Response to reviewers

We thank the reviewers for their careful reading of our manuscript. Several of the comments by reviewers helped us significantly improve the paper so that is great. In response to their comments we have made several significant changes to our paper.

1. We have replaced the AMSR data set with the MACLWP data set. This reduces uncertainty related to diurnally averaging the dataset. Full details of the improvement in MACLWP data relative to AMSR data are given in Elsaesser et al. (2017).
2. We have expanded the methods section to more completely describe what has been done in the paper.
3. Based on comments from the reviewers we have found that use of the albedo from CERES from all solar zenith angles (SZAs) was introducing a substantial bias due to SZA effects. This effect is discussed in detail in Bender, Engström, Wood, and Charlson (2017) and we have taken steps to show sensitivity to this effect in our analysis.
4. Based on further analysis of the Holuhraun eruption we have decided to remove this case study from our paper for more in-depth evaluation at a future time. Our previous results were based on AMSRE/2 data, which changed overpass time in the transition from AMSRE to AMSR2, spuriously creating differences in cyclone properties within the data record. Use of MACLWP data shows that our previous analysis is less robust once the diurnal cycle is more carefully accounted for.

We will answer each reviewers comments in detail below- reviewer comments are in italics.

Reviewer 1:
The second part is a set of observations are drawn from several sources, and probably need a bit more discussion. This is especially true because the 'observations' include essentially diagnostic aerosol fields from a reanalysis system that does not have aerosol-cloud interactions and are used as a very coarse proxy (without explanation really) for aerosols.

We did not explain this aspect of the paper sufficiently. In fact, in the paper we emphasized the reanalysis aerosol giving the impression that our results are completely dependent on this. We have rewritten the methods and paper to show that our results are not qualitatively sensitive to whether we use MODIS or MERRA2 to look at CDNC. We have also tried to provide a more thorough evaluation of how MERRA2, MODIS and the OMI SO2 product covary in McCoy et al. (2017) to support these observations further. Of course the paper should stand on its own and our results using MODIS and MERRA2 are presented in parallel now. We have also expanded our discussion of the data sets and methodology.

Secondly, the model simulations are confusing and not fully evaluated. As noted below, I question 'convective permitting' simulations as being in a really bad part of the 'gray zone'.

Thank you- good point. We have inserted a discussion regarding pros and cons of the simulation resolution. Overall, we find the same results in a qualitative sense at both the convection permitting resolution and the GCM resolution so our results are not hugely sensitive to our choice of resolution and the contrast between the GCM and 6.8km resolution demonstrates how sensitive a single model might be to these choices in terms of its cyclone behavior.. We have added a reference to a paper showing that within a resolution range of 1-16km the mean field statistics of parameters such as broadband fluxes and water path do not change much (Field et al., 2017).

Finally, the volcano work needs more sensitivity tests and does not seem robust. Intro- ducing a totally different model to this work is a bit complicated and it's not clear what can be learned from the langrangian dispersion model.

See comment above, we have removed this work as we believe our results are robust without it and the analysis appears to be sensitive to how much ocean coverage is required in the cyclone composites and how far we assume the lagrangian dispersion model is accurate for. We will return to this analysis in a future study.

Page 1, L14: surrogate climate model? What is that? This might need a bit more description in the abstract.

We feel that the main focus of our paper should not be examining the contrast between the GCM-resolution and convection permitting simulation because this contrast may not be highly representative of the population of GCMs and this has been deleted from the abstract. The simulation discussion in the methodology has been expanded. The surrogate climate model is just a coarsened (140km) version of the high resolution model with parametrized convection switched on. The use of parametrized convection at coarse resolution makes it more similar to a current climate GCM than the 6.8km with explicit convection. The coarse model with parametrized convection provides a convenient comparison to suggest what climate model might do, but without too many changes that make it difficult to disentangle the cause of the difference.

Page 1, L30: it might be useful here to separate out the first and second effects in these studies for clarity.

We noted that the first group of papers address the first indirect effect and the second group address the lifetime effect. We have rewritten the introduction to try and make it clear which papers refer to each effect.

Page 2, L14: I think you need to reference or show some model validation for the cyclone composites. Maybe it is later but it should be noted here.

We have added comparisons of the cyclone composites across WCB regimes between the observations, simulations, and MERRA2. There are big differences, although the convection permitting simulations look generally ok. Since the WCB moisture flux- rain rate relation appears to be a feature of GCMs and observations (Field, Bodas-Salcedo, & Brooks, 2011) and we hypothesize that the inhibition of rain via aerosol-cloud interactions leads to divergence between low and high CCN simulations the mean-state of the simulated cyclones does not have to perfectly match the observations. The comparisons in the paper discuss the differences and similarities between the observations and simulations now.

Page 2, L25: does MERRA2 assimilate AMSRE or MODIS? If so, then these are co dependent.

This is a very good point- we have re-examined the MERRA2 documentation. In (McCarty et al., 2016) a list of assimilated radiances and data is made. SSM/I rain rates are retrieved through 1987-2009. SSM/I radiances are also used for this period and are cloud-cleared following (Derber & Wu, 1998). AOD from MODIS is assimilated in the aerosol analysis, but cloud properties are not (Randles et al., 2016). We have noted this in the text, but we also note that all else being equal higher LWP should imply a higher rain-rate and lower aerosol mass so the co-dependence should lead to anti-correlation between LWP and SO4. We have added analysis of the MERRA2 total precipitable liquid water path and indeed see that this is the case. That is to say, higher CDNC (from the MERRA2 SO4) corresponds to lower CLWP cyclones and vice versa. Thank you for suggesting this.

Also, the statement here about CDNC from a sulfate regression I think means you are baking in an indirect effect.

As noted above we use MODIS and MERRA2 SO4 as proxies for CDNC and both direct observations and the MERRA2 proxy produce similar results.

Page 3, L20: so these simulations prescribe an indirect effect. How was this tuned?

This indirect effect was not really tuned. In a sense the simulations are more of a 'fixed CDNC' set of simulations than a 'fixed CCN' simulations (as in Lu and Deng (2015)). Another Twomey-type activation scheme would have produced results that show the same qualitative result. We have added some verbage to the text to explain this more clearly. We have added a note that while the CDNC is sensitive to both vertical motions and CCN, the sensitivity to CCN contributes the vast majority of variability in CDNC. Thanks.

Page 3, L28: how do these emissions compare to Malavelle et al 2017 and Gettelman et al 2015?

Although this material has been removed we note that these emissions are somewhat more in keeping with observations than the fixed 40kT scenario in Malavelle 2017. The time varying flux is somewhat more realistic based on observed fluxes (Schmidt et al., 2015).

Page 4, L8, but also without variable aerosol sources from land that might co vary with meteorology.

The aerosol in the aquaplanet was constant so it can't vary.

Page 4, L9: This is not resolving, it may be barely even permitting. Most mesoscale meteorologists I know would not run a model in this Gray zone between 3km and 10- 15km, the latter with a convection scheme, and usually 3km even with some sort of vertically coherent parameterized turbulence.

I hope there is validation of this model somewhere? How about some cyclone com- posite maps to compare to observations, not just WCB flux.

Please see comments above regarding model resolution and additional plots of composites.

Page 4, L20: The ice assumption is simplistic. Is there significant ice in your cyclones? It

Depends on the temperature in your aquaplanet run.

There is significant ice, but it does not seem to be a strong function of WCB moisture flux or aerosol (see Fig S2 of the original SM). The ice number is linked to 'Cooper temperature dependence'. We do not change the ice number representation when we change the CCN. We have added a note to this effect in the text in section 3.1.

Page 4, L30: To what extent might that be a consequence of the model formulation.

Perhaps this figure needs to be in the main text?

This is a common function of GCMs and observations (Field et al., 2011; Field et al., 2008) and not a function of this particular model formulation and appears to be a consequence of mass conservation within the midlatitude cyclone systems.

Page 5, L2: Again, the frozen water path is not aerosol aware. If the LWP but not the IWP changes, then what does this say about how the IWP is formed in the model? This should be assessed by looking at microphysical process rates.

Please note that the aerosol exponentially decreases with height. IWP is aerosol aware, but the assumption here is that aerosol is not uniform with height throughout the atmosphere.

Page 5, L8: What does it mean that the slopes are very different, and the mean values for the model at low WCB strength are off by a factor of 2.

Do you mean the slopes in the observations or the simulations? It seems like the model is more sensitive to WCB at high aerosol, which seems reasonable if precipitation is being inhibited. The model mean is much lower than the observations we now note this in the paper and this is likely just because we have not used a cloud scheme, which would add additional complexity and make it harder to understand the differences between the low- and high-resolution simulation. We should note that the goal of this study was not to create a simulation with perfectly realistic cyclones, but to examine how cyclones respond to aerosol in a qualitative sense and use that insight to analyze observations in a sensible way.

Page 5, L14: However, you presume that the convective permitting simulations can capture the real physics of convection at 6.5km. I don't think this is true.

We don't say that the convection permitting simulation is perfectly representing convection in this section. We note that the convection parameterization, which is not aerosol-aware, is not acting on any of the clouds.

Many studies with high resolution limited area models (1km horizontally and finer, with higher vertical resolution) note competing processes such as enhanced cloud depth that may offset some of the impacts. Can you comment on this?

We argue that our simple model formulation appears to be consistent with the observations. We have added a note to this effect to the discussion. Thank you.

Bottom line: without showing that your aquaplanet simulations are realistic (and they look pretty weak from the one evaluation in Figure 1), you have oversold this conclu- sion.

As suggested we have added more evaluation of the aquaplanet. The goal of the aquaplanet is to offer some sort of framework to analyze the models in. The main framework is the strong dependence of rain rate and by extension CLWP on WCB moisture flux. This appears to be a feature of many GCMs (Field et al., 2008). In our idealized simulations we have been able to run high and low aerosol perturbation simulations. When observations are analyzed in the same way high and low CDNC cyclones behave in a qualitatively similar sense to both our high resolution and low-resolution simulations. Based on Reviewer 1's careful analysis we have tried to prune back the conclusions and shore up the methodology.

Page 5, L16: you need to explain the statement that climate sensitivity is too low. I understand the point: but this implies that people adjust the climate sensitivity to match the forcing. It may imply that sensitivity is higher in reality if the forcing is higher. It needs to be rephrased for a reader who does not understand the direction you are going in.

This comment has been removed. Given the spread in aerosol-cloud forcing it is likely many models also have too strong an aerosol cloud indirect effect. Admittedly, if all climate models without aerosol-aware convection miss this effect this does make it a systematic error, but going through explaining this is convoluted.

Page 6, L3: I understand that you have a published reference for this, but taking so4 from a reanalysis and trying to use that as a CCN proxy with observations is problem- atic. If MERRA2 assimilates AMSRE, then you have a potential co-variance problem.

See comment in regards to this above.

Page 6, L9: I think you need to show cyclone composites in the main text.

See comment related to P2 L14 above- we have added figures for models and observations to the text.

Page 6, L10: Didn't Field and wood use a 2 d cyclone composite? Check.

They did, as does this paper. I don't understand how it relates to the material here. High and low aerosol cyclones are for the cyclone as a whole, not for a specific part of the cyclone.

Page 6, L13: Clarify what is observations and what is reanalysis here. It might be interesting to also look at the CLWP in MERRA without ACI: if it also changes with CCN then there might be an issue with co variance here.

Thank you for your suggestion. We did this and MERRA2 implies the opposite of the observations.

Page 6, L21: These radiation fluxes need uncertainty estimates.

Great point- one of the major things that we have changed in the paper is the radiation calculation. We realized that the dependence of albedo on solar zenith angle was yielding unrealistic results (high albedo cyclones having the lowest water path because they had very high SZA). We have added additional analysis exploring this to the paper. We have also added uncertainty to our analysis.

Page 6, L34: except that the analysis says ice is not important but we ignore ice. So if ice was doing something you would not see it. I think it is a bit dangerous to make this assumption. How much IWP Is in the cyclones?

I am not clear what you mean by this, but we do show IWP for the simulations in the SM.

Page 7, L5: Where do all these values come from? Are you just looking at observations + MERRA here or is there something from the model. Also, does CLWP include a rain rain which is part of the WCB metric? What does that mean?

This is from the observations. As noted in the methods section the CLWP is precipitating and non-precipitating liquid. We have chosen to look at this because it is what the microwave radiometer is sensitive to.

Page 7, L14: I am still confused about this metric. Maybe a better figure would help.

Thank you- we have remade this figure and tried to explain it better in the text. The idea was to point out that in the context of the regression model not all cyclones are equally sensitive to aerosol forcing. Not a hugely surprising result and really just restating Carslaw et al. (2013), but we thought it was important. We have added notation to show where different ocean regions fall on this figure.

Page 7, L19: what about co-variation?

This is just the weighted mean of the figure so it takes covariation into account. We have noted in the text that this analysis is hopefully illustrative.

Page 8, L6: The regression model needs uncertainties on it if you are going to do this. It is not clear whether the results are significant.

Removed- see above.

Page 8, L9: Can you describe the 'Lagrangian model' NAME in more detail? What met fields drive NAME? Are these MERRA2 cyclones or observed cyclones. I think this needs a more complete treatment. This seems like the beginning of a different paper.

Removed- see above.

Page 8, L13: I'm not sure that there is a relationship here, or that the anomalies are statistically significant. Shouldn't the line increase with sulfate (i.e. CCN).

See above- removed.

Page 8, L18: doesn't this depend on the uncertainty in the regression model (which I do not think you have described)

See above, removed

Page 8, L20: Malavelle et al 2017 also used 10 years of data for a climatological comparison.

Sorry- we phrased that poorly. Removed.

Page 8, L20: how are the convective permitting simulations used here? They were aquaplanet?

This material has been removed.

Page 8, L25: But the sensitivity of convective permitting simulations is higher than observations I think?

This material has been removed.

Page 8, L29: I think a more thorough sensitivity test is necessary. Also, please check your emissions against earlier work as noted below. Page 9, L8: But the Holuhraun simulations seem sensitive to emissions, and I'm not sure your statistics are robust. Again, this conclusion does not seem robust Page 20, Fig8: is this statistically significant? The most 'polluted' storm is not signifi- cant. Take out 3 points and there is nothing here. I do not think this is robust.

Agreed- we found sensitivity to our assumptions in this analysis. This does not affect the conclusions in the remainder of the paper.

Page 9, L4: but you largely prescribe these effects in idealized models: fixed CCN, no scavenging and infinite sources, fixed relationships. Of course you are going to find this.

This is a good point. Our model by default tends toward a positive lifetime effect on LWP, although this is not a given because of interactions with other clouds and the environment – see Fig 6 and 7 of Miltenberger et al. (2017). We have added to text to make this clearer.

Page 9, L6: you have said nothing about radiative forcing over the 20th century. As before: I see your logic but you need to explain it in several sentences with references.

The argument here is just that the forcing is sufficiently large to be non-negligible. We have expanded our discussion to make it clearer that this is all we are saying.

Page 9, L10: I'm not fully clear what is idealized and what is observations in this study,

We have tried to expand our methods and discussion to make the analysis clearer.

Page 9, L15: Vague, and of course there is one in the simuLations. The observational part is interesting but needs a more careful treatment.

See above. We have tried to more clearly articulate the analysis and acknowledge that the model set up we have will enhance LWP with enhanced CCN, all else being equal.

Page 9, L17: as noted, I think this statement is not defended by the analysis and ignores a lot of previous literature on the complexities of aerosols in convection. It needs a much more thorough analysis of the simulations.

We have clarified this statement to reflect the fact that we have shown this via observations, for the first time.

Page 14, Fig2: Are cyclones composited only over the ocean here?

That is correct, we have expanded our methodology to more clearly articulate this.

Page 16, Figure 4 needs some discussion of error and/or error bars: are these lines significantly different?

We have updated our analysis of the albedo effect.

Page 18, Fig6: the presentation is not that effective. It is hard to read the color scale when you use a 2 color gradient. I'm not entirely clear what this is trying to show.

See above. We have tried to more clearly explain the figure.

Reviewer 2:

1. There are not enough details and a lot is left for the reader to find in other publi- cations. The accuracy of the various derived observations would be very helpful. The paper is rather succinct, and some figures were moved to a supplement document, as if it were intended as a letter or short publication.

This is correct- we have tried to more clearly articulate our research in the revised submission.

2. I am not convinced by the work done with CERES on the impact of the aerosols on the storm albedo (Figure 4 and associated discussion), possibly because there are not enough explanations on how the results are obtained. First it is not clear whether the WCB is constrained in the figure, then there is very succinct discussion on what actually might impact the albedo: with the warm frontal and warm conveyor belt regions of the cyclone dominating the signal and their large amount of high level, mostly ice clouds, there is little signal to be expected from changes in *aerosols or low level clouds.*

This is a good point- we have significantly reworked our evaluation of the albedo effect, primarily due to difficulties resulting from SZA bias (see previous reviewer comments). The reviewer also makes a good point regarding ice cloud, however, assuming that the ice cloud effect on albedo is more or less randomly distributed and is unaffected by CCN then it should just add variability to the populations of low and high CCN cyclones. We have compared albedo from the high and low CCN cyclones in the SM of the original submission and we show that most of the effect in the observations and simulations is in the post cold frontal clouds. This is consistent with the proposed effect (eg CCN affecting liquid cloud cover). We have expanded this analysis and moved it to the main text.

In addition, if all cyclones are included, then the CDNC classification can be highly correlated with the cyclone properties and this would mask any impact aerosols direct and indirect ef- fect might have.

This is a good point regarding the direct effect. To try and better understand the possibility that we are somehow aliasing this effect in we looked at cyclone composites of CF and CLWP differences between high and low CDNC cyclones. We see that there is a general agreement between the regions where CF and CLWP enhance and where the all-sky albedo increases. Of course the partitioning of this radiative effect into components owing to changes in CF, LWP, CDNC is difficult, but it does seem like changes in cloud macrophysical properties and changes in albedo are happening in the same area so that supports the idea that these changes are driving the changes in albedo.

3. More details are needed on the work of section 3.2, especially the method, the whole section is confusing and so the importance of the results somewhat degraded

This has been expanded- thanks.

4. In the title, and in the conclusions, the "aerosol-cyclone indirect effect" is mentioned. This is misleading, as this would entail an observational evidence of an impact of aerosols on the cyclone dynamics. This study is about aerosol-cloud in- teractions in the midlatitude using extratropical cyclones to constrain the large scale environment.

Admittedly it's an aerosol-cloud effect, but we are using the cyclone as a constraint to order the meteorology so it seems reasonable to call it this since we are referring to the cyclone as the clouds that compose it. Please note that we have altered the main body of the text to reflect that it is the clouds within the cyclone changing to avoid any confusion, but we feel that completely spelling this out in the title would make it clunky.

Detailed comments: 5. Page 1, line 21-22: Here you introduce the role of extratropical cyclones: why not include their role for precipitation in the midlatitude which would be appropriate with the rest of the paper? Reference to the work of Hawcroft et al (GRL 2012), and Catto et al (GRL 2012) would make sense here.

Thank you. These are very good references to add. One thing we did find is that the precipitation is controlled by the WCB, not aerosol so the effect we show should not alter the total rainfall.

6. Page 2, line 1: here refer to Igel et al., 2013 before Malavelle et al. 7.

Done.

7. Page 2, line 20: "the algorithm of Field and Wood (2007)", please provide some details of what it is.

This section has been expanded- thanks.

8. Page 2, line 21 onward: when you introduce the CDNC product of MODIS, some details of what it is, its strengths and limitations should be included. The same is true of the other observations/products introduced in this section. There are many observations of the same parameter that are available, so it would be good to justify a bit more why these particular ones are used. For example, cloud fraction is from CERES, why not from MODIS (which the CERES product is in fact retrieved from if I am not mistaken)? How good is the MERRA-2 reanalysis for the sulfate mass concentration product?

This would be good to do. As you point out the CERES product is partially from MODIS- what was meant is that we used the data included as part of the CERES product. This has been altered to specify that the cloud fraction is originally from MODIS and geostationary satellites and the SYN1DEG product has been referenced. We have tried to provide some additional background and more complete citations on the other data products.

9. Page 2, last paragraph: how accurate is this rain water path estimate?

We have redone this part of the paper to use the MAC-LWP data set, which comes with the total (precipitating+non-precipitating) liquid water path already calculated. In the original paper we were just inverting the algorithm used by RSS to calculate the rain rate because the quantity that the microwave actually measures is the total liquid water path, not the liquid water path and rain rate. The rain-cloud partitioning from RSS was based on SST alone, which added ambiguity to the results since it covaries with WVP.

10. Page 3, section 2.2.2: more details on the model would be helpful. What does "NAME" stand for?

This has been removed.

11. Page 4, line 28: you write that the cyclone-centered mean is used to obtain the cyclone moisture flux. You should justify this a bit more, as the link to warm conveyor belt is not obvious: this is not the definition used typically. One argument is that cloud and precipitation occur predominantly in the warm conveyor belt and the warm frontal region, so the signal averaged in the entire cyclone region would be dominated by these two areas. Another is that cyclone cloud and precipitation depend strongly on the strength of the cyclone (here characterized by the surface wind) and the amount of moisture ingested in the cyclone (here characterized by the total *water path). Refer- ences are many, but as an example, one could be given to the Field and Wood paper (2007), and/or Bauer and Del Genio (JCLI 2006) and/or Rudeva and Gulev (MWR 2011).*

We referenced the Field and Wood 2007 paper in this paragraph, but we will add the other references in addition to the paper used to justify the simple model used in the original FW07 paper.

12. Page 5, line 1-2: "indicating that this aerosol-cyclone indirect effect acts through

the warm rain process." This is quite a leap, how do we know this is not model-specific? Also, in Igel et al. (2013), even though the total ice mass in a warm front shows very small changes with an increase in aerosol concentration, the microphysical processes differed such that the aerosol had a compensating impact on vapor depostion and riming efficiency in the mixed phase region. So could it be the case here as well? in which case you might want to change this statement as the indirect effect here would not just act through the warm rain process. And this is not an aerosol-cyclone relation, but an aerosol-cloud relation.

This was meant to refer to the model alone, and indeed this might be different in a different model. We have updated this to reflect this.

13. Page 5, line 14-16. In figure S3, how do we know that the change in ToA SW flux is not caused by the direct aerosol effect instead of the effect on liquid water path?

See discussion above.

14. Page 5, line 22: "allowing for accurate observations", how accurate? There are issues in heavy rain situations with microwave radiometer retrievals of water path and wind speed, which could impact the estimate of the moisture flux and the classification used in the paper. This should be discussed, preferably as early as section 2.

We have deleted 'accurate' as this is not a quantifiable statement. We have extended the discussion of biases in WVP and wind speed from microwave observations.

15. Page 6, line 4: "highly consistent" is vague, could you be more quantitative? How is CDNC obtained when clouds are present? How often do you have retrievals in the southwest quadrant? Do you have a threshold on this number below which you do not consider the cyclone in question?

We did not fully explain- CDNC is retrieved from cloud top effective radius and optical depth. It is only retrieved for overcast 1°x1° regions. We have added additional text to clarify how the retrieval is performed. Since the SW quadrant is highly cloudy it is not too difficult to perform the retrieval. There is no lower-bound on the number of retrievals required. We also perform the same analysis with MERRA2 SO4, which never has missing data since it is reanalysis and get the same results so this does not appear to be an issue. The inter-cyclone correlation between the

$CDNC_{SW}$ calculated using MERRA2 and MODIS is shown in the supplementary figures along with the effects of resampling MERRA2 SO4 so it is only sampled when MODIS can perform a retrieval. We have added text discussing differences in cyclone composited CDNC.

16. Page 6, line 5: because the cold front is moving with respect to the center of the cyclones, sometimes it is in the southwest quadrant, other times in the southeast quadrant, and so the aerosols could be ingested in either quadrant. Have you tried to use the southeast quadrant instead to see if the results change?

We have expanded the discussion related to this uncertainty to also include a recalculation of our results (characterized as the low-high CDNC plot as a function of WCB) using the cyclone mean CDNC and using the SE, and South part of the composite. The difference between high and low CDNC cyclones narrows when the cyclone-mean CDNC is used. This seems reasonable as it is adding a lot of noise to the calculation. However, if the SE part of the composite is used the separation between high and low CDNC cyclones narrows considerably. If just the south part of the composite is used it narrows much less. This seems like a case that would be improved by front identification, but for the purposes of this article we will stay with our simple cyclone compositing algorithm and note that there is some sensitivity to the sector of the cyclone composite used to calculate CDNC in the cyclone. We have also added these figures to the SM and additional discussion to the main text.

17. Page 6, line 9: Figure S4: the two composites look rather different, the two color bars should match to make the comparison easier, and a 1-1 line should be added to the (c) scatter plot. Also, why not add these three plots to Figure 2 and make it a 4-panel figure?

Thank you- we have moved all of these figures into a 1 panel figure and have added discussion regarding why they look somewhat different in structure. Overall the inter cyclone variability agrees well and once differences in sampling are accounted for MERRA2 and MODIS agree decently well in the region where there is abundant low, liquid cloud that MODIS can retrieve CDNC from.

18. Page 6, line 16-17: why not discuss the very obvious differences in the southeast quadrant between observations and model?

We have expanded the discussion and comparison between model simulations and observations. Thank you.

19. Page 6, lines 18-26: so here the albedo is estimated with the CERES data, correct? and so is the cloud fraction? how can you have 100% cloud fraction in your cyclone area? you did not explain how this is obtained. Also, if you really have 100%CF, how do you have C4

MODIS CDNC? Finally the differences between MERRA-2 and MODIS are not that different in magnitude from the differences between high/low CDNC, how significant is this effect on albedo?

We found that the SZA significantly impacted these results and we have redone this calculation. The MODIS CDNC retrieval is at cloud top, not in clear sky.

20. Page 6, lines 27-33: This is not very convincing, as there is no mention of the moisture flux being constrained, which means that the albedo effect could come from cyclone with low vs. high CDNC having different mean moisture flux and thus different cloud cover caused by this instead.

We have now constrained our estimate by WCB. See above.

21. Page 7, the regression model work: I am not sure I se the link between the albedo discussion and this work. Why not introduce this before the albedo work? This re- gression model is obtained how, based on Figure 5? Finally, I am not sure what the implication of these results is? In particular the very last sentence is unclear, please elaborate. Line 15, and line 17, large and small cyclones do not really mean anything, you do not know anything about their spatial extent. You could use strong/weak maybe, but you would need to specify that this is in term of moisture flux strength, not winds alone.

We train the regression model using the observational record. Figure 5 just shows a summary of this showing the average of all the observations in predictor space to show that the lines curve a little more sharply at low CDNCSW. We have specified that by large and small we mean large and small moisture flux. We have also remade the plot and tried to make it clearer that it is just a plot summarizing the regression models fit to the data.

22. Page 8, line 10: "both simulations", not clear what the two simulations are, only one is mentioned above.

Removed- see above.

23. Page 8, lines 22-26: This feels out of place, why mention the convection permitting simulations in this context? It just repeats what has been said a few times already about the merits of the high resolution vs GPM-resolution simulations.

This has been removed.

24. Page 9, line 15: "an aerosol indirect effect on midlatitude storms" is not accurate, maybe add "clouds" after "storm"

That is a good point. Done.

Reviewer 3:

The manuscript by McCoy et al. investigates aerosol-cloud interactions in midlatitude cyclones over the North Atlantic using modelling and the Hohluraun eruption. I think as such the topic is interesting, but the uncertainty has to be discussed much better. My recommendation is to name the motivation and discuss major limitations of the different approaches such that the scientific evaluation of the work is easier. I hope my comments will be useful for improving the *manuscript.*

Thanks- we have tried to more fully explore the uncertainty in our analysis and appreciate the reviewers help in improving our paper.

General comments

I recommend to provide more information/discussion on uncertainty and the motivation of some specific choices in the methods for this work. I understand that one would want to highlight the positive results that seem to provide a conclusive story, but I recommend to more openly discuss the uncertainty in such work.

In my opinion, meteorological variability has a large impact on the perceived aerosol-cloud interaction, no matter whether we look at observations or modelling.

We agree that meteorology controls the majority of cyclone behavior, but it does seem like once we remove variability associated with meteorological variability there is still a signal associated with aerosols. This result appears to be present in both highly idealized simulations and observations. Overall we estimate an impact of aerosols from a standardized perturbation in CDNC that is less than 30% of the response to meteorology- so meteorology still dominates cyclone behavior.

My suggestion is to clearly high- light it for supporting an open debate and helping the reader in assessing the results. When we look for instance at the Holuhraun case, we have very few cyclones that have been affected by excessive amounts of sulphate, i.e., 10 cyclones in total according to Fig. 8. Half of these cyclones show an increase in CLWP, but that is within the range of CLWP anomalies that also naturally occur in the absence of SO4 perturbations. The other half of the cyclones with above-threshold perturbations in SO4 show, however, almost no change in CLWP and this includes the cyclone with the largest SO4 pertur- bation. I would state this explicitly in the text.

See comments above, we have removed this.

In addition to meteorological variability, I wonder how the regionally limited increase in aerosol affects the radiation transfer, thus the temperature gradients and possibly the cyclone/WCB statistics, based on which you construct your argument that aerosol- cloud interaction is the driver of CLWP increases. Have you analysed the changes in the temperature distributions? This would be important for understanding the physical mechanisms behind the model results.

Removed. See above.

The abstract could be a little longer, e.g., it does not state which model and satellite data has *been used, and should more clearly state the uncertainty assessments, e.g., uncertainty in assumptions about the eruption.*

Very true- we have expanded the abstract- thanks.

p.1, l. 20: "liquid water amount and thus the albedo" The cloud albedo depends on the number and size of droplets. I also wonder whether "constraining predictions of the 21st century warming" is a good word choice as the warming will depend not only on the physics, but also on the socio-economic development. As such we will always have a spread in long-term projections into the future. In any case, citing of references would be useful here.

Optical depth is a function of both liquid content and CDNC, not counting whether the lifetime effect changes cloud fraction. All else being equal we should expect that increasing liquid water amount should increase albedo. Since the range of possible climate sensitivity is strongly affected by the assumed strength of aerosol indirect effects if we had a tighter constraint on indirect effects we should have a tighter constraint on climate sensitivity. This statement implies that it is for a given emissions scenario, which should give a more constrained prediction based on a better constrained climate sensitivity. We have tried to expand this section to better explain what we meant.

p.1, l. 22: "thermal contrasts" alone are not enough to form a cyclone. It might be best to just delete that sentence.

Removed.

p.3, l.7: I am a little bit surprised that both the configurations with and without explicit convection use the same vertical resolution. Maybe you can explain why you have made that choice. Would you expect the results to differ when you also change the vertical resolution?

We are using the model configuration based on the operational model used for CMIP and trying to make as few changes as possible.

p.3, l. 14-15: Do you mean that the exponential decay starts at the surface or above 5km? Both seems to be tricky, unless there is observational evidence for it, since aerosol is typically well mixed in the boundary layer, but only few places have a deep BL of 5 km. Maybe use cm-3 instead of /cc to be consistent with your results section.

The exponential decay starts at 5km. Given that it is constant and non-interacting it is not intended to be highly physical. We also tried a constant vertical profile, but this generated a great deal of ice due to the highly simplistic ice nucleation used in the simulations at high altitudes and the results were clearly unrealistic.

p.3, l.18: Please clarify "non-interacting". I guess you mean that no complex aerosol parameterisation is coupled to the atmospheric model, but you prescribe the aerosol *concentration as function of vertical velocity and let the aerosol interact with the radia- tion and clouds in the model (such a setup could also be interpreted as "interacting").*

Thank you- yes- what we meant was that precipitation does not deplete the aerosol and no new aerosol is generated. 'Fixed' would have been better. We have changed this to clarify what was meant.

p.3, l.19: Is there a reason why you have chosen to increase the aerosol just in this channel? Such a setup generates a steep (artificial) gradient in aerosol that might change your temperature gradients and thereby the cyclones.

This setup was chosen because it was simple to implement and didn't require any assumptions regarding the aerosol gradient. The SST is fixed in the simulations so the aerosol shouldn't change the overall temperature profile beyond changing the atmospheric temperature. We do not think that the step function of CCN should affect our results since their purpose is to give us insight into how to analyze the observations- however, this is something that we plan to look at more in the future with more simulations so we will be looking at step function and gradiated channels for the purpose of understanding model behavior more clearly.

Section: 2.2.2: I think if you could add the uncertainty range of these estimates and maybe even systematically test the effect of such a range on your results, the work could be a much better contribution. Later in the results section you touch on that type of uncertainty. Maybe you could motivate it here already.

Removed- see above. We will consider this in a future paper more fully examining these results.

p.4, The first paragraph is partly redundant with the method section. Maybe you can merge the text.

We have substantially re written both the methods section and this section and hopefully it flows better now.

p.5, l.1-2: Is this due to the simple parameterisation that you have implemented into the model? In either case I would mention it here again, because the way it is currently written suggests that what your model tells us is a fact and that fact "warm rain process" seems to contradict what one would expect for precipitation formation in midlatitude cyclones (in reality).

That was poorly worded, we have altered the text to make it clearer we just meant the model.

p.6, l.8-9, Fig. 2: It is not clear why you get large CDNC in the cyclone center, typically a decline going outward, but than again an increase in CDNC to the southwest. Could you argue that this is something you would expect? Here I would also want to read more about the comparison of the CDNC of MERRA and MODIS to judge the quality of the re-analysis.

We have expanded this discussion, but the CDNC retrieval outside of the regions of low lying cloud (such as the cold front) are not reliable and the increase toward the center of the cyclone is not likely to be robust.

p.7, l.26: "enhancement of cyclone properties" I would speak of changes of cyclone properties.

Altered- thanks.

p.8, l.5-6: MERRA assimilates, however, observations that have experienced a poten- tial effect of the aerosol on the meteorology. So, this might not be as conclusive as one would hope.

Very good point- we were concerned about this and have undertaken a separate and more extensive analysis of the MERRA2-MODIS CDNC products (McCoy et al., 2017). It does seem like there is a reasonable agreement between these products and long term changes due to pollution and volcanic degassing that cannot be explained by meteorology. We also added evaluation of the CLWP-WCB relationship partitioned into high and low CDNCSW populations (inferred from MERRA2) to our analysis and see that MERRA2 CLWP actually has the opposite behavior to the observations suggesting that the MERRA2 analysis is not baking in the increase in CLWP with increasing CDNC.

p.9, l.18-20: These are big implications, but how could we know that we would get the same answer when we used other models or other volcanic eruptions, given the uncertainties and variability?

You are correct that assigning a systematic bias to GCMs was not fair. Some GCMs have extremely strong lifetime effects that are sure to exceed the observations shown in our study. This sentence has been removed.

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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. Using high-resolution, convection-permitting global aquaplanet simulations we 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 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 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. 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).

In general for warm rain processes, enhanced aerosol that can act as cloud condensation nuclei (CCN) should enhance cloud droplet number concentration (CDNC, the first indirect, or Twomey effect) (McCoy et al., 2017b;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, 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), 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. 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 the effects of aerosols and meteorology on cloud properties. 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.

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). 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 located over ocean are considered. All observations that are over land are removed from the composite.

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.

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 to assess the robustness of our analysis.

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 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). 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 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 source will yield insight into the observational uncertainty surrounding CDNC.

2.2.5 Albedo

Finally, we examine changes in cyclone albedo due to changes in cloud properties. We utilize albedo from the CERES 3-hourly observations, where the 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 cloud property retrievals(Marchand et al., 2010) and potentially allowing examination of broken cloud cover, which is a prominent feature in midlatitude cyclones and has the ability to substantially influence their brightness (McCoy et al., 2017c).

The 3-hourly albedo is averaged to create a daily-mean albedo. This data is provided in the CERES SYN1DEG data set edition 4(Wielicki et al., 1996). Cloud fraction (CF) from MODIS and geostationary satellites provided in the CERES SYN1DEG data set were used to examine total cloud cover and were averaged in the same way as albedo to create a daily mean cloud fraction. To calculate the shortwave (SW) forcing consistent with changing albedo 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).

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. 1. 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 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-aerosol indirect effect to model resolution in an idealized aquaplanet setting. These models were a GCM-surrogate model and a convection-permitting model. The GCM-surrogate model provides a comparison to the resolution of a typical GCM and was run at 1.89°x1.25°. 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 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 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 intercomparison of simulations at scales ranging from 1-16km show minimal change to the mean statistics of simulated cloud fields (Field et al., 2017) giving 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) was used for all clouds in the convection-permitting simulation and for large-scale cloud cover in the GCM-surrogate simulation. 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 $CDNC = 0.5Nw^{0.25}$ with N being accumulation mode aerosol number concentration and w being updraft velocity limited such that at w=16m/s CDNC=N, and 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.

It is important to note that an increase in CDNC with increasing CCN 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 a ‘fixed-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 above. In these simulations 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 effect, but our results are unlikely to be qualitatively dependent on the simplistic activation scheme chosen here.

3. Results and discussion

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

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.

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 2000km 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). The relationship between WCB flux and rain rate appears to be relatively invariant as a function of model resolution in the aquaplanet simulations analyzed in this study and aerosol concentration (Fig. S3). This is also supported by analysis of other GCMs(Field et al., 2011). Use of this 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.

Essentially, 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. It is interesting to note that the frozen water path in the cyclones did not change between control and enhanced aerosol experiments, indicating that this aerosol-cyclone indirect effect acts through the warm rain process within these 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 lifetime effects 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 MAC-LWP observations of cyclone-composited CLWP and aquaplanet simulations are shown in Fig. 2. To compare cyclone composites in similar meteorology conditions the cyclone composites are shown stratified by WCB moisture flux into regimes of 1-3, 3-5, and 5-7 mm/day. 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 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. Use of a cloud scheme would increase the CLWP and bring

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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. 2f and agree qualitatively with MAC-LWP observations, although the difference between different WCB moisture flux regimes is not as strong and the cyclones are significantly more diffuse.

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One consistent behavior observed across the aquaplanet simulations and observations in Fig. 2 is the enhancement in CLWP with increasing WCB moisture flux. By stratifying the cyclones simulated in the UM at GCM-surrogate and convection-permitting resolutions by the WCB moisture flux we find that WCB moisture flux plays a significant role in determining the CLWP (Fig. 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 CCN available to the cyclone and hence CDNC result in a different CLWP?

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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.

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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.

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. 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 as a proxy for CCN (McCoy et al., 2017b;McCoy et al., 2015;McCoy et al., 2017a). This use of the sulfate mass proxy is advantageous because it is not susceptible to retrieval error and because MERRA2 does not have a parameterized cloud-aerosol indirect effect, further simplifying its interpretation as a proxy of CCN.

Using the daily mean MERRA2 SO4 we calculate a CDNC proxy within cyclones following the relationship established in previous studies (McCoy et al., 2017b). An enhancement in proxy CDNC observed by MODIS and inferred from MERRA2 occurs in the southwest quadrant that is likely to be the source of moisture and aerosol for the cyclone (Fig. 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 $CDNC_{SW}$. 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, MERRA2 are shown in Fig. S6ab. While MERRA2 infers enhancement in CDNC in the southwest quadrant, MODIS shows a higher CDNC in the north part of the composite, which is likely due to retrieval bias at low sun angles and from heterogeneous cloud. However, the inter-cyclone variability in both cyclone-mean CDNC and CDNC$_{SW}$ observed by MODIS and inferred from MERRA2 are in agreement (Fig. S6d) and when MERRA2 is sampled where MODIS can perform a retrieval, the pattern of CDNC within the mean cyclone composite is in better agreement (Fig. S6cd).

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 enhancement. When we compare the observational record of cyclone-mean CLWP by stratifying it into the top and bottom third of observed CDNC$_{SW}$ inferred from MERRA2 and observed by MODIS (Fig. 4cd) a systematic separation in mean CLWP between high and low CCN cyclones becomes apparent (Fig. 3ab).

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One potential caveat to this approach is 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, if the same procedure 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 low CDNC$_{SW}$ cyclones have a higher CLWP. That is to say, they have the opposite behavior of the observations from MAC-LWP (Fig. 3d). 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.

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It should be noted that there is some sensitivity to whether the cyclone-mean CDNC is used to stratify the cyclones or if CDNC$_{SW}$ is used (Fig. S7ab), but the CLWP for high CDNC cyclones is still higher than the CLWP for low CDNC cyclones if the cyclone-mean CDNC is used. Additional sensitivity tests of use of the southern half of the composite and the south-east quadrant of CDNC to stratify cyclones are shown in Fig. S8 and Fig. S9. Only use of the south-eastern quadrant (Fig. S9) for stratification results in large portions of the high and low CDNC cyclone population being indistinguishable within the standard error. 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 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 $\mathrm{CDNC_{SW}}$ offers a better overall representation of the importation of CCN into the cyclone and it will be used for the remainder of the analysis.

Having examined the difference in cyclone-mean properties between high and low $\mathrm{CDNC_{SW}}$ populations we now examine differences in cyclone-centered cloud structure between these populations. The difference in cloud properties between high and low $\mathrm{CDNC_{SW}}$ cyclones share features between observations and modelling, primarily an increase in the MAC-LWP CLWP in the south-west sector of the cyclone (Fig. 5 and Fig. 6). 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).

Examination of the differences in observed cloud extent between high and low $\mathrm{CDNC_{SW}}$ cyclones exhibit a similar pattern of differences to CLWP with enhanced cloud cover in the south west quadrant of the composites (Fig. 7). It is also interesting to note that at high WCB moisture flux cyclones with high $\mathrm{CDNC_{SW}}$ 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 $\mathrm{CDNC_{SW}}$ data set is used.

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As shown above systematic differences in cyclone coverage and liquid content seem to exist between low and high $\mathrm{CDNC_{SW}}$ populations, but to offer a more accurate prediction of 21st century climate change the key variable to constrain is the change in reflected shortwave radiation due to aerosol indirect effects (Forster, 2016;Stevens, 2015). The difference in cyclone-composited albedo observed by CERES between cyclones whose $\mathrm{CDNC_{SW}}$ are in the top and bottom third of the population is shown in Fig. 8. Albedo increases with increasing $\mathrm{CDNC_{SW}}$ in the western side of the cyclone and is roughly consistent with the regions whose CLWP and CF increased (Fig. 5 and Fig. 7). Low moisture-flux cyclones (WCB<3 mm/day) show relatively little difference in albedo if MERRA2-inferred $\mathrm{CDNC}_{\mathrm{SW}}$ is used to stratify the observational record. However, if MODIS observations are used to stratify the record the difference in albedo is much more pronounced across all WCB moisture flux categories. Overall, the consistency in differences in CLWP, cloud fraction, and albedo support the idea that the difference in albedo between low and high $\mathrm{CDNC}_{\mathrm{SW}}$ 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 simulations bear some general similarities to the observations (Fig. 9) although albedo increases are much more uniform throughout the entire cyclone region. This may reflect the extremely large difference in CCN imposed on the simulations leading to a saturation of the aerosol-cloud lifetime effect.

Having inspected the differences in the structure of cloud properties and albedo between high and low CDNCSW populations we will now examine the difference in cyclone-mean albedo. To do this we first divide the high and low CDNCSW 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. 10a). The mean albedo is higher for the high $\mathrm{CDNC}_{\mathrm{SW}}$ population. When MODIS is used to retrieve CDNC the albedo is on average 0.03 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.

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To calculate the difference in mean albedo between high and low $\mathrm{CDNC}_{\mathrm{SW}}$ 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 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 in the difference between high and low $CDNC_{SW}$ populations as a function of WCB is shown in Fig. 10b.

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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°. This yields a difference in reflected SW between high and low $CDNC_{SW}$ 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. 10b). 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 5.58±0.48 and 4.23±0.63 Wm^{-2} for MODIS and MERRA2-inferred CDNC, respectively (Fig. S10)). A maximum SZA of 60° yields values of 6.50±0.27 Wm-2 and 2.88±0.28 Wm^{-2} (Fig. S11). If all SZAs are included this reverses the position of the low and high $CDNC_{SW}$ cyclones (Fig. S12). 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 considerably higher albedo (Fig. S1). 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.

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3.1.3 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). Using the observational record from 2003-2015 we train a regression model

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$$CLWP = aWCB^{b}CDNC_{SW}^{c} - d \qquad (1)$$

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 1. The regression model explains 62%-67% of the variance in the observed CLWP.

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 1 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. 12 for equation 1 trained using MODIS $CDNC_{SW}$.

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Based on the simple visualization in Fig. 12 (and Fig. S14 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.

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4. Conclusions

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.

Based on the idealized simulations and analysis of observations shown here we have developed and tested the hypothesis that *enhanced CCN 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.

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.

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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).

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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), 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. All authors contributed ideas and helped edit the paper.

Acknowledgments

MERRA2 data was downloaded from the Giovanni data server. CERES data was downloaded through the ceres.larc.nasa.gov ordering interface. DTM and PRF acknowledge support from the PRIMAVERA project, funded by the European Union's Horizon 2020 programme, Grant Agreement no. 641727.

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Table 1 The coefficients for equation 1 based on using CDNC$_{SW}$ observed by MODIS and inferred from MERRA2 sulfate. Coefficients (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	0.95	0.11	18.52	37837	0.79
MERRA2	19.23	0.86	0.19	4.53	49361	0.82

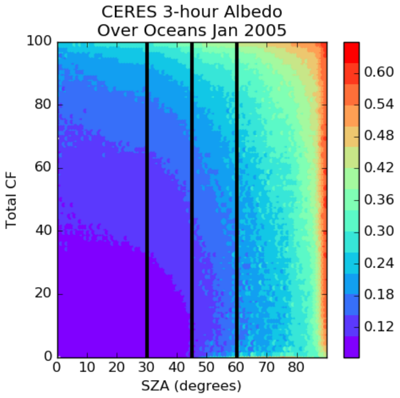
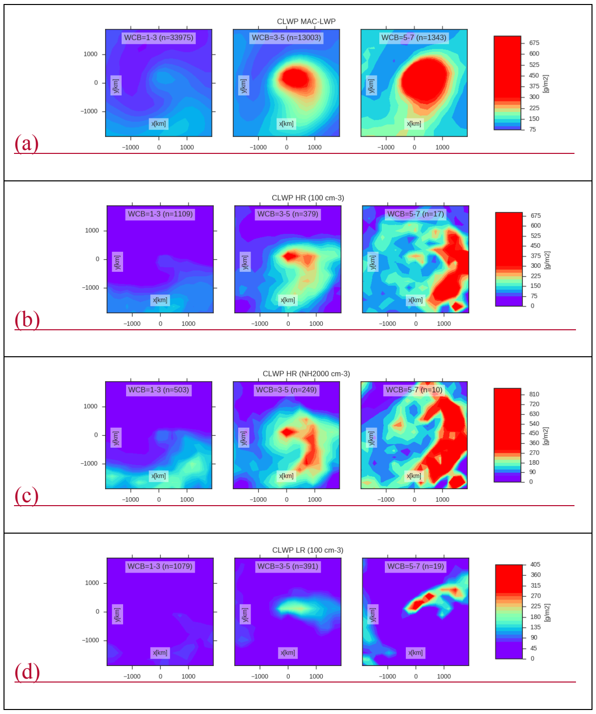


Fig. 1 CERES albedo over oceans binned as a function of cloud fraction and solar zenith angle (SZA) during January 2005. Above a SZA of 45° a strong dependence of albedo on SZA is seen. The SZA cut offs used in this study of 30°, 45° and 60° are shown with vertical black lines. Example CERES albedo is shown in Fig. S1.


CLWP MAC-LWP
WCB=1-3 (n=33975)
WCB=3-5 (n=13003)
WCB=5-7 (n=1343)
1000
0
−1000
y[km]
x[km]
−1000
0
1000
675
600
525
450
375
300
225
150
75
[g/m2]
(a)
CLWP HR (100 cm-3)
WCB=1-3 (n=1109)
WCB=3-5 (n=379)
WCB=5-7 (n=17)
675
600
525
450
375
300
225
150
75
0
[g/m2]
(b)
CLWP HR (NH2000 cm-3)
WCB=1-3 (n=503)
WCB=3-5 (n=249)
WCB=5-7 (n=10)
810
720
630
540
450
360
270
180
90
0
[g/m2]
(c)
CLWP LR (100 cm-3)
WCB=1-3 (n=1079)
WCB=3-5 (n=391)
WCB=5-7 (n=19)
405
360
315
270
225
180
135
90
45
0
[g/m2]
(d)

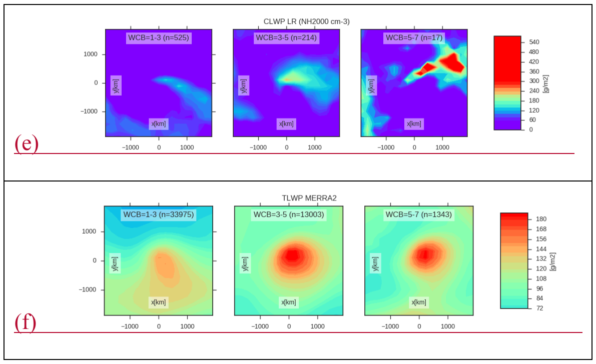


Fig. 2 Cyclone composites showing CLWP from (a) MAC-LWP, (b-c) the convection-permitting simulation in the control and enhanced CCN experiments, (d-e) the GCM-surrogate simulation in the control and enhanced aerosol experiments, and (f) MERRA2. All composites are shown in three bins of WCB moisture flux 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.

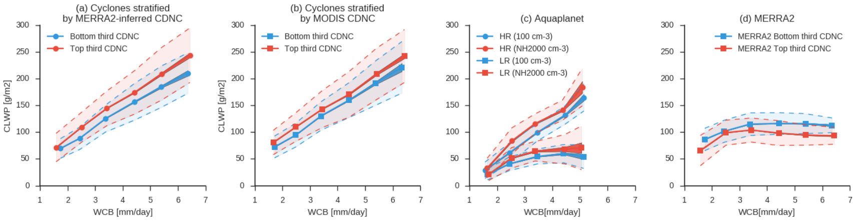


Fig. 3 Comparison between the dependence of cyclone mean liquid water path (CLWP) on warm conveyor belt (WCB) moisture flux as a function of increasing aerosol. The CLWP is binned by WCB moisture flux. The standard deviations in CLWP across bins are shown as shading. The standard error in the mean is shown thick solid lines. Figures (a) and (b) show MAC-LWP observations from 2003 to 2015 stratified by 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 simulations. In the aquaplanet simulations a high aerosol channel is added to the northern hemisphere to investigate the response of cyclone properties and surface 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. 4) are indicated by red and blue lines.

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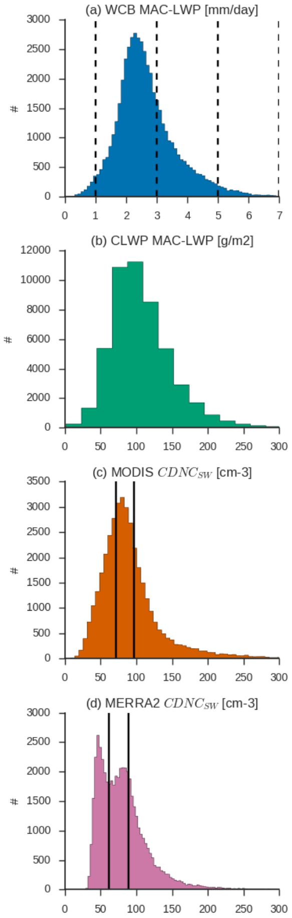

Fig. 4 Distributions of cyclone-mean properties within the 2003-2015 observational record. Units are noted for each variable. The number of composite cyclones with that value is indicated on the ordinate. Warm conveyor belt moisture flux is shown in (a). Cyclone LWP (precipitating and non-precipitating 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) WCB bins are shown with dashed lines.

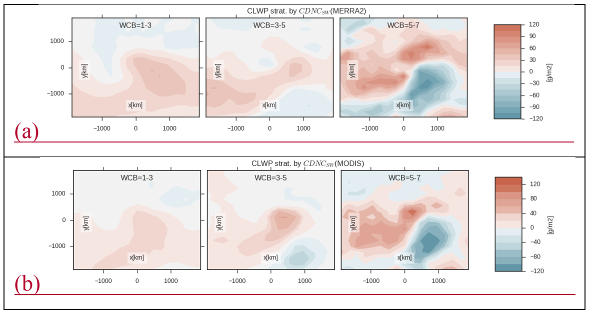

Fig. 5 The difference in cyclone composited MAC-LWP CLWP between the top and bottom third of CDNC$_{SW}$ inferred from MERRA2 (a) and observed by MODIS (b). Composites are shown split into WCB categories as in Fig. 2.

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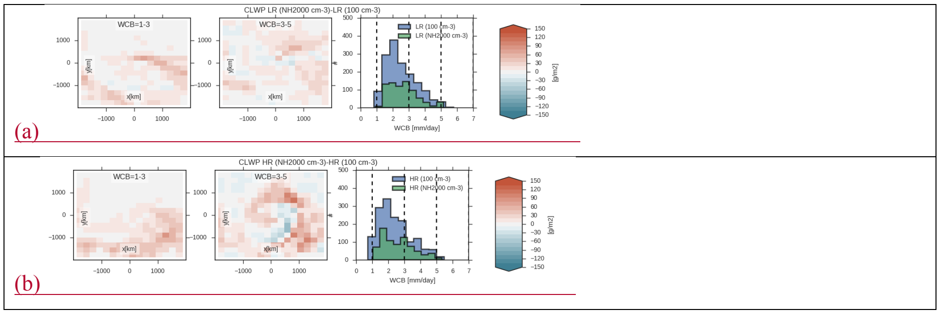

Fig. 6 The difference in mean cyclone composites between the high and low CCN simulations for (a) the GCM-surrogate low-resolution (LR) simulation and (b) the convection-permitting (HR) simulation. Differences in mean cyclone composites for different WCB regimes are shown. It should be noted that the relatively short integration time (relative to the observations) of the simulations did not yield a large number of cyclones with WCB>5 mm/day and only the first two WCB regimes are shown in contrast to Fig. 5. The distribution of cyclones by WCB in the simulations is shown on the rightmost plot.

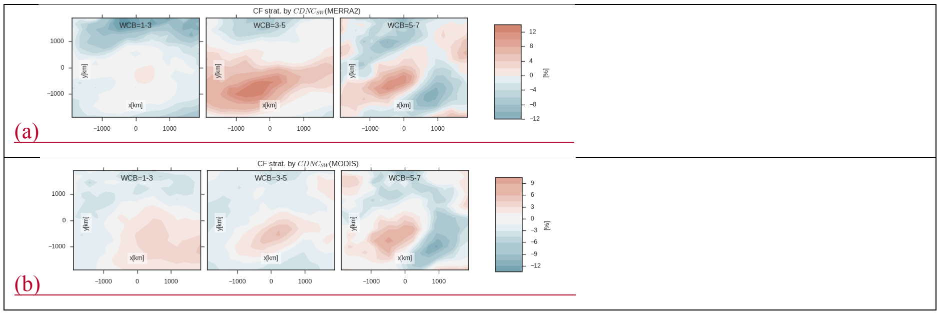

Fig. 7 As in Fig. 5, but showing differences in cloud fraction.

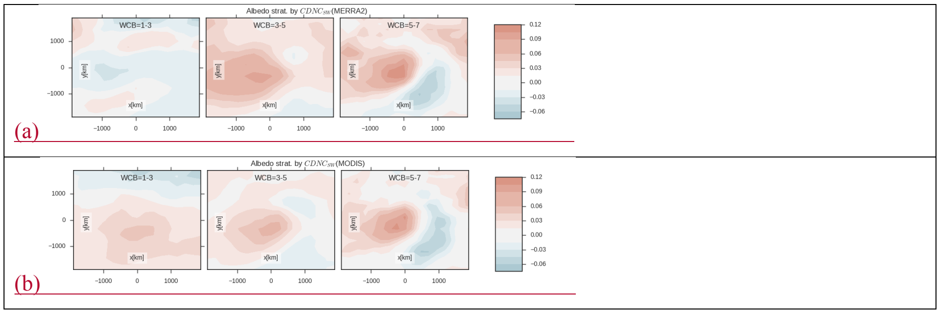

Fig. 8 As in Fig. 5, but showing differences in albedo.

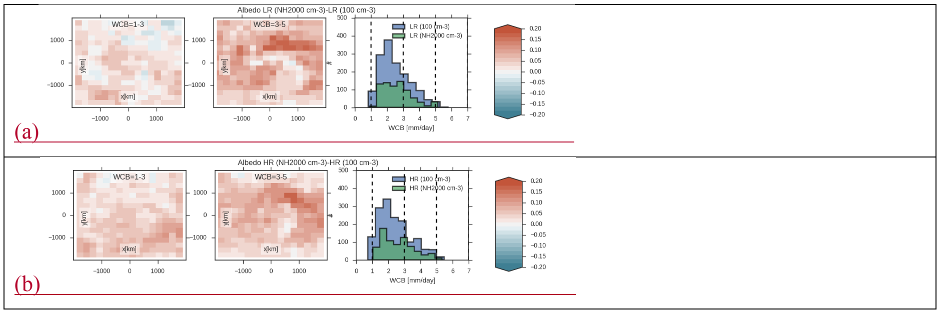

Fig. 9 As in Fig. 6, but showing differences in albedo.

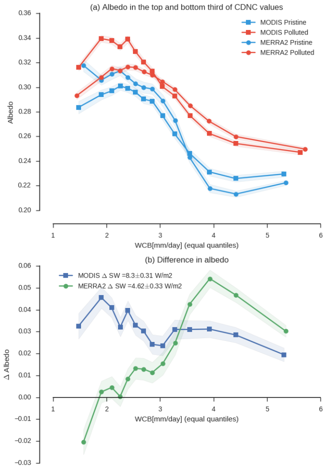

Fig. 10 (a) Cyclone-mean albedo from CERES as a function of WCB moisture flux. Data is shown binned into equal quantiles of WCB moisture flux and separated into the top and bottom third of observed CDNC$_{SW}$. Standard error in the mean is shown using shading. Both MERRA2 sulfate and MODIS-observed 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 mean difference in reflected SW based on this difference in albedo is noted in the legend. The 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. S10,11,12).

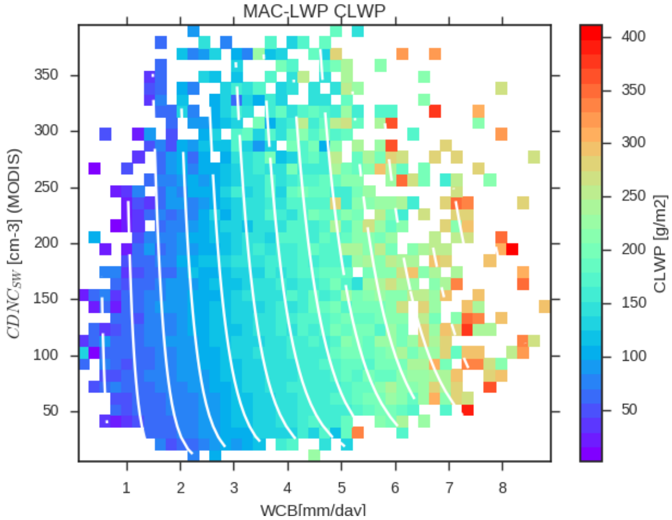

Fig. 11 The cyclone-mean CLWP in units of mm of liquid water observed by MAC-LWP binned as a function of WCB moisture flux and the CDNC$_{SW}$ observed by MODIS. Data is binned into equal size bins for the purpose of visualizing the data record. White lines show contours of constant CLWP as predicted by Eq. 1 and the coefficients listed in Table 1. The dependence of CLWP on CDNC$_{SW}$ inferred from MERRA2 is shown in Fig. S13.

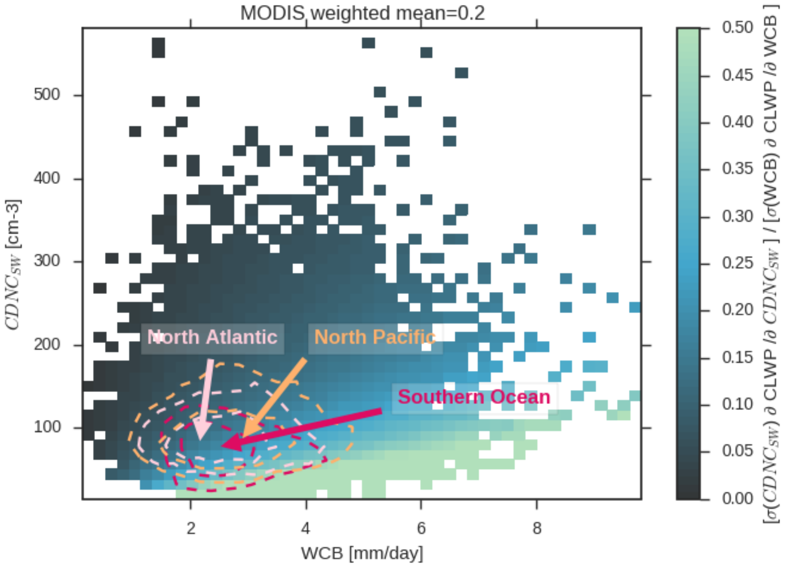

Fig. 12 The relative contribution to CLWP of perturbations in CDNC$_{SW}$ and perturbations in WCB as estimated using Eq. 1 and the standard deviation of each predictor over the historical record. The regression model was trained using MODIS-observed CDNC$_{SW}$ (the same plot is shown for MERRA2-inferred CDNC$_{SW}$ in Fig. S14). 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.