observations than we utilize here, ~~these~~we agree with the results shown in these ~~studies found a consistent prediction of enhanced enhancement in cloud cover~~ with ~~with enhanced aerosol column optical depth~~studies. These regime-sorted analyse~~i~~s agree with global analysis in Gryspeerdt et al. (2016), which inferred that enhanced CCN enhanced CF in the midlatitudes. We also note that our statement that enhanced CDNC, driven by aerosol emissions, should enhance CLWP, CF, and albedo in cyclones appears to be in contradiction to the ~~multi-pronged~~ analysis conducted by Malavelle et al. (2017), which showed little response in LWP to a transient volcanic emission of sulfur from the 2014-2015 eruption of Holuhraun in Iceland. ~~However, we find that this small change in LWP is not inconsistent with the dependence of CLWP inferred in our study from 13 years of global data~~~~Malavelle et al. (2017).~~ We performed dispersion ~~modelling~~model simulations of the volcanic ~~plume~~sulfate aerosol to determine which cyclone systems ~~had interacted with the plume~~were affected by Holuhraun. This analysis indicated that affected cyclones had high CLWP given their meteorological environment~~,~~. ~~but s~~Sensitivity to assumptions regarding emissions height above the volcanic fissure;~~,~~ sulfur flux from the fissure;~~,~~ and the efficiency with which precipitation removes aerosol in the dispersion model necessitates a more complete validation of this analysis in a future work.

While we suggest that there is a measurable difference in cyclone properties that is driven by microphysical changes, most of the variability in extratropical cyclones is still driven by meteorology. A regression model representation of CLWP as a function of WCB moisture flux and CDNC in the southwest quadrant of the cyclone (CDNC$_{SW}$) explains the majority (~~approximately~~more than 60%) of observed variability in CLWP. This regression model allows us to estimate the relative importance of WCB moisture flux and CDNC$_{SW}$ to CLWP variability. The response of CLWP as inferred by the regression model to a standard deviation change in CDNC$_{SW}$ can be a significant fraction of the response to a standard deviation in WCB moisture flux when CDNC$_{SW}$ is low in pristine regions (Fig. 16~~Fig. 16~~), consistent with Carslaw et al. (2013). The average contribution of CDNC$_{SW}$ relative to WCB moisture flux to CLWP variability is estimated to be 20-30%.

~~It is worth noting that w~~While we should not expect to explain all of the variability in CLWP no matter how many predictors we use, it is likely that the explained variability in our regression model could be improved by (1) a more skillful metric for moisture flux into the cyclone, (2) a more accurate observation of CDNC$_{SW}$, or (3) additional information regarding ice and mixed-phase cloud properties. In regards to point (1): we have chosen to predict moisture flux in this way so that we may observe it utilizing microwave radiometers. In regards to points (2) and (3): we note that both of these retrievals are difficult and are likely to improve as the remote sensing community examines them in more depth. Overall, explaining the majority of extratropical cyclone liquid water path variability utilizing two predictors is a ~~a significant achievement~~useful contribution to our understanding of the midlatitudes.

Comparison of cyclone properties in the top and bottom third of the CDNC$_{SW}$ population correspond to different mean CLWP for a given WCB moisture flux, but also significant changes in cyclone cloud fraction and albedo. All-sky albedo difference between the top and bottom third of all observed CDNC$_{SW}$ is 0.018±0.002 (95% confidence) when MERRA2 reanalysis SO4 is used to infer CDNC and 0.032±0.002 when CDNC is observed by MODIS. These differences in the cyclone-mean albedo observed by CERES contribute to an in-cyclone enhancement in outgoing top of atmosphere shortwave radiation between 4.6 Wm^{-2} and 8.3 Wm^{-2} if the change in albedo is scaled by the annual-mean downwelling shortwave radiation between 30°-80° (Fig. 13~~Fig. 13~~).

The results presented here ~~we~~ suggest that cloud adjustments in midlatitude cyclones will not reduce the negative forcing resulting from the first indirect effect~~ and thus not lead us to infer a lower climate sensitivity based on the observed warming signal (Andreae et al., 2005;Forster, 2016).(Stevens and Feingold, 2009)~~ A more complete evaluation of aerosol transport into cyclones in the pre-industrial era would be necessary to offer an estimate of the forcing, but it appears that the forcing is negative in order for it to be consistent with observed covariability between microphysics and cloud properties. ~~(Andreae et al., 2005;Forster, 2016)Insight gained by the investigation of high-resolution, convection-permitting idealized global simulations has allowed us to hypothesize that cyclone liquid water path is substantially influenced by aerosol perturbations providing an important key to evaluating and~~

~~constraining the second indirect effect in models and offering a more tightly-constrained estimate of anthropogenic radiative forcing and of 21st century warming.~~

~~A regression model representation of CLWP as a function of WCB moisture flux and CDNC in the southwest quadrant of the cyclone (CDNC$_{SW}$) explains approximately 60% of observed variability in CLWP. Based on this regression model we infer that, as one would expect, meteorology (as characterized by WCB moisture flux) dominates CLWP variability. However, the response of CLWP as inferred by the regression model to a standard deviation change in CDNC$_{SW}$ can be a significant fraction of the response to a standard deviation in WCB moisture flux (Fig. 12). A mean relative contribution of CDNC$_{SW}$ variability to CLWP variability of 20-30% is estimated.~~

~~Comparison of cyclone properties in the top and bottom third of the CDNC$_{SW}$ population correspond to different mean CLWP for a given WCB moisture flux, but also significant changes in cyclone cloud fraction and albedo. Differences in the cyclone-mean albedo observed by CERES equate to an in-cyclone enhancement in outgoing top of atmosphere shortwave between 4.6 Wm^{-2} and 8.3 Wm^{-2} if the change in albedo is scaled by the annual-mean downwelling shortwave between 30°-80° (Fig. 10).~~

~~An aerosol indirect effect on the clouds in midlatitude storms has been predicted by simulations of the North Pacific (Wang et al., 2014;Joos et al., 2016), and observed in the intensification of the North Pacific storm track(Zhang et al., 2007)(Naud et al., 2017), but this is the first time it has been demonstrated using observations from across the extratropics.~~

Author contributions

DTM and PRF planned the paper and wrote the text. DTM performed data analysis and calculations. PRF created simulations in the Unified Model. DPG created the CDNC data set. BJS, AAH, and JMW created the CASIM microphysics package. GSE created the MAC-LWP dataset. AS ran the NAME ~~simulations of plume diffusion~~dispersion modelling. All authors contributed ideas and helped edit the paper.

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References

Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227-1230, 10.1126/science.245.4923.1227, 1989.

Andreae, M. O., Jones, C. D., and Cox, P. M.: Strong present-day aerosol cooling implies a hot future, Nature, 435, 1187-1190, 10.1038/nature03671, 2005.

Bellouin, N., Quaas, J., Morcrette, J. J., and Boucher, O.: Estimates of aerosol radiative forcing from the MACC re-analysis, Atmos. Chem. Phys., 13, 2045-2062, 10.5194/acp-13-2045-2013, 2013.

Bender, F. A. M., Charlson, R. J., Ekman, A. M. L., and Leahy, L. V.: Quantification of Monthly Mean Regional-Scale Albedo of Marine Stratiform Clouds in Satellite Observations and GCMs, Journal of Applied Meteorology and Climatology, 50, 2139-2148, 10.1175/jamc-d-11-049.1, 2011.

Bender, F. A. M., Engström, A., and Karlsson, J.: Factors Controlling Cloud Albedo in Marine Subtropical Stratocumulus Regions in Climate Models and Satellite Observations, Journal of Climate, 29, 3559-3587, 10.1175/jcli-d-15-0095.1, 2016.

Bender, F. A. M., Engström, A., Wood, R., and Charlson, R. J.: Evaluation of Hemispheric Asymmetries in Marine Cloud Radiative Properties, Journal of Climate, 30, 4131-4147, 10.1175/JCLI-D-16-0263.1, 2017.

Bennartz, R., Fan, J., Rausch, J., Leung, L. R., and Heidinger, A. K.: Pollution from China increases cloud droplet number, suppresses rain over the East China Sea, Geophys. Res. Lett., 38, n/a-n/a, 10.1029/2011gl047235, 2011.

Blender, R., and Schubert, M.: Cyclone tracking in different spatial and temporal resolutions, Monthly Weather Review, 128, 377-384, 2000.

Bodas-Salcedo, A., Webb, M. J., Bony, S., Chepfer, H., Dufresne, J. L., Klein, S. A., Zhang, Y., Marchand, R., Haynes, J. M., Pincus, R., and John, V. O.: COSP: Satellite simulation software for model assessment, Bulletin of the American Meteorological Society, 92, 1023-1043, 10.1175/2011BAMS2856.1, 2011.

Bodas-Salcedo, A., Williams, K. D., Field, P. R., and Lock, A. P.: The Surface Downwelling Solar Radiation Surplus over the Southern Ocean in the Met Office Model: The Role of Midlatitude Cyclone Clouds, Journal of Climate, 25, 7467-7486, 10.1175/jcli-d-11-00702.1, 2012.

Bodas-Salcedo, A., Williams, K. D., Ringer, M. A., Beau, I., Cole, J. N. S., Dufresne, J. L., Koshiro, T., Stevens, B., Wang, Z., and Yokohata, T.: Origins of the Solar Radiation Biases over the Southern Ocean in CFMIP2 Models, Journal of Climate, 27, 41-56, 10.1175/jcli-d-13-00169.1, 2014.

Bosilovich, M., Akella, S., Coy, L., Cullather, R., Draper, C., and Gelaro, R.: MERRA-2. Initial evaluation of the climate, Tech. Rep. Ser., Global Modeling and Data Assimilation, RD Koster, ed., NASA/TM-2015-104606, 2015.

Boucher, O., and Lohmann, U.: The sulfate-CCN-cloud albedo effect, Tellus B, 47, 281-300, 10.1034/j.1600-0889.47.issue3.1.x, 1995.

Boucher, O., Randall, D. A., Artaxo, P., Bretherton, C., Feingold, G., Forster, P. M., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S. C., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols
Climate Change 2013 - The Physical Science Basis, Cambridge University Press, 2014.

Carslaw, K. S., Lee, L. A., Reddington, C. L., Pringle, K. J., Rap, A., Forster, P. M., Mann, G. W., Spracklen, D. V., Woodhouse, M. T., Regayre, L. A., and Pierce, J. R.: Large contribution of natural aerosols to uncertainty in indirect forcing, Nature, 503, 67-71, 10.1038/nature12674, 2013.

Catto, J. L., Shaffrey, L. C., and Hodges, K. I.: Can climate models capture the structure of extratropical cyclones?, Journal of Climate, 23, 1621-1635, 2010.

Catto, J. L., Jakob, C., Berry, G., and Nicholls, N.: Relating global precipitation to atmospheric fronts, Geophysical Research Letters, 39, n/a-n/a, 10.1029/2012GL051736, 2012.

Catto, J. L.: Extratropical cyclone classification and its use in climate studies, Reviews of Geophysics, 54, 486-520, 10.1002/2016RG000519, 2016.

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

Cooper, O. R., Forster, C., Parrish, D., Trainer, M., Dunlea, E., Ryerson, T., Hübler, G., Fehsenfeld, F., Nicks, D., Holloway, J., de Gouw, J., Warneke, C., Roberts, J. M., Flocke, F., and Moody, J.: A case study of transpacific warm conveyor belt transport: Influence of merging airstreams on trace gas import to North America, Journal of Geophysical Research: Atmospheres, 109, n/a-n/a, 10.1029/2003JD003624, 2004.

Cooper, W. A.: Ice initiation in natural clouds, in: Precipitation Enhancement—A Scientific Challenge, Springer, 29-32, 1986.

Doelling, D. R., Loeb, N. G., Keyes, D. F., Nordeen, M. L., Morstad, D., Nguyen, C., Wielicki, B. A., Young, D. F., and Sun, M.: Geostationary Enhanced Temporal Interpolation for CERES Flux Products, Journal of Atmospheric and Oceanic Technology, 30, 1072-1090, 10.1175/jtech-d-12-00136.1, 2013.

Doelling, D. R., Haney, C. O., Scarino, B. R., Gopalan, A., and Bhatt, R.: Improvements to the Geostationary Visible Imager Ray-Matching Calibration Algorithm for CERES Edition 4, Journal of Atmospheric and Oceanic Technology, 33, 2679-2698, 10.1175/jtech-d-16-0113.1, 2016.

Eckhardt, S., Stohl, A., Wernli, H., James, P., Forster, C., and Spichtinger, N.: A 15-Year Climatology of Warm Conveyor Belts, Journal of Climate, 17, 218-237, 10.1175/1520-0442(2004)017<0218:aycowc>2.0.co;2, 2004.

Elsaesser, G. S., O'Dell, C. W., Lebsock, M. D., Bennartz, R., Greenwald, T. J., and Wentz, F. J.: The Multi-Sensor Advanced Climatology of Liquid Water Path (MAC-LWP), Journal of Climate, 0, null, 10.1175/jcli-d-16-0902.1, 2017.

Engstrom, A., Bender, F. A. M., Charlson, R. J., and Wood, R.: The nonlinear relationship between albedo and cloud fraction on near-global, monthly mean scale in observations and in the CMIP5 model ensemble, Geophys. Res. Lett., 42, 9571-9578, 10.1002/2015gl066275, 2015a.

Engstrom, A., Bender, F. A. M., Charlson, R. J., and Wood, R.: Geographically coherent patterns of albedo enhancement and suppression associated with aerosol sources and sinks, Tellus, 2015b.

Feingold, G., Koren, I., Yamaguchi, T., and Kazil, J.: On the reversibility of transitions between closed and open cellular convection, Atmos. Chem. Phys., 15, 7351-7367, 10.5194/acp-15-7351-2015, 2015.

Field, P. R., and Wood, R.: Precipitation and Cloud Structure in Midlatitude Cyclones, Journal of Climate, 20, 233-254, doi:10.1175/JCLI3998.1, 2007.

Field, P. R., Gettelman, A., Neale, R. B., Wood, R., Rasch, P. J., and Morrison, H.: Midlatitude Cyclone Compositing to Constrain Climate Model Behavior Using Satellite Observations, Journal of Climate, 21, 5887-5903, doi:10.1175/2008JCLI2235.1, 2008.

Field, P. R., Bodas-Salcedo, A., and Brooks, M. E.: Using model analysis and satellite data to assess cloud and precipitation in midlatitude cyclones, Quarterly Journal of the Royal Meteorological Society, 137, 1501-1515, 10.1002/qj.858, 2011.

Field, P. R., Brožková, R., Chen, M., Dudhia, J., Lac, C., Hara, T., Honnert, R., Olson, J., Siebesma, P., de Roode, S., Tomassini, L., Hill, A., and McTaggart-Cowan, R.: Exploring the convective grey zone with regional simulations of a cold air outbreak, Quarterly Journal of the Royal Meteorological Society, 143, 2537-2555, 10.1002/qj.3105, 2017.

Forster, P. M.: Inference of climate sensitivity from analysis of Earth's energy budget, Annual Review of Earth and Planetary Sciences, 44, 85-106, 2016.

Gassó, S.: Satellite observations of the impact of weak volcanic activity on marine clouds, Journal of Geophysical Research: Atmospheres, 113, n/a-n/a, 10.1029/2007JD009106, 2008.

Gettelman, A., Schmidt, A., and Egill Kristjansson, J.: Icelandic volcanic emissions and climate, Nature Geosci, 8, 243-243, 10.1038/ngeo2376
http://www.nature.com/ngeo/journal/v8/n4/abs/ngeo2376.html#supplementary-information, 2015.

Goren, T., and Rosenfeld, D.: Satellite observations of ship emission induced transitions from broken to closed cell marine stratocumulus over large areas, Journal of Geophysical Research: Atmospheres, 117, 2012.

Grandey, B. S., Stier, P., Grainger, R. G., and Wagner, T. M.: The contribution of the strength and structure of extratropical cyclones to observed cloud–aerosol relationships, Atmos. Chem. Phys., 13, 10689-10701, 10.5194/acp-13-10689-2013, 2013.

Grosvenor, D. P., and Wood, R.: The effect of solar zenith angle on MODIS cloud optical and microphysical retrievals within marine liquid water clouds, Atmos. Chem. Phys., 14, 7291-7321, 10.5194/acp-14-7291-2014, 2014.

Grosvenor, D. P., Field, P. R., Hill, A. A., and Shipway, B. J.: The relative importance of macrophysical and cloud albedo changes for aerosol-induced radiative effects in closed-cell stratocumulus: insight from the modelling of a case study, Atmos. Chem. Phys., 17, 5155-5183, 10.5194/acp-17-5155-2017, 2017.

Gryspeerdt, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, Journal of Geophysical Research: Atmospheres, n/a-n/a, 10.1002/2015JD023744, 2016.

Gryspeerdt, E., Quaas, J., Ferrachat, S., Gettelman, A., Ghan, S., Lohmann, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M., and Zhang, K.: Constraining the instantaneous aerosol influence on cloud albedo, Proceedings of the National Academy of Sciences, 114, 4899-4904, 10.1073/pnas.1617765114, 2017.

Harrold, T. W.: Mechanisms influencing the distribution of precipitation within baroclinic disturbances, Quarterly Journal of the Royal Meteorological Society, 99, 232-251, 10.1002/qj.49709942003, 1973.

Hartmann, D. L.: Global physical climatology, Newnes, 2015.

Hawcroft, M. K., Shaffrey, L. C., Hodges, K. I., and Dacre, H. F.: How much Northern Hemisphere precipitation is associated with extratropical cyclones?, Geophysical Research Letters, 39, n/a-n/a, 10.1029/2012GL053866, 2012.

Hill, A. A., Feingold, G., and Jiang, H.: The Influence of Entrainment and Mixing Assumption on Aerosol–Cloud Interactions in Marine Stratocumulus, Journal of the Atmospheric Sciences, 66, 1450-1464, 10.1175/2008jas2909.1, 2009.

Hill, A. A., Shipway, B. J., and Boutle, I. A.: How sensitive are aerosol-precipitation interactions to the warm rain representation?, Journal of Advances in Modeling Earth Systems, 7, 987-1004, 10.1002/2014MS000422, 2015.

Hoskins, B. J., and Hodges, K. I.: New perspectives on the Northern Hemisphere winter storm tracks, Journal of the Atmospheric Sciences, 59, 1041-1061, 2002.

Igel, A. L., van den Heever, S. C., Naud, C. M., Saleeby, S. M., and Posselt, D. J.: Sensitivity of Warm-Frontal Processes to Cloud-Nucleating Aerosol Concentrations, Journal of the Atmospheric Sciences, 70, 1768-1783, 10.1175/JAS-D-12-0170.1, 2013.

Jiang, J. H., Su, H., Zhai, C., Perun, V. S., Del Genio, A., Nazarenko, L. S., Donner, L. J., Horowitz, L., Seman, C., Cole, J., Gettelman, A., Ringer, M. A., Rotstayn, L., Jeffrey, S., Wu, T., Brient, F., Dufresne, J.-L., Kawai, H., Koshiro, T., Watanabe, M., LÉcuyer, T. S., Volodin, E. M., Iversen, T., Drange, H., Mesquita, M. D. S., Read, W. G., Waters, J. W., Tian, B., Teixeira, J., and Stephens, G. L.: Evaluation of cloud and water vapor simulations in CMIP5 climate models using NASA "A-Train" satellite observations, Journal of Geophysical Research: Atmospheres, 117, D14105, 10.1029/2011JD017237, 2012.

Jones, A., Thomson, D., Hort, M., and Devenish, B.: The U.K. Met Office's Next-Generation Atmospheric Dispersion Model, NAME III, in: Air Pollution Modeling and Its Application XVII, edited by: Borrego, C., and Norman, A.-L., Springer US, Boston, MA, 580-589, 2007.

Joos, H., Madonna, E., Witlox, K., Ferrachat, S., Wernli, H., and Lohmann, U.: Effect of anthropogenic aerosol emissions on precipitation in warm conveyor belts in the western North Pacific in winter – a model study with ECHAM6-HAM, Atmos. Chem. Phys. Discuss., 2016, 1-20, 10.5194/acp-2016-722, 2016.

Jung, T., Gulev, S. K., Rudeva, I., and Soloviov, V.: Sensitivity of extratropical cyclone characteristics to horizontal resolution in the ECMWF model, Quarterly Journal of the Royal Meteorological Society, 132, 1839-1857, 10.1256/qj.05.212, 2006.

Khairoutdinov, M., and Kogan, Y.: A new cloud physics parameterization in a large-eddy simulation model of marine stratocumulus, Monthly weather review, 128, 229-243, 2000.

King, M. D., Menzel, W. P., Kaufman, Y. J., Tanre, D., Bo-Cai, G., Platnick, S., Ackerman, S. A., Remer, L. A., Pincus, R., and Hubanks, P. A.: Cloud and aerosol properties, precipitable water, and profiles of temperature and water vapor from MODIS, Geoscience and Remote Sensing, IEEE Transactions on, 41, 442-458, 10.1109/TGRS.2002.808226, 2003.

Koren, I., Kaufman, Y. J., Rosenfeld, D., Remer, L. A., and Rudich, Y.: Aerosol invigoration and restructuring of Atlantic convective clouds, Geophysical Research Letters, 32, 2005.

Koren, I., and Feingold, G.: Aerosol–cloud–precipitation system as a predator-prey problem, Proceedings of the National Academy of Sciences, 108, 12227-12232, 10.1073/pnas.1101777108, 2011.

Loeb, N. G., Wielicki, B. A., Doelling, D. R., Smith, G. L., Keyes, D. F., Kato, S., Manalo-Smith, N., and Wong, T.: Toward Optimal Closure of the Earth's Top-of-Atmosphere Radiation Budget, Journal of Climate, 22, 748-766, 10.1175/2008jcli2637.1, 2009.

Löptien, U., Zolina, O., Gulev, S., Latif, M., and Soloviov, V.: Cyclone life cycle characteristics over the Northern Hemisphere in coupled GCMs, Climate dynamics, 31, 507-532, 2008.

Lu, Y., and Deng, Y.: Initial Transient Response of an Intensifying Baroclinic Wave to Increases in Cloud Droplet Number Concentration, Journal of Climate, 28, 9669-9677, 10.1175/jcli-d-15-0251.1, 2015.

Lu, Y., and Deng, Y.: Impact of Environmental Aerosols on a Developing Extratropical Cyclone in the Superparameterized Community Atmosphere Model, Journal of Climate, 29, 5533-5546, 10.1175/JCLI-D-16-0157.1, 2016.

Malavelle, F. F., Haywood, J. M., Jones, A., Gettelman, A., Clarisse, L., Bauduin, S., Allan, R. P., Karset, I. H. H., Kristjánsson, J. E., Oreopoulos, L., Cho, N., Lee, D., Bellouin, N., Boucher, O., Grosvenor, D. P., Carslaw, K. S., Dhomse, S., Mann, G. W., Schmidt, A., Coe, H., Hartley, M. E., Dalvi, M., Hill, A. A., Johnson, B. T., Johnson, C. E., Knight, J. R., O'Connor, F. M., Partridge, D. G., Stier, P., Myhre, G., Platnick, S., Stephens, G. L., Takahashi, H., and Thordarson, T.: Strong constraints on aerosol–cloud interactions from volcanic eruptions, Nature, 546, 485-491, 10.1038/nature22974 http://www.nature.com/nature/journal/v546/n7659/abs/nature22974.html#supplementary-information, 2017.

Marchand, R., Ackerman, T., Smyth, M., and Rossow, W. B.: A review of cloud top height and optical depth histograms from MISR, ISCCP, and MODIS, Journal of Geophysical Research-Atmospheres, 115, D16206
10.1029/2009jd013422, 2010.

McCarty, W., Coy, L., R, G., A, H., Merkova, D., EB, S., M, S., and K, W.: MERRA-2 Input Observations: Summary and Assessment, Technical Report Series on Global Modeling and Data Assimilation, 46, 2016.

McCoy, D. T., Burrows, S. M., Wood, R., Grosvenor, D. P., Elliott, S. M., Ma, P. L., Rasch, P. J., and Hartmann, D. L.: Natural aerosols explain seasonal and spatial patterns of Southern Ocean cloud albedo, Sci Adv, 1, e1500157, 10.1126/sciadv.1500157, 2015.

McCoy, D. T., and Hartmann, D. L.: Observations of a substantial cloud-aerosol indirect effect during the 2014–2015 Bárðarbunga-Veiðivötn fissure eruption in Iceland, Geophys. Res. Lett., n/a-n/a, 10.1002/2015GL067070, 2015.

McCoy, D. T., Bender, F. A. M., Grosvenor, D. P., Mohrmann, J. K., Hartmann, D. L., Wood, R., and Field, P. R.: Predicting decadal trends in cloud droplet number concentration using reanalysis and satellite data, Atmos. Chem. Phys. Discuss., 2017, 1-21, 10.5194/acp-2017-811, 2017a.

McCoy, D. T., Bender, F. A. M., Mohrmann, J. K. C., Hartmann, D. L., Wood, R., and Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA, and AeroCom, Journal of Geophysical Research: Atmospheres, 122, 1779-1796, 10.1002/2016jd026141, 2017b.

McCoy, I. L., Wood, R., and Fletcher, J. K.: Identifying Meteorological Controls on Open and Closed Mesoscale Cellular Convection Associated with Marine Cold Air Outbreaks, Journal of Geophysical Research: Atmospheres, n/a-n/a, 10.1002/2017JD027031, 2017c.

McCoy, I. L., Wood, R., and Fletcher, J. K.: Identifying Meteorological Controls on Open and Closed Mesoscale Cellular Convection Associated with Marine Cold Air Outbreaks, Journal of Geophysical Research: Atmospheres 2017d.

Mears, C., Smith, D. K., and Wentz, F. J.: Comparison of special sensor microwave imager and buoy-measured wind speeds from 1987 to 1997, Journal of Geophysical Research: Oceans, 106, 11719-11729, 2001.

Mechem, D. B., Yuter, S. E., and De Szoeke, S. P.: Thermodynamic and aerosol controls in southeast Pacific stratocumulus, Journal of the Atmospheric Sciences, 69, 1250-1266, 2012.

Meissner, T., Smith, D., and Wentz, F.: A 10 year intercomparison between collocated Special Sensor Microwave Imager oceanic surface wind speed retrievals and global analyses, Journal of Geophysical Research: Oceans, 106, 11731-11742, 2001.

Meskhidze, N., and Nenes, A.: Phytoplankton and Cloudiness in the Southern Ocean, Science, 314, 1419-1423, 10.1126/science.1131779, 2006.

Miltenberger, A. K., Field, P. R., Hill, A. A., Rosenberg, P., Shipway, B. J., Wilkinson, J. M., Scovell, R., and Blyth, A. M.: Aerosol-cloud interactions in mixed-phase convective clouds. Part 1: Aerosol perturbations, Atmos. Chem. Phys. Discuss., 2017, 1-45, 10.5194/acp-2017-788, 2017.

Minnis, P., Sun-Mack, S., Young, D. F., Heck, P. W., Garber, D. P., Chen, Y., Spangenberg, D. A., Arduini, R. F., Trepte, Q. Z., and Smith, W. L.: CERES edition-2 cloud property retrievals using TRMM VIRS and Terra and Aqua MODIS data—Part I: Algorithms, IEEE Transactions on Geoscience and Remote Sensing, 49, 4374-4400, 2011.

Nakajima, T., Higurashi, A., Kawamoto, K., and Penner, J. E.: A possible correlation between satellite-derived cloud and aerosol microphysical parameters, Geophys. Res. Lett., 28, 1171-1174, 10.1029/2000GL012186, 2001.

Naud, C. M., Posselt, D. J., and van den Heever, S. C.: Observational Analysis of Cloud and Precipitation in Midlatitude Cyclones: Northern versus Southern Hemisphere Warm Fronts, Journal of Climate, 25, 5135-5151, 10.1175/jcli-d-11-00569.1, 2012.

Naud, C. M., Booth, J. F., and Del Genio, A. D.: Evaluation of ERA-Interim and MERRA Cloudiness in the Southern Ocean, Journal of Climate, 27, 2109-2124, 10.1175/jcli-d-13-00432.1, 2014.

Naud, C. M., Posselt, D. J., and van den Heever, S. C.: Aerosol optical depth distribution in extratropical cyclones over the Northern Hemisphere oceans, Geophysical Research Letters, 43, 10,504-510,511, 10.1002/2016GL070953, 2016.

Naud, C. M., Posselt, D. J., and van den Heever, S. C.: Observed Covariations of Aerosol Optical Depth and Cloud Cover in Extratropical Cyclones, Journal of Geophysical Research: Atmospheres, 122, 10,338-310,356, 10.1002/2017JD027240, 2017.

Pfahl, S., and Sprenger, M.: On the relationship between extratropical cyclone precipitation and intensity, Geophysical Research Letters, 43, 1752-1758, 10.1002/2016GL068018, 2016.

Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol climate forcing, Journal of Geophysical Research: Atmospheres, 113, n/a-n/a, 10.1029/2007JD008962, 2008.

Quaas, J.: Evaluating the “critical relative humidity” as a measure of subgrid-scale variability of humidity in general circulation model cloud cover parameterizations using satellite data, Journal of Geophysical Research: Atmospheres, 117, n/a-n/a, 10.1029/2012JD017495, 2012.

Randles, C., AM, d. S., V, B., A, D., PR, C., V, A., H, B., EP, N., X, P., A, S., H, Y., and R, G.: The MERRA-2 Aerosol Assimilation, Technical Report Series on Global Modeling and Data Assimilation, 45, 2016.

Rogers, R., and Yau, M.: A short course of cloud physics. Pregamon, in, Oxford, 1989.

Rosenfeld, D., and Woodley, W. L.: Deep convective clouds with sustained supercooled liquid water down to-37.5 C, Nature, 405, 440, 2000.

Rosenfeld, D., Kaufman, Y. J., and Koren, I.: Switching cloud cover and dynamical regimes from open to closed Benard cells in response to the suppression of precipitation by aerosols, Atmos. Chem. Phys., 6, 2503-2511, 10.5194/acp-6-2503-2006, 2006.

Schmidt, A., Leadbetter, S., Theys, N., Carboni, E., Witham, C. S., Stevenson, J. A., Birch, C. E., Thordarson, T., Turnock, S., Barsotti, S., Delaney, L., Feng, W., Grainger, R. G., Hort, M. C., Höskuldsson, Á., Ialongo, I., Ilyinskaya, E., Jóhannsson, T., Kenny, P., Mather, T. A., Richards, N. A. D., and Shepherd, J.: Satellite detection, long-range transport, and air quality impacts of volcanic sulfur dioxide from the 2014–2015 flood lava eruption at Bárðarbunga (Iceland), Journal of Geophysical Research: Atmospheres, n/a-n/a, 10.1002/2015JD023638, 2015.

Schneider, T., Smith, K. L., O’Gorman, P. A., and Walker, C. C.: A Climatology of Tropospheric Zonal-Mean Water Vapor Fields and Fluxes in Isentropic Coordinates, Journal of Climate, 19, 5918-5933, 10.1175/jcli3931.1, 2006.

Seifert, A., Köhler, C., and Beheng, K.: Aerosol-cloud-precipitation effects over Germany as simulated by a convective-scale numerical weather prediction model, Atmospheric Chemistry and Physics, 12, 709, 2012.

Seifert, A., Heus, T., Pincus, R., and Stevens, B.: Large-eddy simulation of the transient and near-equilibrium behavior of precipitating shallow convection, Journal of Advances in Modeling Earth Systems, 7, 1918-1937, 2015.

Sekiguchi, M., Nakajima, T., Suzuki, K., Kawamoto, K., Higurashi, A., Rosenfeld, D., Sano, I., and Mukai, S.: A study of the direct and indirect effects of aerosols using global satellite data sets of aerosol and cloud parameters, Journal of Geophysical Research: Atmospheres, 108, n/a-n/a, 10.1029/2002JD003359, 2003.

Shipway, B. J., and Hill, A. A.: Diagnosis of systematic differences between multiple parametrizations of warm rain microphysics using a kinematic framework, Quarterly Journal of the Royal Meteorological Society, 138, 2196-2211, 10.1002/qj.1913, 2012.

Sinclair, M. R.: An objective cyclone climatology for the Southern Hemisphere, Monthly Weather Review, 122, 2239-2256, 1994.

Sourdeval, O., Labonnote, L. C., Baran, A. J., Mülmenstädt, J., and Brogniez, G.: A Methodology for Simultaneous Retrieval of Ice and Liquid Water Cloud Properties. Part II: Near-global Retrievals and Evaluation against A-Train Products, Quarterly Journal of the Royal Meteorological Society, 2016.

Stevens, B., Vali, G., Comstock, K., Wood, R., Zanten, M. C. V., Austin, P. H., Bretherton, C. S., and Lenschow, D. H.: POCKETS OF OPEN CELLS AND DRIZZLE IN MARINE STRATOCUMULUS, Bulletin of the American Meteorological Society, 86, 51-57, 10.1175/bams-86-1-51, 2005.

Stevens, B., and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461, 607-613, 2009.

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

Thompson, G., and Eidhammer, T.: A Study of Aerosol Impacts on Clouds and Precipitation Development in a Large Winter Cyclone, Journal of the Atmospheric Sciences, 71, 3636-3658, 10.1175/JAS-D-13-0305.1, 2014.

Toll, V., Christensen, M., Gassó, S., and Bellouin, N.: Volcano and Ship Tracks Indicate Excessive Aerosol-Induced Cloud Water Increases in a Climate Model, Geophysical Research Letters, 44, 12,492-412,500, 10.1002/2017GL075280, 2017.

Trenberth, K. E., and Stepaniak, D. P.: Covariability of Components of Poleward Atmospheric Energy Transports on Seasonal and Interannual Timescales, Journal of Climate, 16, 3691-3705, 10.1175/1520-0442(2003)016<3691:cocopa>2.0.co;2, 2003.

Trenberth, K. E., Fasullo, J., and Smith, L.: Trends and variability in column-integrated atmospheric water vapor, Climate dynamics, 24, 741-758, 2005.

Twomey, S.: INFLUENCE OF POLLUTION ON SHORTWAVE ALBEDO OF CLOUDS, Journal of the Atmospheric Sciences, 34, 1149-1152, 10.1175/1520-0469(1977)034<1149:tiopot>2.0.co;2, 1977.

Walters, D., Boutle, I., Brooks, M., Melvin, T., Stratton, R., Vosper, S., Wells, H., Williams, K., Wood, N., Allen, T., Bushell, A., Copsey, D., Earnshaw, P., Edwards, J., Gross, M., Hardiman, S., Harris, C., Heming, J., Klingaman, N., Levine, R., Manners, J., Martin, G., Milton, S., Mittermaier, M., Morcrette, C., Riddick, T., Roberts, M., Sanchez, C., Selwood, P., Stirling, A., Smith, C., Suri, D., Tennant, W., Vidale, P. L., Wilkinson, J., Willett, M., Woolnough, S., and Xavier, P.: The Met Office Unified Model Global Atmosphere 6.0/6.1 and JULES Global Land 6.0/6.1 configurations, Geosci. Model Dev., 10, 1487-1520, 10.5194/gmd-10-1487-2017, 2017.

Wang, H., and Feingold, G.: Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part II: The microphysics and dynamics of the boundary region between open and closed cells, Journal of the atmospheric sciences, 66, 3257-3275, 2009a.

Wang, H., and Feingold, G.: Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part I: Impact of drizzle on the formation and evolution of open cells, Journal of the Atmospheric Sciences, 66, 3237-3256, 2009b.

Wang, Y., Wang, M., Zhang, R., Ghan, S. J., Lin, Y., Hu, J., Pan, B., Levy, M., Jiang, J. H., and Molina, M. J.: Assessing the effects of anthropogenic aerosols on Pacific storm track using a multiscale global climate model, Proceedings of the National Academy of Sciences, 111, 6894-6899, 10.1073/pnas.1403364111, 2014.

Wentz, F. J.: A 17-yr climate record of environmental parameters derived from the Tropical Rainfall Measuring Mission (TRMM) Microwave Imager, Journal of Climate, 28, 6882-6902, 2015.

Wielicki, B. A., Barkstrom, B. R., Harrison, E. F., III, R. B. L., Smith, G. L., and Cooper, J. E.: Clouds and the Earth's Radiant Energy System (CERES): An Earth Observing System Experiment, Bulletin of the American Meteorological Society, 77, 853-868, 10.1175/1520-0477(1996)077<0853:catere>2.0.co;2, 1996.

Wood, R., Kubar, T. L., and Hartmann, D. L.: Understanding the Importance of Microphysics and Macrophysics for Warm Rain in Marine Low Clouds. Part II: Heuristic Models of Rain Formation, Journal of the Atmospheric Sciences, 66, 2973-2990, 10.1175/2009jas3072.1, 2009.

Wood, R.: Stratocumulus Clouds, Monthly Weather Review, 140, 2373-2423, 10.1175/MWR-D-11-00121.1, 2012.

Xue, H., and Feingold, G.: Large-Eddy Simulations of Trade Wind Cumuli: Investigation of Aerosol Indirect Effects, Journal of the Atmospheric Sciences, 63, 1605-1622, 10.1175/jas3706.1, 2006.

Xue, H., Feingold, G., and Stevens, B.: Aerosol effects on clouds, precipitation, and the organization of shallow cumulus convection, Journal of the Atmospheric Sciences, 65, 392-406, 2008.

Zhang, R., Li, G., Fan, J., Wu, D. L., and Molina, M. J.: Intensification of Pacific Storm Track Linked to Asian Pollution, Proceedings of the National Academy of Sciences of the United States of America, 104, 5295-5299, 2007.

Table 1 Acronyms used in this article.

Acronym	**Description**
CDNC	Cloud droplet number concentration within clouds.
$CDNC_{SW}$	Cloud droplet number concentration average in the south west quarter circle of each cyclone composite. Note that southern hemisphere cyclones are flipped so that their orientation is consistent with northern hemisphere cyclones.
CF	Cloud fraction.
CLWP	Cyclone liquid water path, defined as the sum of precipitating and non-precipitating liquid.
LWP	Liquid water path, the integrated mass concentration of liquid in a column of atmosphere.
N_{acc}	Accumulation mode aerosol number concentration.
SW	Shortwave radiation
SZA	Solar zenith angle.
WCB	Warm conveyor belt.
WVP	Water vapor path

Table 2 The coefficients –for equation 1 based on using CDNC$_{SW}$ observed by MODIS and inferred from MERRA2 sulfate. Coefficients and 95% confidence intervals (a-d) are listed for each. The number of observations used to train the model is listed as n. The correlation coefficient, r, between predicted and observed CLWP is also listed for each model.

	a	b	c	d	n	r
MODIS	21.79±1.75	0.95±0.030	0.11±0.0062	18.52±3.25	37837	0.79
MERRA2	19.23±1.28	0.86±0.021	0.19±0.0062	4.53±2.69	49361	0.82

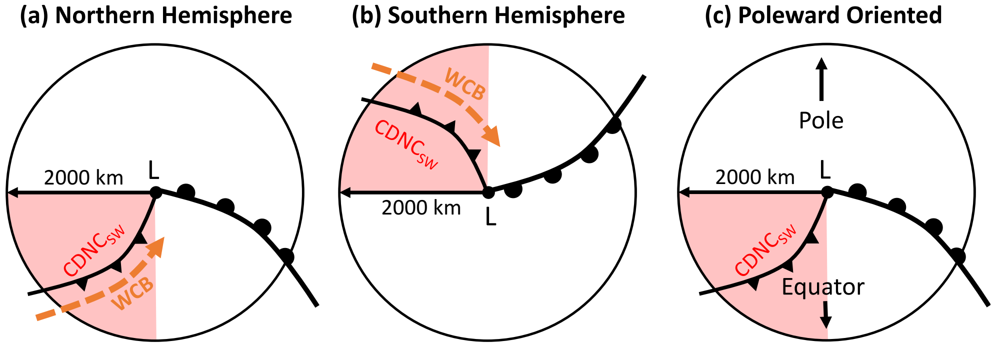


Fig. 1 Schematic illustrations of an idealized cyclone in the northern hemisphere (a), southern hemisphere (b), and flipped so they are poleward oriented (c). All cyclone composites in this study are presented in poleward oriented format. The approximate location of the cold front is shown with triangles and the warm front is shown with half-circles. The approximate warm conveyor belt (WCB) location is indicated in orange and the low is indicated with an L. The 2000 km radius of averaging is indicated. The averaging region used to calculate CDNC$_{SW}$ is shown using red shading.

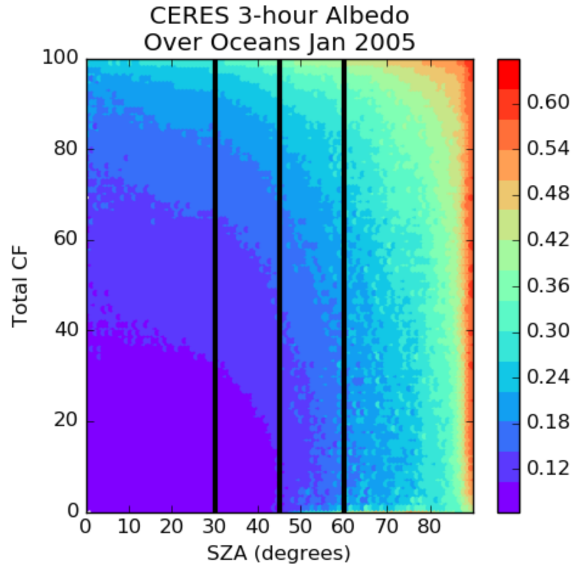
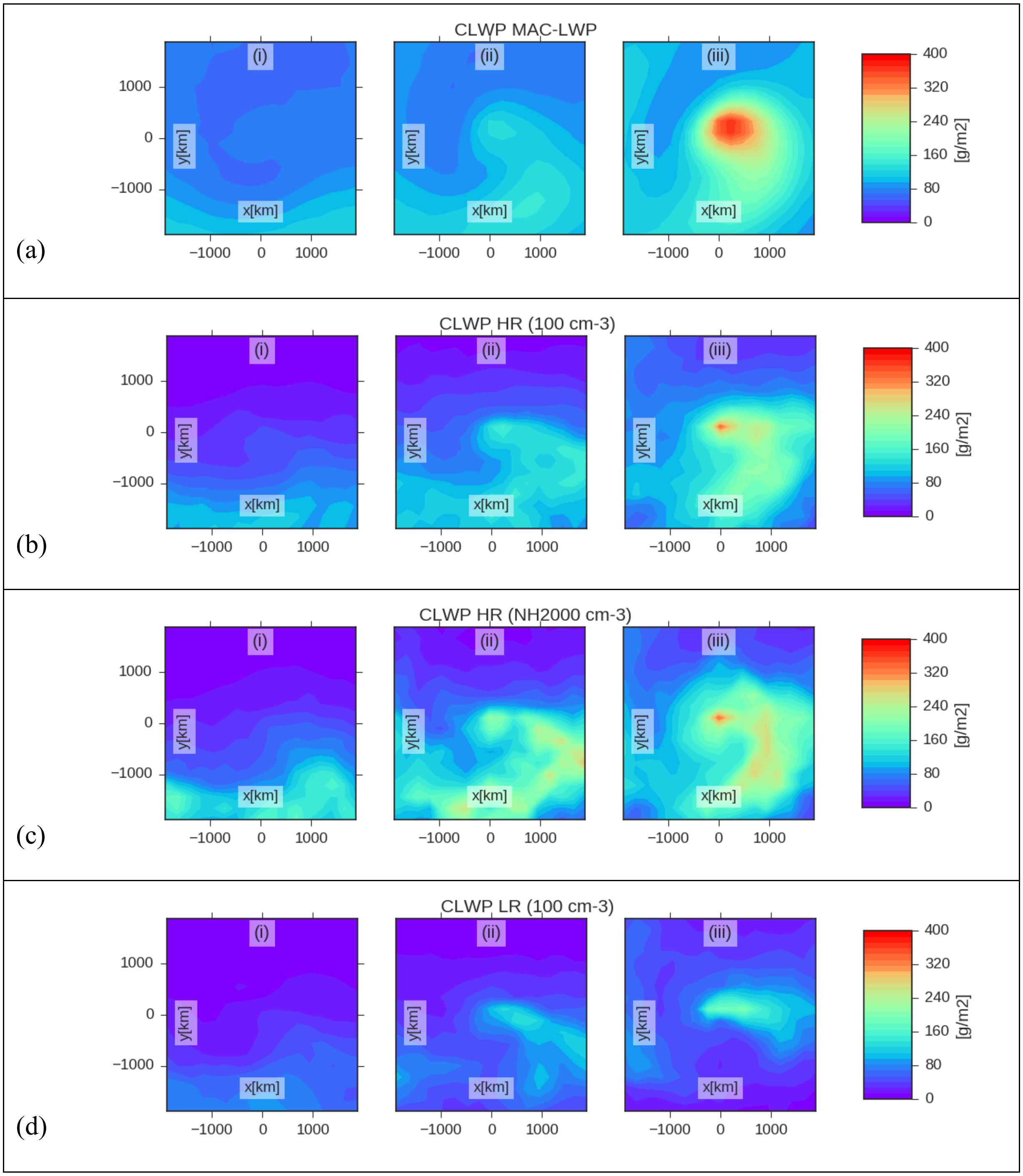


Fig. 2 CERES 3-hourly albedo over oceans binned as a function of cloud fraction and solar zenith angle (SZA) during January 2005. Above a SZA of 45° a strong dependence of albedo on SZA is seen. The SZA cut offs used in this study of 30°, 45° and 60° are shown with vertical black lines. Example CERES albedo is shown in Fig. S1.


CLWP MAC-LWP
CLWP HR (100 cm-3)
CLWP HR (NH2000 cm-3)
CLWP LR (100 cm-3)
(a)
(b)
(c)
(d)
(i)
(ii)
(iii)
y[km]
x[km]
1000
0
−1000
[g/m2]
400
320
240
160
80
0

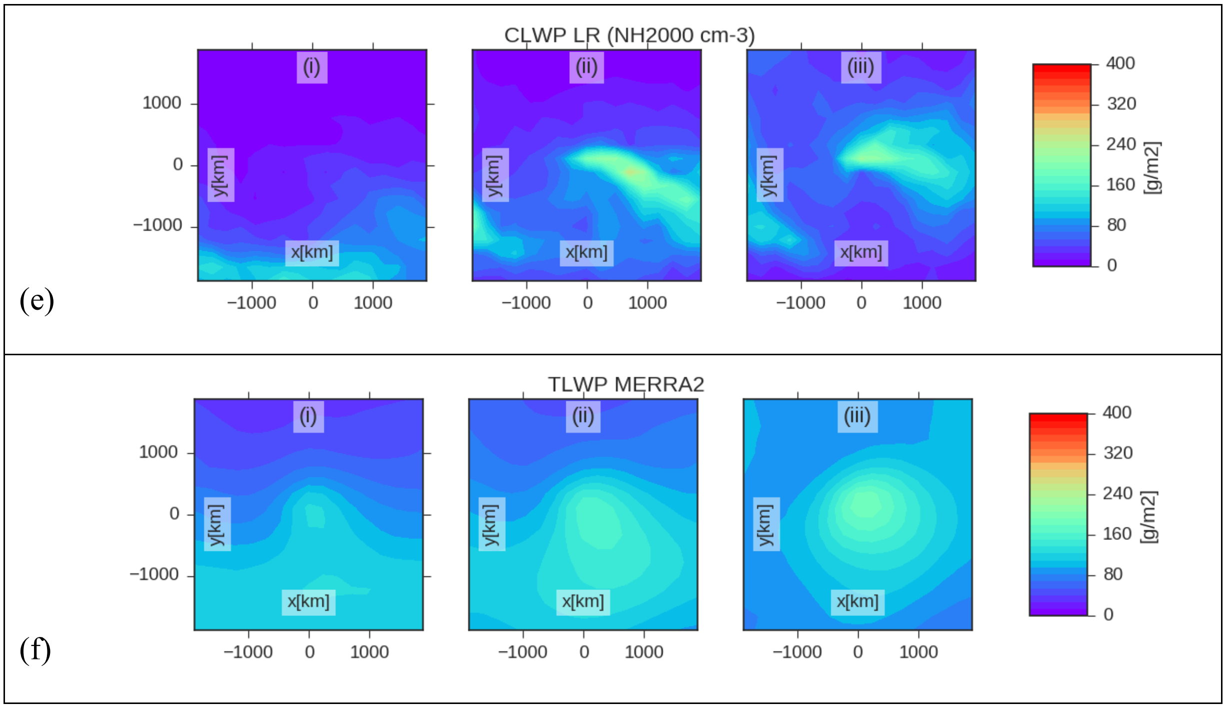


Fig. 3 Cyclone composites showing CLWP from (a) MAC-LWP, (b-c) the convection-permitting simulation in the control and enhanced ~~CCN~~ N_{acc} experiments, (d-e) the GCM-surrogate simulation in the control and enhanced ~~aerosol~~ N_{acc} experiments, and (f) MERRA2. All composites are shown in three bins of WCB moisture flux so that cyclones with similar meteorology can be compared. ~~Note that plotted ranges differ between models and observations. The number of cyclones contributing to the mean composite are noted in each subplot~~ The bins are ~~calculated using the~~terciles of observed WCB moisture flux~~ distribution and calculating the bins so that each contains an equal number of cyclones~~. Bins are shown in Fig. 5~~Fig. 5~~a and are noted in each subplot by (i)-(iii). It should be noted that the bin edges are not recalculated for the simulations~~.~~

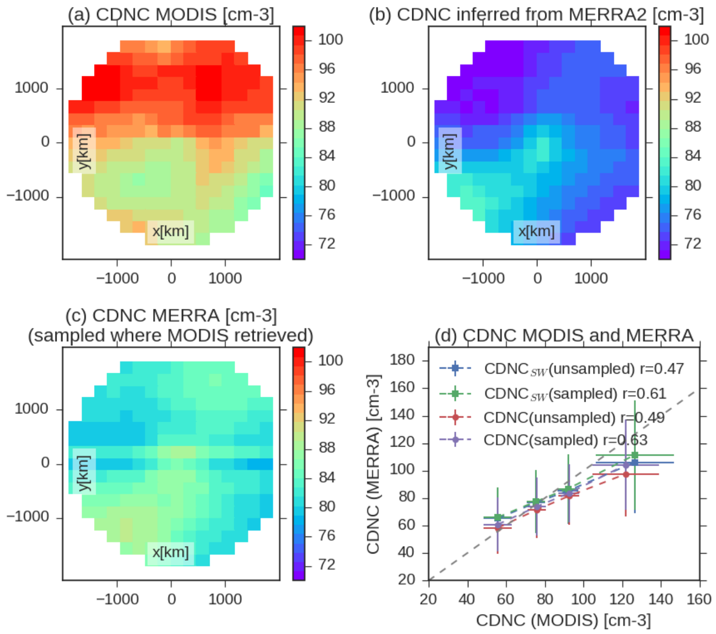


Fig. 4 Cyclone composited CDNC (a) observed by MODIS, (b) inferred from MERRA2, (c) inferred from MERRA2, but sampled so that failed retrievals from MODIS are not used in calculation of the mean composite from MERRA2. Figure (d) Shows the covariability between MERRA2-inferred and MODIS-observed cyclone-mean and southwest quadrant (poleward-oriented) CDNC (CDNC$_{SW}$). The CDNC from MERRA2 is plotted on the y-axis and is shown binned by MODIS CDNC. Error bars show one standard deviation over each bin. The correlation between the CDNC (and CDNC$_{SW}$) from MODIS and from MERRA2 for all cyclones in the observational record is noted in the legend. CDNC (and CDNC$_{SW}$) from MERRA2 is calculated when all data is used (unsampled) and when it is sampled to correspond to MODIS.

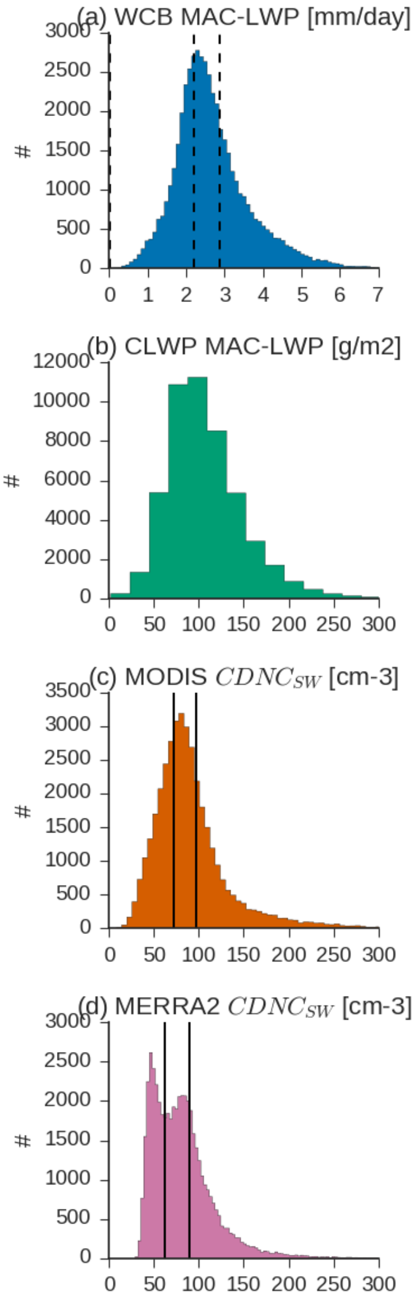


Fig. 5 Distributions of cyclone-mean properties within the 2003-2015 observational record. Units are noted for each variable. The number of composite cyclones with that value is indicated on the ordinate. Warm conveyor belt (WCB) moisture flux is shown in (a). Cyclone LWP (precipitating and non-precipitating liquid) is shown in (b). Observations and MERRA2-inferred values of CDNC in the southwest quadrant of the cyclone ($CDNC_{SW}$) are shown in (c-d). In (c) and (d) the top and bottom third of distribution are indicated with dashed lines. In (a) edges of the WCB ~~bins~~ terciles used in Fig. 3~~Fig. 3~~ are shown with dashed lines.

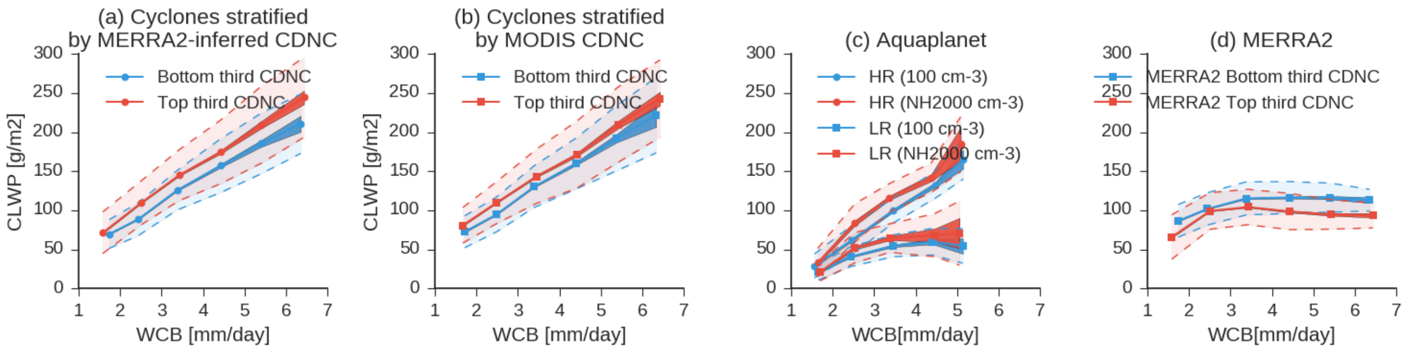


Fig. 6 Comparison between the dependence of cyclone mean liquid water path (CLWP) on warm conveyor belt (WCB) moisture flux as a function of increasing aerosol. The CLWP is binned by WCB moisture flux. The standard deviations in CLWP across bins are shown as shading with a dashed border. The ~~standard error~~95% confidence ~~in~~ interval on the mean is shown as thick solid lines. Figures (a) and (b) show ~~(a)~~ MAC-LWP observations from 2003 to 2015 stratified by (a) MERRA2-inferred CDNC$_{SW}$ and (b) stratified by observations of CDNC$_{SW}$ from MODIS. Panel (c) shows the simulated CLWP in a suite of global aquaplanet simulations split into low and high ~~CCN~~ N_{acc} simulations. ~~.~~ In the aquaplanet simulations a high aerosol channel is added to the northern hemisphere to investigate the response of cyclone properties and surface N_{acc} is ~~accumulation mode aerosol concentrations are~~ noted in the legend. Aquaplanet simulations are run at convection-permitting (HR) and GCM-surrogate resolution (LR). Panel (d) shows MERRA2 total precipitable liquid stratified by MERRA2-inferred CDNC$_{SW}$. In (a,b, and d) cyclones with CDNC$_{SW}$ in the top and bottom third of observed CDNC$_{SW}$ (see Fig. 5~~Fig. 5~~) are indicated by red and blue lines.

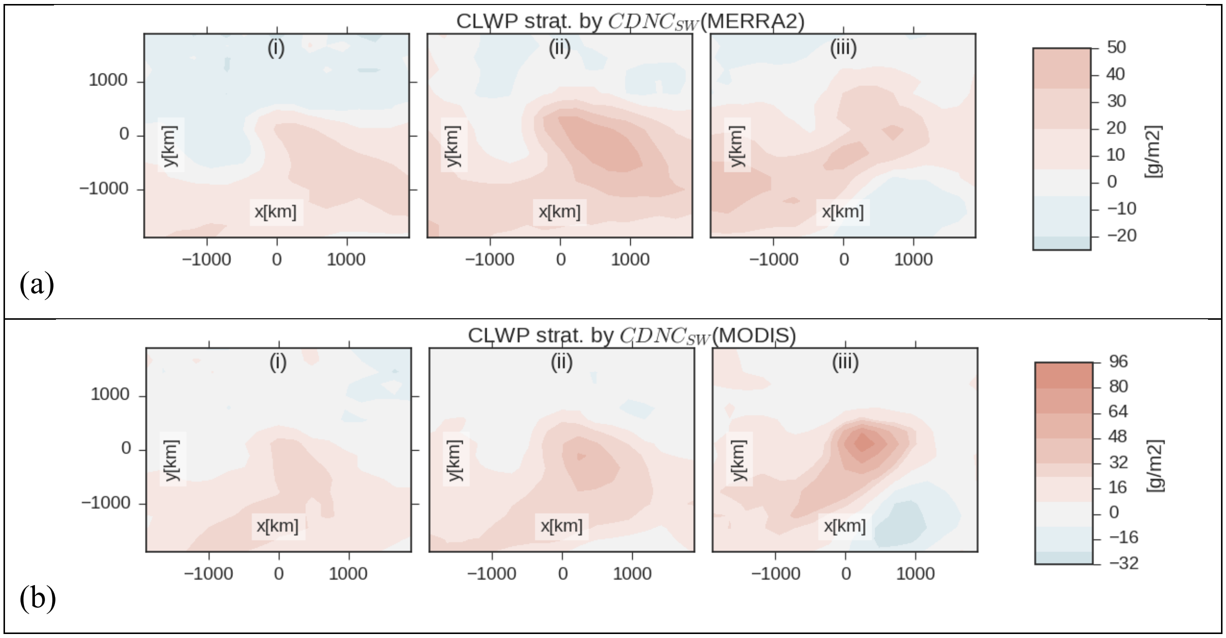


Fig. 7 The difference in cyclone composited MAC-LWP CLWP between the top and bottom third of CDNC$_{SW}$ inferred from MERRA2 (a) and observed by MODIS (b). Composites are shown split into WCB ~~categories~~ quantiles as in Fig. 3~~Fig. 3~~.

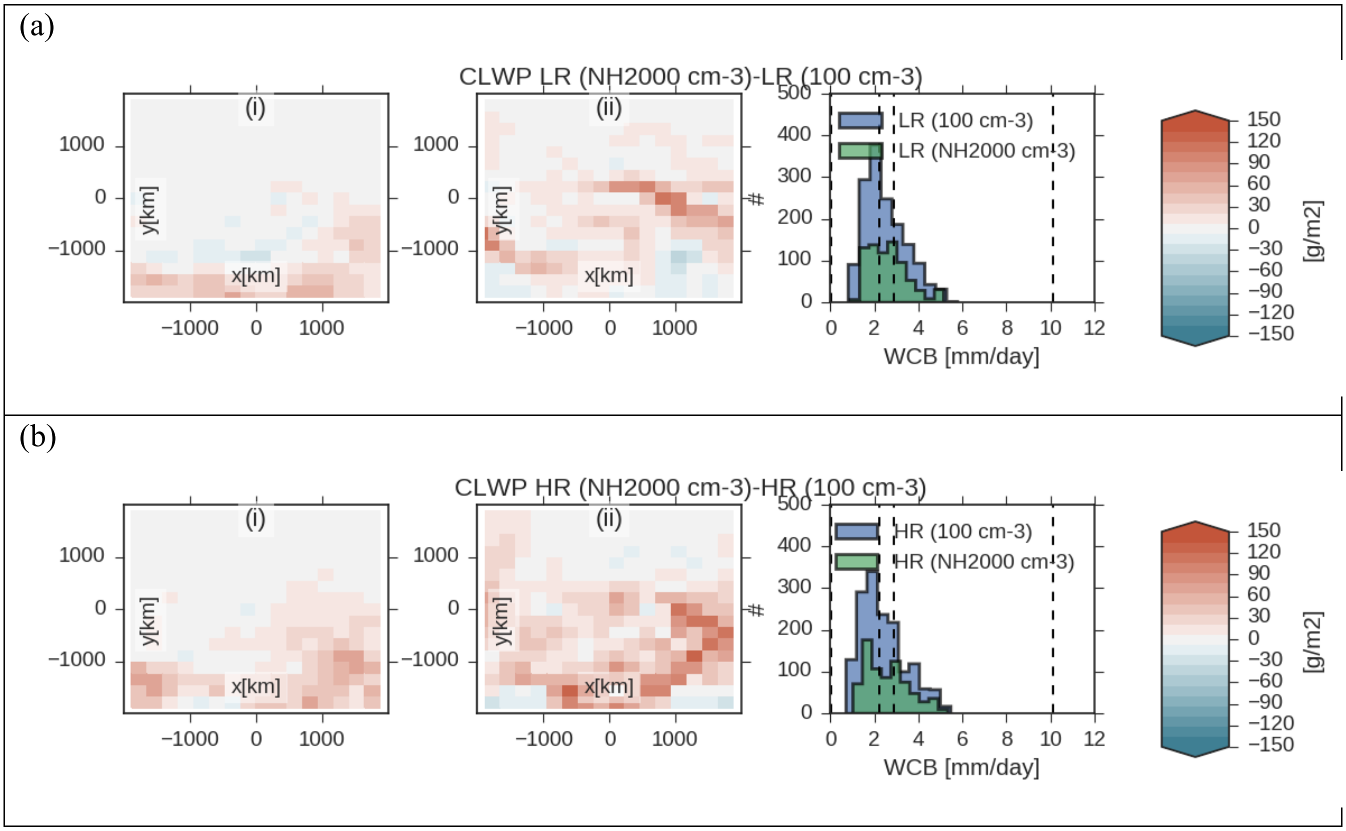


Fig. 8 The difference in mean cyclone composites of CLWP between the high and low ~~CCN~~ N_{acc} simulations for (a) the GCM-surrogate low-resolution (LR) simulation and (b) the convection-permitting (HR) simulation. Differences in mean cyclone composites for different WCB regimes are shown. It should be noted that the relatively short integration time (relative to the observations) of the simulations did not yield a large number of cyclones ~~with WCB>5 mm/day~~in the top tercile of observations and only the first two WCB regimes are shown in contrast to Fig. 7~~Fig. 7~~. The distribution of cyclones by WCB in the simulations is shown on the rightmost plot.

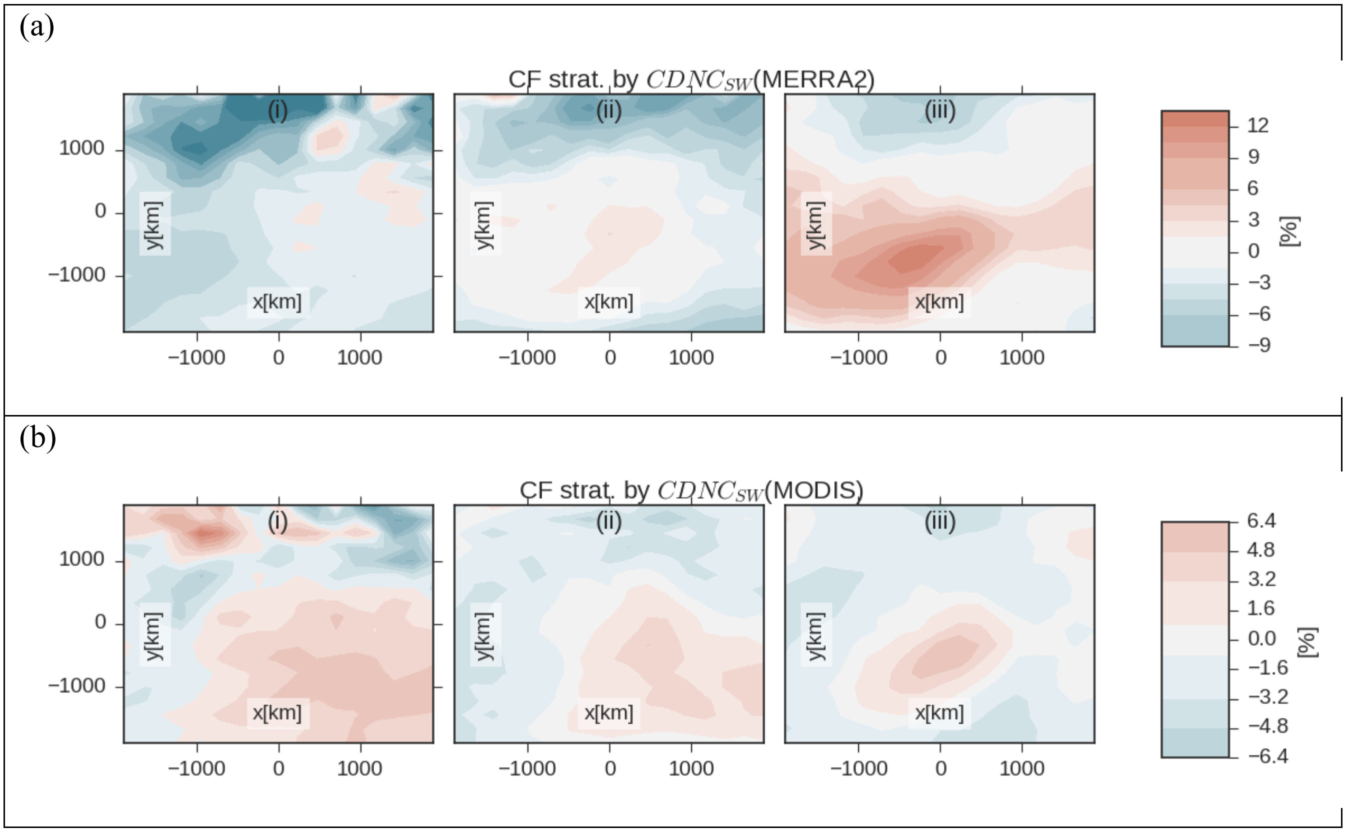


Fig. 9 As in Fig. 7~~Fig. 7~~, but showing differences in cloud fraction.

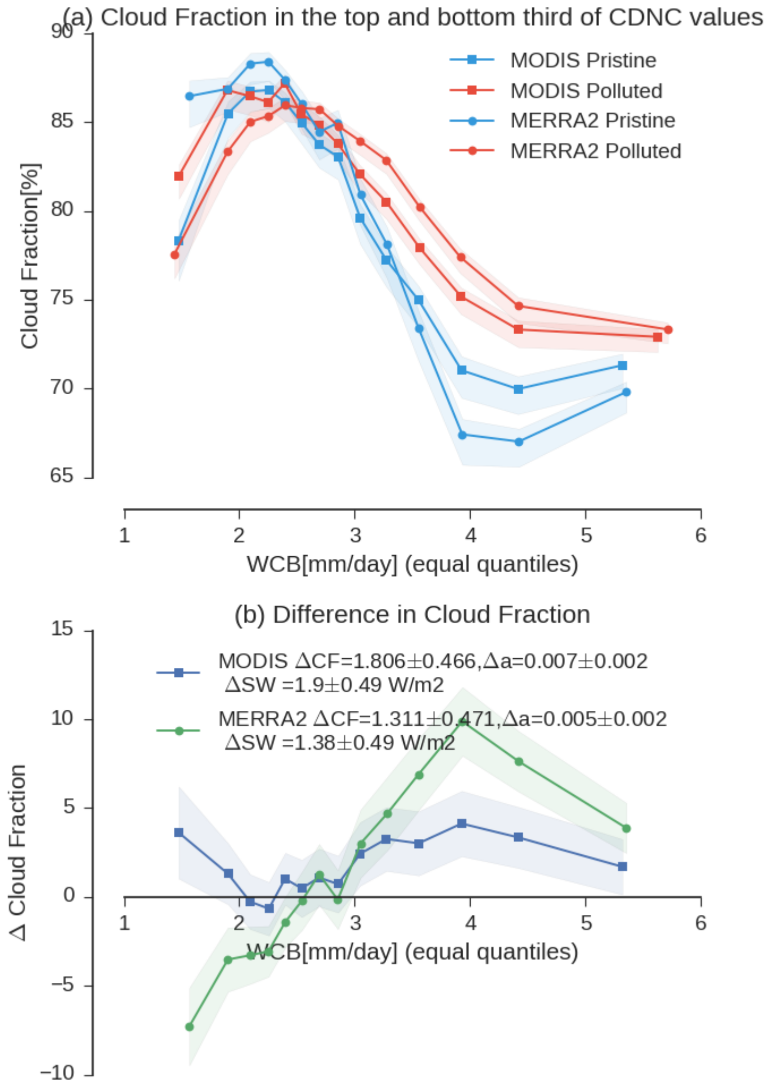


Fig. 10 (a) Cyclone-mean c~~C~~loud fraction split into 15 equal quantiles ~~in cyclones identified~~and split into ~~as~~ high and low CDNC$_{SW}$ populations. Shaded areas show 95% confidence range in the mean in each quantile. The CDNC data set used to partition the cyclone population is noted in the legend. (b) shows the difference between the high and how CDNC$_{SW}$ populations. The difference in cloud fraction between the populations and 95% confidence in the difference are noted in the legend. The relation between cloud fraction and all-sky albedo for the midlatitudes from Bender et al. (2017) is used to approximate the difference in albedo consistent with this difference in cloud cover. The difference in reflected shortwave (SW) is calculated by scaling the albedo by the annual-–mean insolation between 30° and 80° latitude.

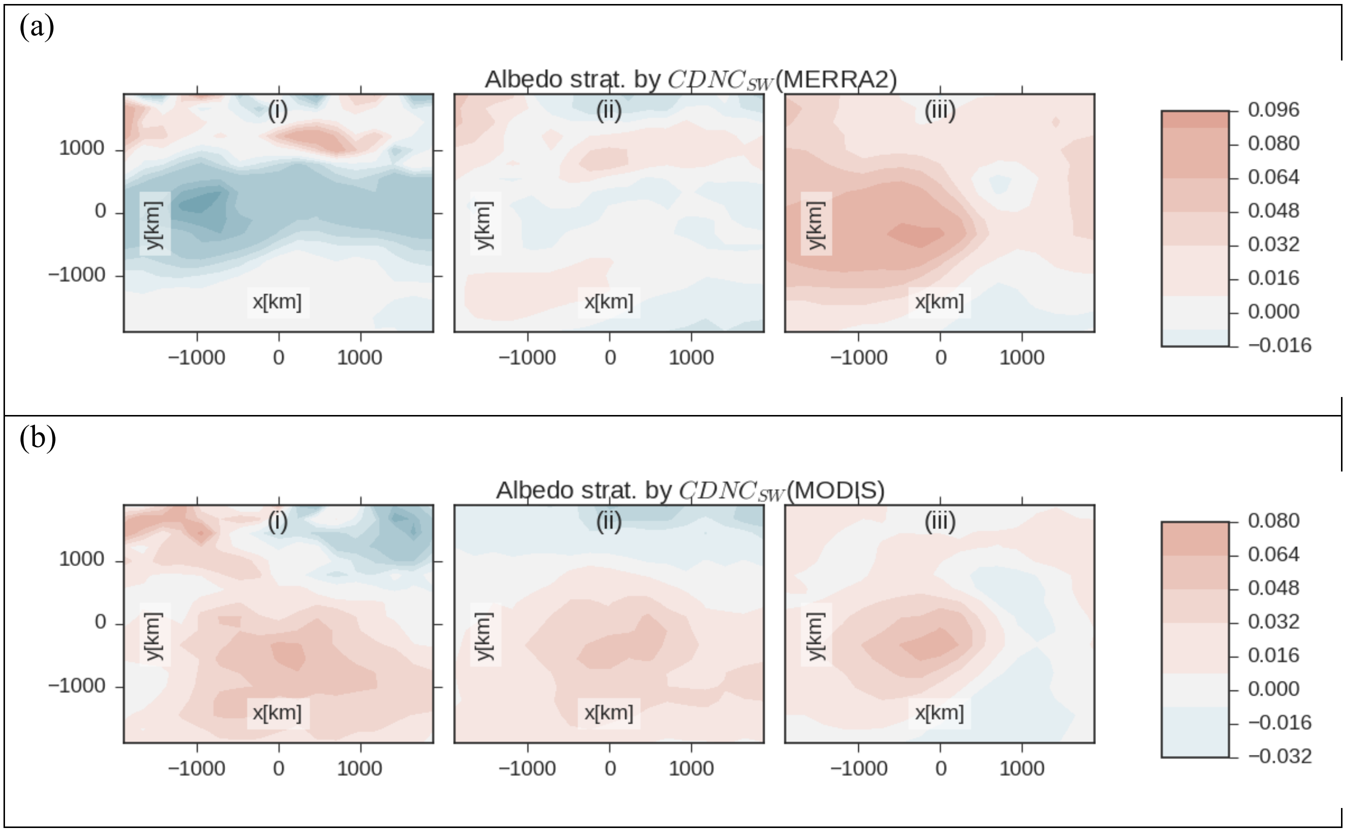


Fig. 11 As in Fig. 7~~Fig. 7~~, but showing differences in albedo.

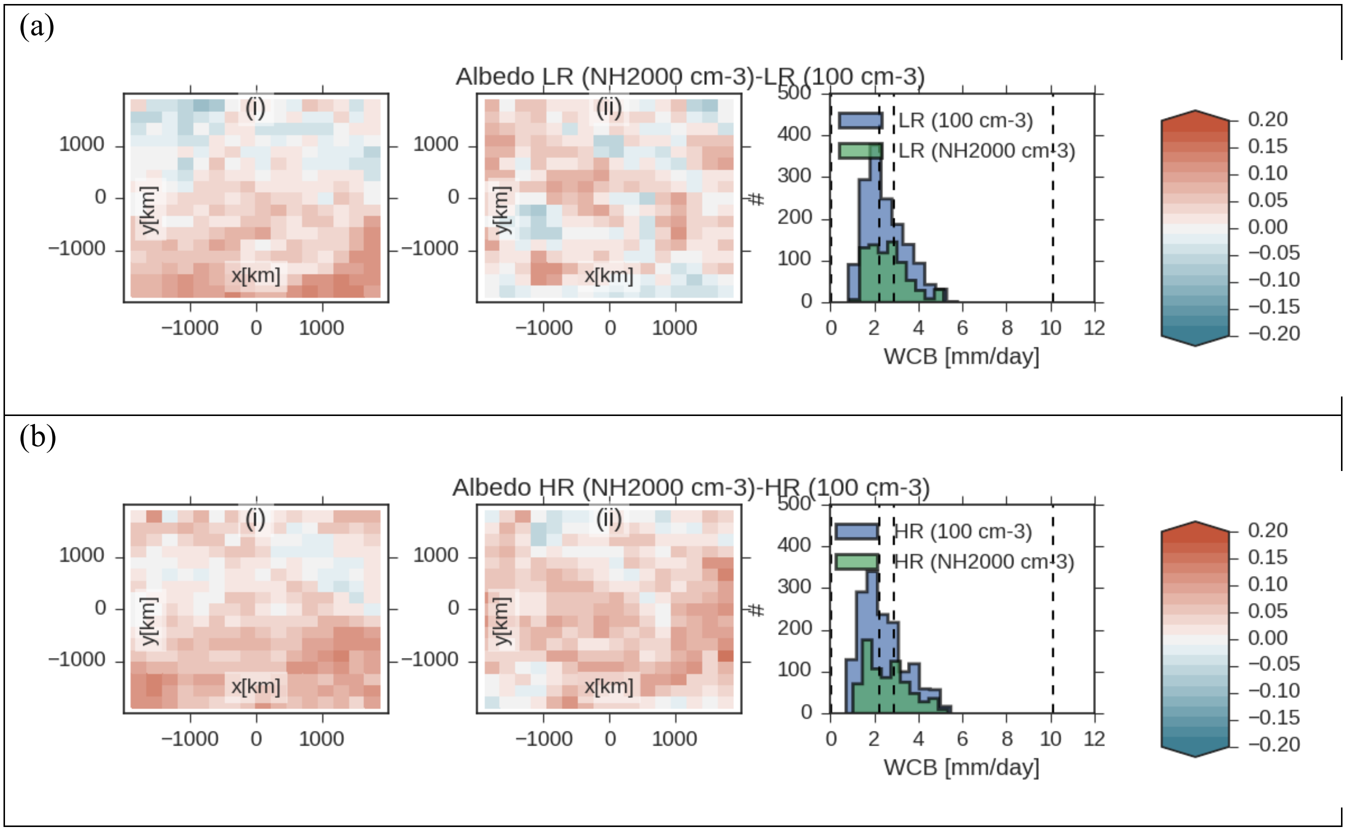
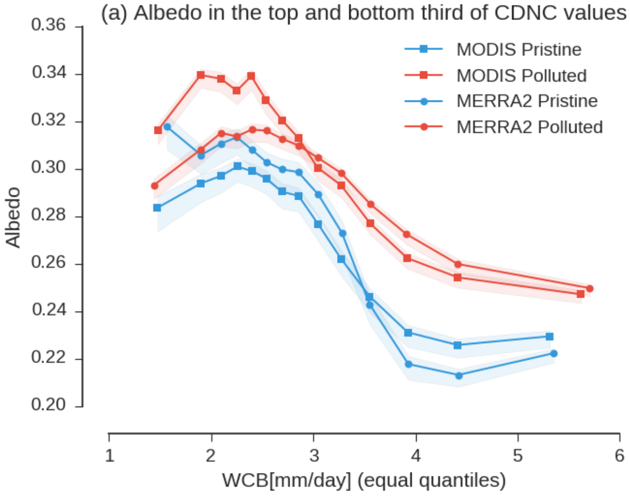


Fig. 12 As in Fig. 8~~Fig. 8~~, but showing differences in albedo.



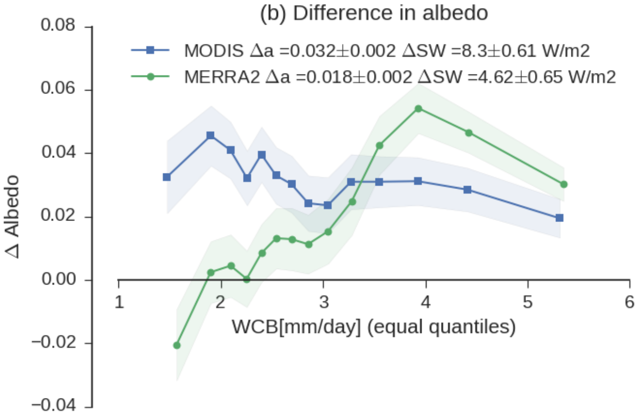


Fig. 13 (a) Cyclone-mean albedo from CERES as a function of WCB moisture flux. Data is shown binned into equal quantiles of WCB moisture flux and separated into the top and bottom third of observed CDNC$_{SW}$. The ~~Standard error~~95% confidence intervals in the mean ~~is~~ are shown using shading. Both MERRA2-inferred ~~sulfate~~ and MODIS-observed CDNC$_{SW}$ are used to partition the top and bottom third of CDNC$_{SW}$ and are noted in the legend. (b) shows the difference in albedo between the top and bottom third of observed CDNC$_{SW}$ as a function of WCB moisture flux. The 95% confidence interval on the difference in each quantile is shown using shading. The mean difference and 95% confidence range on the difference in albedo and estimated reflected SW based on this difference in albedo ~~is~~ are noted in the legend. To calculate reflected SW t~~T~~he difference in albedo is scaled by the annual-mean climatological insolation between 30°-80° ~~to calculate the reflected SW~~. Because albedo is a strong function of solar zenith angle

(SZA) only 3-hourly measurements with SZA<45° are considered here (similar calculations using cut-offs of 30°, 60°, and 90° are shown in Fig. S9~~10~~,10~~1~~,11~~2~~).

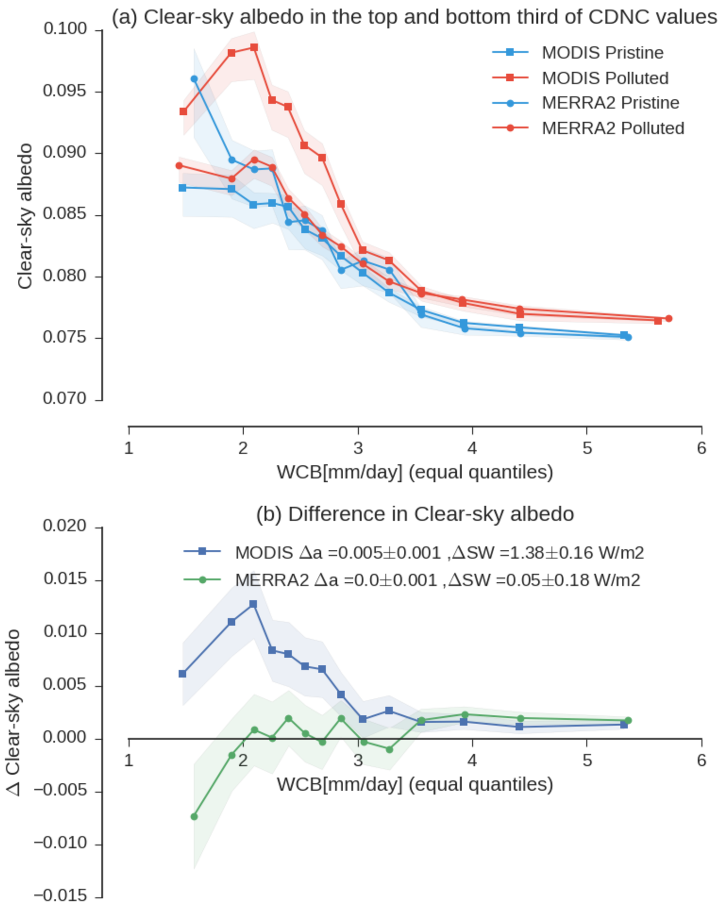


Fig. 14 As in Fig. 13~~Fig. 13~~, but showing differences in cyclone-mean ~~aerosol~~clear-sky albedo. The estimated difference in clear-sky SW is calculated based on the annual-mean insolation between 30-80°~~ optical depth (AOD) from MERRA2 reanalysis.~~

~~**Bender et al. (2017)**~~

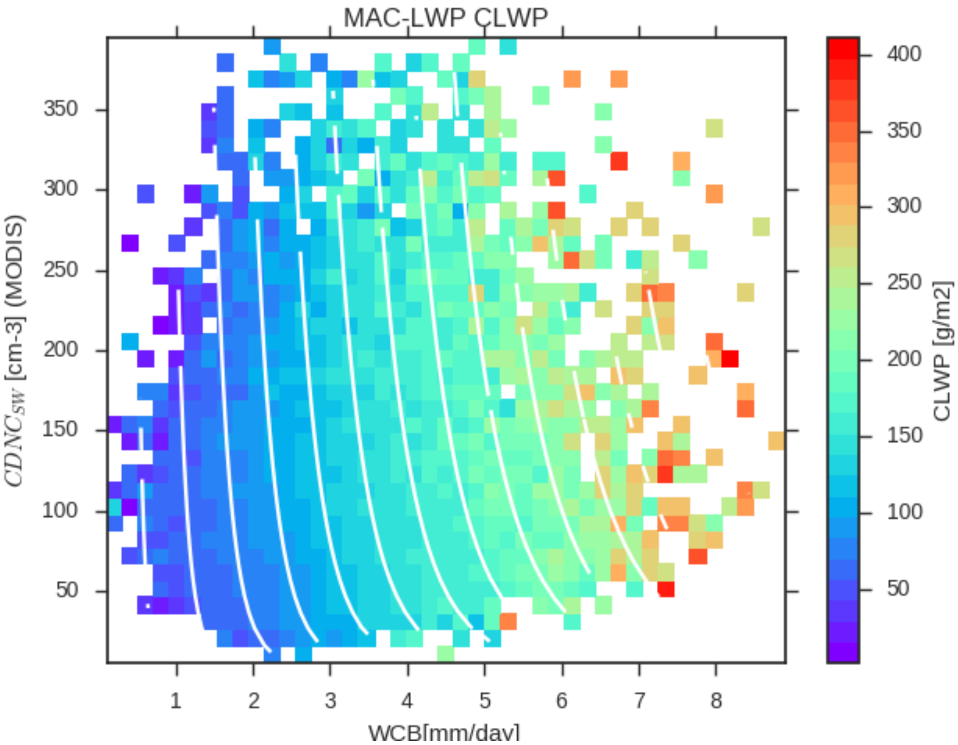


Fig. 15 The cyclone-mean CLWP in units of ~~mm~~ g/m^2 of liquid water observed by MAC-LWP binned as a function of WCB moisture flux and the $CDNC_{SW}$ observed by MODIS. Data is binned into equal size bins for the purpose of visualizing the data record. White lines show contours of constant CLWP as predicted by Eq. 1 and the coefficients listed in Table 2~~Table 2~~. The dependence of CLWP on $CDNC_{SW}$ inferred from MERRA2 is shown in Fig. S12~~3~~.

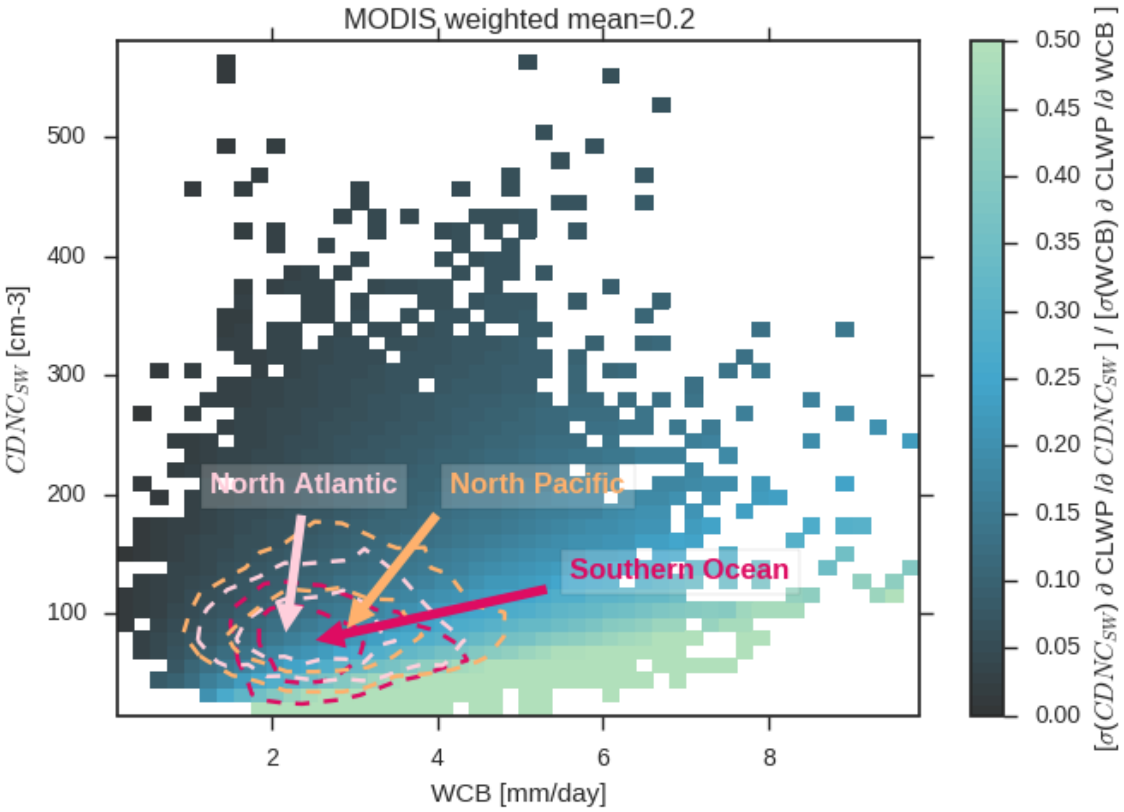


Fig. 16 The relative contribution to CLWP of perturbations in $CDNC_{SW}$ and perturbations in WCB as estimated using Eq. 1 and the standard deviation of each predictor over the historical record. The regression model was trained using MODIS-observed $CDNC_{SW}$ (the same plot is shown for MERRA2-inferred $CDNC_{SW}$ in Fig. S134). The partial derivative of Eq. 1 is taken with respect to each predictor and scaled by the standard deviation of that predictor. The ratio of the partial derivative scaled by standard deviations in each of the predictors is shown using colors. The joint probability distribution of cyclones during the observational record for different ocean regions are roughly indicated using dashed lines. The joint probability distribution of all observations is used to calculate the weighted mean of the fractional contribution of perturbations in $CDNC_{SW}$ and WCB over the range of WCB and $CDNC_{SW}$ in the observational record.

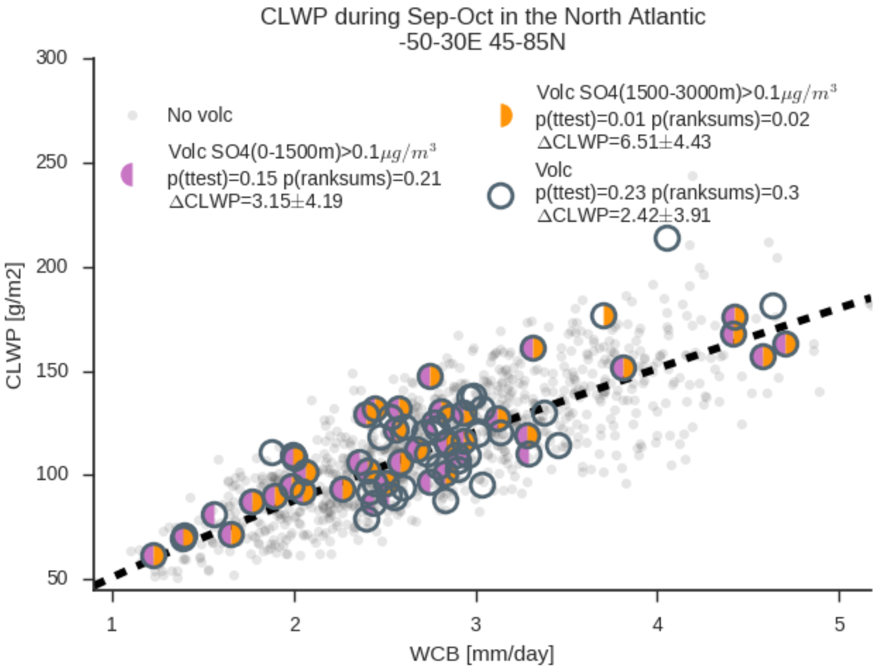


Fig. 17 The behavior of cyclones as a function of WCB moisture flux are contrasted for September and October during the eruption of Holuhraun and during all other years. The CLWP in the North Atlantic (50°W-30°E, 45°N-85°N) in September and October is shown as a function of WCB moisture flux during all years except 2014 (grey dots), during 2014 (grey circles), and for cyclones that the NAME dispersion model predicted to have interacted with the sulfur plume from Holuhraun (purple and orange half-circles). NAME was used to simulate the Holuhraun plume assuming it extended from 0-1500m at its source (purple) or from 1500m-3000m (orange). The mean sulfate mass within the southwest quadrant of each cyclone was calculated. Only cyclones with a sulfate mass of 0.1µg/m3 were considered to have interacted with the plume. A power law fit of climatological CLWP to WCB moisture flux in the region is shown as a dashed line. This fit was used to calculate anomalies in CLWP for cyclones in September and October except 2014;~~,~~ for September and October of 2014;~~,~~ and for the cyclones that NAME ~~indicated~~predicted ~~had~~to have interacted with the plume. A t-test and a non-parametric rank sum test were used to evaluate the difference in means between the climatological anomalies in CLWP and the anomalies in 2014 and for the cyclones that NAME predicted had interacted with the plume. The p-values for these tests are given in the legend. Differences in means and 95% confidence intervals, assuming a normal distribution, are also given. Latitude ranges of 35°N-90°N, and 30°N-90°N are shown in Fig. S14~~6~~ and S15~~7~~.