Response to reviewer 1

: The impact of aerosols on the climate system can be quantified through the framework of radiative forcing (RF), whereby aerosols exert an energetic imbalance on top-of-atmosphere (TOA) radiation, which the climate system attempts to restore. Aerosol radiative forcing (RF) is highly uncertain in both models and observations, so deter-mining how to compare and weight the relative value of these estimates remains an open question. This work shows observations and models are in better agreement than previously documented, when care is taken to properly decompose aerosol RF into contributions from aerosol-radiation interactions and aerosol-cloud interactions;both of which can be further decomposed into contributions from direct forcing and rapid adjustments.I found this work to be novel, relevant for the ACPD reader base, and well written. However, some clarifications in the methodology and results, and improvements to the discussion surrounding the presented results, would help improve the manuscript. If the authors can address the minor comments below, I recommend the work for publication.

Reply: We thank the reviewer for their comments and have addressed them below in turn. We note that a few changes have been made to the method and results in this work which are not mentioned below:

1. Correction of the bug in the LWP adjustment calculation - The original LWP adjustment calculation included scenes with no cloud as having zero LWP. This biased both the PD-PI change in LWP and the linear regression of LWP against cloud albedo. In addition, the adjusted forcing from cases with thin overlying ice cloud was applied incorrectly to the RFaci only (and not the LWP adjustment). Correcting these issues results in a small change for the RFaci and LWP adjustment in some models, but the main conclusions remain unchanged.

2. Inclusion of the constraint from Toll et al. (2019) - A new constraint on the LWP adjustment has been included in Figs. 1 and 2. This does not affect the conclusions, but provides an important new constraint that can be compared to the models.

3. Inclusion of the RFaci constraint from (Hasekamp et al., subm) - This is a new constraint based on an improved aerosol retrieval. Again, this does not affect the conclusions, but provides an important new constraint that can be compared to the models.

4. Update of the forcing values from Andersen et al. (2017) - The forcing values from Andersen et al. (2017) were incorrectly determined from the data available in the paper. These have now been updated, but do not affect the conclusions drawn.

5. Inclusion of UKESM1 - To provide an example of how the decomposition can change between model versions, UKESM1 (a descendent of HadGEM2-A and HadGEM3-UKCA) has been included.

Line numbers given are in the “track changes” version of the manuscript.

General Comment 1:: *The authors allude to how previous observational estimates of aerosol RF have generally been smaller than model estimates, and that the presented decomposition brings them into closer agreement. To paint this picture more clearly it would be helpful to provide some numbers from the literature of just how much observations and models were in disagreement previously. One can nitpick and say most of the observational estimates are still on the high end of the presented RFaci model range (Fig 1a). Putting their general agreement into clearer context with previous work,however, will help explain to the reader just how much of an improvement this is.*
Reply: Thank you for this suggestion. We have included the ranges from (Boucher et al., 2013) in the introduction section and modified the conclusion to highlight the better agreement in this work.

General Comment 2:: *The authors briefly allude to the notable divide in the magnitude of the adjustments (and their enhancement of RFaci) between CMIP5 models and AeroCom models, but it would be useful to present some comments on why CMIP5 models are so much smaller, beyond just stating that their LWP change is smaller. Presumably this occurs because most of the CMIP5 models did not really include treatment of indirect effects. Outside of the analysis of RFaci, should we ignore these models then?For the analysis of adjustments, that would only leave us with a few truly different AeroCom models (since most of the data points are just variations of the base same model). Where the authors make claims about the level of agreement/disagreement in the adjustments between models and observations, it would be good to clarify these points.*
Reply: We agree that the number of models used in this analysis is limited and that the number of model families involved is smaller still. However, only three of these families (CanESM2, IPSL, MPIESM) exclude liquid cloud adjustments. Even within a single model family, there is significant variation in the cloud adjustments (CAM5) and the RFaci (HadGEM/UKESM). Although this does not ensure the uncertainty is fully spanned, the agreement between the models and observations (where it exists), is encouraging evidence of progress in constraining the ERFaer and components.

Although the liquid cloud adjustments in the CMIP5 models are typically smaller than those from the AeroCom simulations, it is not clear that this can be explained purely by a lack of sophistication of the parametrisations. Of the six model families in the CMIP5 collection, half of them are a level two (liquid cloud adjustments) or higher. The end of this paragraph (P11L31) has been modified to mention this. “The CMIP5 models tend to have less sophisticated aerosol schemes (Tab. 1), which may explain these weaker adjustments. However, as some models with higher levels of sophistication (e.g. UKESM1, MRI-CGCM3) also have weak adjustments, model sophistication is not the only factor influencing the strength of the adjustments.” This is clearly an area of interest for future work and CMIP6/AerChemMIP may provide the chance to do this, with a larger set of harmonised models.

Specific comments

Page 3, line 13:: *An opportunity is missed in this section to explain why this new approach is more suitable than others. The authors could highlight that other methods,like PRP, are too expensive for analyzing an ensemble of models, for example.*
Reply: The suggested improvements have been included at the end of the introduction section (P3L11) and combined with the original paragraph that leads the methods section (P3L24), leading to a more logical separation between the sections.

Page 4, equations.: *The individual terms deserve more explaining. Is the subscript "cs"different than the "clr" subscript? It appears "cs" is some sort of scaling of clear-sky conditions. In that case, is "cld" plus "cs" supposed to be all-sky conditions? Is "c" different from "cld"?*
Reply: This has been amended to harmonise the subscripts

Page 5, line 10:: *repeated word typo*
Reply: Amended

Page 5, line 9-10.: *Recently, Muelmenstadt et al. in ACPD found that using daily or monthly data instead of 3-hourly biases estimates of forcing in the PRP method. Here 3-hourly output is used from AeroCom but only daily output is used from CMIP5. Does this bias the results? I imagine it cannot fully explain differences between the AeroCom and CMIP5 models, but it may not be a negligible effect. I recommend the authors test this, or at least explain why it may/may not matter with their methodology.*
Reply: We have repeated this decomposition for the AeroCom models using daily data. The change in the values from the decomposition is small. The IWP limit has to be adjusted, with only gridboxes with an IWP more than $40\,\mathrm{g m^{-2}}$ being considered as ice clouds (Table 2.). This is noted in the discussion of the IWPlim (P8L11). Following on from the previous submission, results from the UKESM1 model have been included. This model has similarities to both the HadGEM3-UKCA and HadGEM2-A models, but it was run in a similar setup to the AeroCom simulations. This shows that the negative ERFaci_ice (Fig. 4b) is not due to the setup of the CMIP5 simulations. This hints that the difference between the AeroCom and the CMIP5 simulations may be a chance occurrence based on the chosen ensemble, rather than a consequence of the slight variations in experimental setup.

Page 7, Table 2:: *Are the results presented in this table only from the ECHAM model? If so, that should be specified in the caption.*
Reply: Amended

Page 8, Figure 1a:: *Why is the black circle for C in a different spot along the y-axis for the left-most section of 1a versus the right-most section of 1a? Different studies? Same question for marker B.*

Reply: Thanks for spotting this. The labels referred to the wrong studies - this has now been amended.

Page 10, around line 15.: *Models and observations seem to be in much better agreement on the magnitude of the fl adjustment (Fig, 1b) than on the fl enhancement of RFaci (Fig. 1c) where only one ensemble member of one model is within the lower observational bound of ≈130%. That would infer the models have a larger RFaci in order to match the magnitude of fl adjustment from the observations. That does not seem to be the case, however (Fig. 1a). It would be helpful to clarify this disconnect. Presumably this means models are getting fl adjustment magnitude right but for the wrong reasons, as the authors allude to on Page 10, lines 33-34.*
Reply: Thanks for pointing this out. We have updated the text at P11L12 to highlight this and explain that we believe the agreement on the magnitude of the adjustment is (as suggested) due to the models getting things right for the wrong reasons.

Page 10, line 31:: *It is not clear to me from the figure that models have a larger LWP adjustment and weaker fl adjustment. Are the authors specifically referring to the ECHAM-HAM and CAM5 models? Even for those models alone however, the observations of fl adjustment seem to fall in line with most of the ECHAM ensemble members(Fig 1b).*
Reply: This was intended to refer to the adjustments an enhancements of the RFaci, rather than the absolute values (Fig. 1c). In general, the models have a smaller fl enhancement, even though the magnitude is the same, as they tend towards the upper end of the RFaci estimates. The majority of LWP adjustment estimates suggest a slight decrease in the LWP (e.g Toll et al, (2019). This sentence has now been amended (P12L16).

Supplemental:: *-The final x-axis tick label in Figure S2a is cut off in my copy*
Reply: Amended

Response to reviewer 2

General comment:: *The manuscript covers a topic of high scientific relevance as areduced uncertainty in the aerosol forcing is crucial in order to constrain the anthropogenic impact on climate. The method the authors present may well be valid and interesting and if model and observational estimates do have a closer agreement than what have been presented so far, this is very interesting to the scientific community. Although well written, I found that lack of satisfactory explanations made it hard to fully understand their approach. The research quality is good, but modifications to how it is presented is needed before publication.*
Reply: We thank the reviewer for their comments and have addressed them below in turn. We note that a few changes have been made to the method and results in this work which are not mentioned below:

1. Correction of the bug in the LWP adjustment calculation - The original LWP adjustment calculation included scenes with no cloud as having zero LWP. This biased both the PD-PI change in LWP and the linear regression of LWP against cloud albedo. In addition, the adjusted forcing from cases with thin overlying ice cloud was applied incorrectly to the RFaci only (and not the LWP adjustment). Correcting these issues results in a small change for the RFaci and LWP adjustment in some models, but the main conclusions remain unchanged.
2. Inclusion of the constraint from Toll et al. (2019) - A new constraint on the LWP adjustment has been included in Figs. 1 and 2. This does not affect the conclusions, but provides an important new constraint that can be compared to the models.
3. Inclusion of the RFaci constraint from (Hasekamp et al., subm) - This is a new constraint based on an improved aerosol retrieval. Again, this does not affect the conclusions, but provides an important new constraint that can be compared to the models.
4. Update of the forcing values from Andersen et al. (2017) - The forcing values from Andersen et al. (2017) were incorrectly determined from the data available in the paper. These have now been updated, but do not affect the conclusions drawn.
5. Inclusion of UKESM1 - To provide an example of how the decomposition can change between model versions, UKESM1 (a descendent of HadGEM2-A and HadGEM3-UKCA) has been included.

Line numbers given are in the “track changes” version of the manuscript.

Specific comments:: *Generally, full explanations of how numbers are achieved, what experiments in what models are used etc is lacking. This needs to be fixed before publication. The text would benefit from a bit more focus on reminding the reader what you are doing - linking the results you show to the method and explaining why the results actually show and improved agreement. Not a lot is needed, just a few sentences hereand there.*
Reply: The identification of the experiments is now included in Table 1 and in Fig. 4. Changes to the text have been made to improve readability.

P1, L6:: *“different decompositions” is too vague. Suggest writing this more clearly to get the reader on board with what you are doing.*
Reply: Changes to “different methods of separating the components of aerosol forcing used in model and observational studies.” (P1L6)

P3, L8:: *“...a decomposition is introduced...” Decomposition of what? The reader is left hanging here.*
Reply: This paragraph has been modified, but a relevant sentence now read “for decomposing the ERFaer” (P3L19)

P3, L13:: *"...for decomposing changes...". Suggest changing this to "...for decomposing forcing changes..."?*

Reply: Modified to " for decomposing top of atmosphere radiation changes between a PI and a PD simulation (ERFaer)" (P3L11)

P3, L17:: *"...and second from Nd changes (the RFaci)." Are you saying here that RFaci arises only because of changes in cloud droplet number. This sentence is a bit misleading.*

Reply: The sentence was intended to say that as the main controls on cloud albedo are $\mathcal{L}$ and N_d, the change in albedo can be decomposed into changes in the two quantities. The forcing component (RFaci) is identified with the change in N_d, as this assumes that all the other cloud properties remain constant. This sentence has been modified to make it clearer. (P3L24)

P3, L26-27:: *Use cloud cover and cloud fraction interchangeably. I suggest you stick to one term to avoid confusion.*

Reply: Cover has been replaced with fraction.

P4, Eqs 3 and 4.: *I suggest labelling the terms and refer to these in the explanations from L10.*

Reply: New references to the labelled terms equation have now bee included in the text, which now reads "The aerosol direct effect or RFari can be approximated as SWari$_{cs}$+LWari$_{cs}$. This ignores changes in the surface (ΔSurf.) and the impact of aerosol above cloud (SWari$_{cld}$) but provides..." (P4L20)

P4, L11:: *Approximating RFari in this manner seems to ignore surface albedo changeas well as aerosol above cloud? (alpha_clr_NoA?)*

Reply: That is true, but these terms are small for the majority of the models and this provides a closer comparison with estimates derived from observations.

P4, L27:: *"linear regression": A more thorough description is needed here. For what water amounts does this linearity assumption hold? Cloud albedo, like emissivity, reaches saturation, though of course at much higher liquid water paths. And what is this known change in liquid water path from PI to PD? Is this a global value?*

Reply: While it is true that cloud albedo saturates, for small changes in cloud albedo (as would be expected from aerosol-cloud interactions, the cloud albedo is approximately linear with the liquid water path. This regression is calculated at the gridbox scale (as with the other statistics) to capture the local variations in the relationship. Global averages are only taken at the final step. This has been noted in the methods section - "Note that all of the steps in this decomposition are performed at the gridbox scale." (P4L8). After the correction of the bug in the LWP adjustment calculation, the variation between the adjustments calculated using a log or a linear regression between cloud albedo and liquid water path produces around a 10% difference in the calculated adjustment forcing.

P5, Eq 8:: *Please specify that this is to get the models to resemble the observations*
Reply: Sentence modified to read: "To get a closer agreement between models and observations, the change in liquid cloud fraction (Δf_l) is adjusted in the model output for changes in the ice cloud fraction (Δf_i) following Eq. 8, assuming that the changes in ice cloud fraction are uncorrelated to the occurrence of liquid cloud. " (P6L1)

P5, L9:: *Suggest adding a subheader here, for example "datasets".*
Reply: Amended

P5, L10:: *The descriptions of the experiments should be made clearer. Suggest separating the AeroCom and the CMIP5 explanations. As it reads now it is hard to follow. The experiment name abbreviations are explained two sentences further down.*
Reply: Paragraph has been re-worded

P5, L16.: *Please explain the set up for the anthsca simulations more thoroughly. "...whilst using the same pre-industrial simulation." This is hard to follow.*
Reply: The description has been modified to: "The "anthsca" simulations are the same as the base AeroCom setup, but with present-day anthropogenic aerosol emissions scaled by a factor given in the simulation name." (P6L20)

P5, L17:: *"This demonstrates the impact of changing only the aerosol distribution, rather than also the cloud parametrisations." But you do not change the parametrisations between the runs, do you?*
Reply: The aim was to highlight that both aerosol distributions and cloud parametrisations change between the models included in this study, but in the anthsca runs, only the aerosol distribution changes between simulations. The second clause has been removed, such that the sentence now reads: "While both the aerosol distribution and the parametrisations vary between the models used in this work, the "anthsca" simulations demonstrate the impact of changing the aerosol distribution alone.". (P6L21)

P5, L20:: *"Change is liquid" to "change in liquid"*
Reply: Amended

P5, L24:: *Are these numbers based on numbers from your decomposition method. If so, please specify this*
Reply: They are not, they are taken from the model data itself.

P5, L27:: *How do you calculate this residual? Against what?*
Reply: The residual is calculated as the total ERFaer minus the sum of the components. The sentence now reads "However, the residual of the sum of the components of decomposition compared to the total ERFaer is small (typically less than 10%).."

Caption Table 1:: *The first sentence needs to be more specific. As it reads now, this is total ERF. Also, I would argue that it is the third, not the second column that "identifies the nature...".*

Reply: Caption now reads "The ERFaer (global mean differences between the PI and PD TOA radiation) from the AeroCom (top section) and CMIP5 (bottom) models in $\mathrm{W\,m^{-2}}$. CMIP5 physics ensemble members are shown with the "-p" suffix. The third column..."

Table 2:: *Please specify that RFaci, L and fl refers to three forcing estimates in the de-composition (?). Suggest header above these column and explanation in the caption. In the caption, only RFaci is mentioned. Please add a description of the other two as well.*

Reply: Caption now reads "The impact of ice water path thresholds on the RFaci estimate, the forcing from $\mathcal{L}$ and f_l adjustments and the $\mathcal{L}$ and f_l enhancements of the RFaci. The line in bold..."

Table 2 and text:: *Please explain how you got the numbers in the table, what model and experiments were used*

Reply: The text now links to this table where the IWP limit is discussed and ECHAM-HAM is mentioned in the caption.

Page 7, L2:: *"...aerosol-dependent cloud adjustment (CND)..." The notation used here is misleading. I assume the CND refers to an experiment where these adjustments are removed? Please rephrase and specify with what model and how these simulations were carried out*

Reply: This sentence has been modified to provide a quick explanation of the method, with a longer description included in the methods section: "By removing the aerosol-dependent cloud adjustments using a climatological N_d (CND), .." (P8L6)

P7, L1-3:: *Yes, RFaci is within 10% for the CND runs, but the L and fl are not. Please discuss this in the text.*

Reply: The sentence has been modified to read: "... within 10% of the value calculated through the decomposition in this work, with the forcing from the cloud adjustments decreasing to close to zero as the adjustments are removed (Tab. 2)" (P8L7)

P7, L10-12:: *Does it show that this is a suitable method or that the method is not very sensitive to the choice of threshold?*

Reply: Sentence modified to read: "showing that this method is relatively insensitive to the choice of threshold and hence is a suitable method to account for the effect of thin ice clouds." (P8L20)

Figure 1:: *Please separate the panels more so that the axis labelling becomes more clear.*

Reply: Done

Figure 1, caption:: *Please add "are shown" or similar to "other estimates...".*
Reply: Done

P8, L8:: *You use Delta alpha_c here but this is the same as Delta In AOD in Figure 1a?*
Reply: The $\Delta\alpha_c$ is the forcing from the change in cloud albedo, in this case only liquid clouds are considered. It has been changed to $\Delta\mathrm{SW}_c$ for consistency with Eq. 4.

P9, L1-3:: *Is this shown somewhere? Fig 1.b?*
Reply: This section is modified to read "...observational constraints (Fig. 1a - crossbars). However, when the forcing due to $\mathcal{L}$ adjustments is removed, the variability is reduced, with a lower bound of $-1.26\,\mathrm{W\,m^{-2}}$ and many of the models producing an RFaci estimate around $-0.75\,\mathrm{W\,m^{-2}}$ or smaller (Fig. 1a - markers)." (P9L2)

P9, L4-6:: *Is this shown somewhere? Figure in supplementary?*
Reply: Sentence now reads: "A stronger relationship between ΔN_d and the RFaci is seen for the individual models (Fig. 1a), ..." (P9L7)

Figure 2:: *Is there a residual cloud albedo change? Does the positive value in 2a indicate a negative change in cloud droplet number?*
Reply: There is no residual cloud albedo change, as the Twomey effect/RFaci is the difference between the total cloud albedo change and that from changes in $\mathcal{L}$. The positive values have been removed with the bugfix to the decomposition of the liquid cloud.

P9, L13-14:: *"However, uncertainties from both S1 and S2 are shared with the RFaci estimate." Please write this more clearly.*
Reply: Changed to: "However, uncertainties from both S1 and S2 apply to both the RFaci and the adjustments." (P9L15)

P10, L17:: *"The overall pattern of the forcing from fl changes..." Please add "in the models" or equivalent.*
Reply: Done

P10, L10:: *A bit harsh to state that liquid water path decreases with increased N in observations. This is much debated and state and cloud type dependent. This should be reflected in the text here.*
Reply: This has been softened to read "Several studies have found a $\mathcal{L}$ decrease with increased aerosol or N_d, suggesting a negative adjustment (Chen et al., 2014; Christensen et al., 2017; Sato et al., 2018). However, recent work has suggested that..." (P12L1)

P11, L18:: *The variability of...*
Reply: Amended

Figure 4:*: Please separate the panels more so that the axis labelling becomes more clear.*
Reply: Done

Figure 4:*: Mark which models are AeroCom and which are CMIP?*
Reply: Done

Figure 4:*: Caption (Delta SWc) is this the Delta SWalpha used earlier? If so, chose one notation.*
Reply: The notation has been changed to be consistent. $\Delta\alpha_c$ now refers to the change in cloud albedo, whereas $\Delta\mathrm{SW}_c$ refers to forcing resulting from that change.

Figure 4:*: "longwave changes from cloud properties..." is microphysics a more fitting term here? Fraction is also a cloud property?*
Reply: Changed to "Longwave changes from changes in intrinsic cloud properties"

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Surprising similarities in model and observational aerosol radiative forcing estimates

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Abstract. The radiative forcing from aerosols (particularly through their interaction with clouds) remains one of the most uncertain components of the human forcing of the climate. Observation-based studies have typically found a smaller aerosol effective radiative forcing than in model simulations and were given preferential weighting in the IPCC AR5 report. With their own sources of uncertainty, it is not clear that observation-based estimates are more reliable. Understanding the source of the model-observational difference is thus vital to reduce uncertainty in the impact of aerosols on the climate.

These reported discrepancies arise from the different ~~decompositions of the~~ methods of separating the components of aerosol forcing used in model and observational studies. Applying the observational decomposition to global climate model output, the two different lines of evidence are surprisingly similar, with a much better agreement on the magnitude of aerosol impacts on cloud properties. Cloud adjustments remain a significant source of uncertainty, particularly for ice clouds. However, they are consistent with the uncertainty from observation-based methods, with the liquid water path adjustment usually enhancing the Twomey effect by less than 50%. Depending on different sets of assumptions, this work suggests that model and observation-based estimates could be more equally weighted in future synthesis studies.

1 Introduction

Acting as cloud condensation nuclei (CCN) and ice nucleating particles (INP), aerosols can modify the cloud droplet number concentration (N_d) and the ice crystal number concentration (N_i). An increase in N_d can impact the reflectivity of a cloud (Twomey, 1974), resulting in a cooling effect on the climate known as the radiative forcing from aerosol–cloud interactions

(RFaci) or the “Twomey effect”. A change in N_d may also produce cloud adjustments (Albrecht, 1989; Ackerman et al., 2004), resulting in changes to the cloud fraction (f_c) and the liquid water path ($\mathscr{L}$). Similarly, an aerosol-induced change in N_i may change ice cloud properties. The combination of these adjustments and the RFaci is known as the effective radiative forcing from aerosol-cloud interactions (ERFaci). The sign and magnitude of the forcing from cloud adjustments is highly uncertain (Han et al., 2002; Seifert et al., 2015; Gryspeerdt et al., 2016; Malavelle et al., 2017; McCoy et al., 2018) and is a leading contributor to uncertainty in the overall effective radiative forcing from aerosols (ERFaer).

Most global climate models include some form of parametrisation of aerosol–cloud interactions, allowing the ERFaer to be calculated (e.g. Quaas et al., 2009; Ghan et al., 2016). However, uncertainties in the parametrisation of cloud and aerosol processes have led to a large variation in these GCM-based estimates. Satellite and in-situ observations can be used to constrain the magnitude of the ERFaci, typically focusing on the sensitivity of cloud properties to aerosol perturbations (e.g. Feingold, 2003; Kaufman et al., 2005; Quaas et al., 2008; Gryspeerdt et al., 2017; McCoy et al., 2017). These sensitivities can be either used directly to calculate components of the ERFaer, such as the RFaci (Quaas et al., 2008), or used to constrain processes in global models, improving estimates of the ERFaer (e.g. Quaas et al., 2006). However, in many cases, uncertainties and biases in observations can lead to systematic errors in these observation-based estimates of aerosol–cloud interactions (e.g. Quaas et al., 2010; Gryspeerdt et al., 2016; Stier, 2016; Schutgens et al., 2017; Christensen et al., 2017).

Model-based estimates of the ERFaer tend to be larger (more negative), ~~but despite~~ with Boucher et al. (2013) providing a range of -0.81 to -1.68 $\mathrm{W\,m^{-2}}$, compared to -0.45 to -0.95 $\mathrm{W\,m^{-2}}$for observation based estimates. Despite their uncertainties, observation-based studies have previously been given a stronger weight in expert assessments of the ERFaer, leading to ~~a smaller overall~~ smaller overall assessments of the ERFaer (Boucher et al., 2013). Understanding this difference between methods is necessary to improve future estimates of the ERFaer. Uncertainty in the magnitude of the ERFaer comes from three main sources:

S1. **Anthropogenic and natural aerosol properties** Whilst the present day (PD) CCN and INP burden can be constrained, the composition of the atmosphere of the pre-industrial (PI) earth is much more uncertain, creating a significant source of uncertainty in aerosol forcing estimates (Carslaw et al., 2017).

S2. **The sensitivity of N_d and N_i to an aerosol perturbation**. Most climate models include a parametrisation of the impact of aerosol on N_d through droplet activation and the associated radiative forcing from aerosol–cloud interactions (RFaci/Twomey effect). Variations in the parametrisation of unresolved vertical velocities between models leads to a strong variation in this sensitivity between climate models, despite the similarity of their aerosol activation parametrisations (Gryspeerdt et al., 2017).

S3. **The adjustment of clouds to a change in N_d or N_i**. The magnitude of cloud adjustments (such as changes in f_c, $\mathscr{L}$ or ice water path) are a significant source of uncertainty. The nature of the representation of adjustments varies between models, with some processes (such as those involving ice) being excluded from many models, leading to a large uncertainty in the magnitude and sign of these adjustments (Heyn et al., 2017).

Isolating these different sources of uncertainty is difficult, complicating the use of observations to reduce model biases. Some observation-based studies aim to constrain the entire ERFaer (e.g. Cherian et al., 2014). However, most studies typically estimate components of the ERFaer due to changes in specific cloud properties, such as the RFaci (e.g. Quaas et al., 2008; Gryspeerdt et al., 2017; McCoy et al., 2018), the change in liquid f_c (f_l) (Gryspeerdt et al., 2016; Christensen et al., 2017), $\mathscr{L}$ (Gryspeerdt et al., 2019) or cloud albedo (Lebsock et al., 2008; Christensen et al., 2017) due to the difficulty in isolating specific processes in the atmosphere. In contrast, model studies are able to isolate the radiative forcing due to aerosol impacts on individual processes (e.g. autoconversion (Gettelman, 2015) or aerosol absorption (Zelinka et al., 2014)), but the coupled nature of cloud properties means that the forcing from the RFaci is generally not extracted from the total ERFaer reported (Boucher et al., 2013).

~~In this study, a decomposition is introduced to create a clearer comparison between model and observational estimates of the ERFaer components using minimal computational time and output~~Existing methods of decomposing the top of atmosphere radiation changes between a PI and a PD simulation (ERFaer) into components typically require multiple model simulations with different permutations of model processes activated (e.g. Gettelman, 2015) or repeated calls to the radiation parametrisation, requiring significant modification of the model code (e.g. Mülmenstädt et al., 2019). In contrast, the method presented here requires only a single pair of PI and PD simulations with a minimal set of model output (see S.I.), allowing it to be applied even to existing model ensembles. ~~The decomposition is shown to compare well to more sophisticated methods and highlights significant agreements between the aerosol forcing estimates by global models and through observation-based methods.~~

2 ~~Methods~~

This study presents a method, building on Ghan (2013), for decomposing ~~changes between a PI and a PD simulation~~ the ERFaer into changes in the surface albedo, the direct effect of aerosols (RFari) and changes in the cloud albedo ($\Delta\alpha_c$) and fraction (Δf_c). The changes in cloud properties ~~can in turn be~~ are separated into contributions from liquid and ice clouds (or high and low clouds if cloud phase is not available). Finally, as the primary controls on liquid cloud albedo are $\mathscr{L}$ and N_d (Engström et al., 2015), the changes in liquid cloud albedo ~~can be~~ is further separated into two terms, one from $\mathscr{L}$ changes and a second from N_d changes (the RFaci)~~.~~, which assumes that all other cloud quantities are held constant. This ERFaer decomposition creates a clearer comparison between model and observational estimates of the ERFaer components using minimal computational time and output. The decomposition is shown to compare well to more sophisticated methods and highlights significant agreements between the aerosol forcing estimates by global models and through observation-based methods.

2 Methods

2.1 Forcing decomposition

To decompose the aerosol forcing into components, two separate model simulations are required, one with PI aerosol emissions and another with PD emissions. The ERFaer is taken as the difference in top of atmosphere (TOA) radiation between these two simulations. Cloudy-sky quantities (x_c) are computed from the all-sky (x) and clear sky (x_{clr}) quantities and the cloud fraction (f_c).

$$x_c = \frac{x - x_{clr}(1 - f_c)}{f_c} \tag{1}$$

The ERFaer is split into longwave and shortwave components. The changes in the SW TOA radiation can be attributed to changes in the cloudy-sky albedo, clear-sky albedo ($\Delta\alpha_{clr}$) and changes in the cloud ~~cover~~ fraction (Eq. 3). The change in the longwave component (ΔLW) can be similarly decomposed into a cloudy-sky (ΔOLR_c), clear sky (ΔOLR_{clr}) and cloud fraction change. Throughout this work, a Δ signifies PI to PD changes. NoA indicates an albedo determined in a clean atmosphere (no radiative effect of aerosol; Ghan, 2013). $F^{\downarrow}$ is the TOA incoming solar radiation. Note that all of the steps in this decomposition are performed at the gridbox scale.

$$\begin{align}
\text{ERFaer} &= \Delta\text{SW} + \Delta\text{LW} \tag{2}\\
\Delta\text{SW} &\approx F^{\downarrow}((1 - f_c)\Delta\alpha_{clr}^{NoA} && \Delta\text{Surf.} \notag\\
&\quad + (1 - f_c)\Delta(\alpha_{clr} - \alpha_{clr}^{NoA}) && \cancel{\text{SWari}_{cs}}\text{SWari}_{clr}\ \text{(clear-sky contribution)} \notag\\
&\quad + f_c\Delta(\alpha_c - \alpha_c^{NoA}) && \cancel{\text{SWari}_{cld}}\text{SWari}_{c}\ \text{(cloudy-sky contribution)} \notag\\
&\quad + f_c\Delta(\alpha_c^{NoA}) && \Delta\text{SW}_c \notag\\
&\quad + (\alpha_c - \alpha_{clr})\Delta f_c) && \Delta\text{SW}_{cf} \tag{3}\\
\Delta\text{LW} &\approx (1 - f_c)\Delta\text{OLR}_{clr} && \text{LWari}_{cs} \notag\\
&\quad + f_c\Delta\text{OLR}_c && \Delta\text{LW}_c \notag\\
&\quad + (\text{OLR}_c - \text{OLR}_{clr})\Delta f_c && \Delta\text{LW}_{cf} \tag{4}
\end{align}$$

The terms can then be connected to the decomposition of the aerosol forcing in Boucher et al. (2013). The aerosol direct effect or RFari can be approximated as ~~$(1 - f_c)(F^{\downarrow}\Delta\alpha_{clr} + \Delta\text{OLR}_{clr})$~~$\text{SWari}_{cs}$+$\text{LWari}_{cs}$. This ignores ~~the~~ changes in the surface (ΔSurf.) and the impact of aerosol above cloud (SWari_{cld}) but provides a comparable value to the RFari estimated using observations (e.g. Quaas et al., 2008). The remaining terms can then be considered as the ERFaci (plus cloudy-sky components of the ERFari), with terms due to changes in cloud properties (~~ΔSW_c~~ΔSW_c) and cloud amount (~~ΔSW_{cf}~~ΔSW_{cf}).

These cloud terms can be further decomposed into changes in liquid and ice cloud (~~Eq.10~~Eqs.10-7), resulting in forcings from changes in liquid (ΔSW_{cl}) and ice cloud albedo (ΔSW_{ci}) as well as the forcings from changes in cloud fraction (ΔSW_{cfl}, ΔSW_{cfi}). The liquid cloud albedo is determined using only gridboxes with an ice cloud fraction of less than 2%. A similar criterion is used for the ice cloud albedo. The forcing from changes in liquid cloud albedo (ΔSW_{cl}, the "intrinsic" forcing; Chen et al., 2014) can then be further decomposed into a forcing from changes in $\mathscr{L}$ and a change in N_d. Using the strong dependence of cloud albedo on $\mathscr{L}$ (Engström et al., 2015), the ERFaci due to $\mathscr{L}$ changes can be determined by a linear regression to determine the sensitivity of liquid cloud albedo to $\mathscr{L}$ (Eq. 10), combined with a known PI to PD change in $\mathscr{L}$. Similar results are obtained when using $\ln\mathscr{L}$ instead of $\mathscr{L}$. The forcing due to N_d changes (the RFaci) is the residual of liquid cloud albedo forcing with the $\mathscr{L}$ forcing removed (Eq. 9).

$$f_c\Delta\alpha_c = f_l\Delta\alpha_l + f_i\Delta\alpha_i \tag{5}$$

$$\Delta SW_c = \Delta SW_{cl} + \Delta SW_{ci} \tag{6}$$

$$\Delta SW_{cf} = \Delta SW_{cfl} + \Delta SW_{cfi} \tag{7}$$

$$\Delta\alpha_l^{\mathscr{L}} = \left.\frac{d\alpha_l}{d\mathscr{L}}\right|_{PD} \Delta\mathscr{L} \tag{8}$$

$$\Delta\alpha_l^{N_d} = \Delta\alpha_l - \Delta\alpha_l^{\mathscr{L}} \tag{9}$$

In many situations, the ice cloud fraction includes clouds with a low optical depth. This means that in situations where a thin ice cloud overlies a thick low-level liquid cloud, changes in the low-level liquid cloud albedo might be mis-attributed as changes in the ice cloud albedo. To avoid this issue, a threshold in-cloud ice water path (IWP) of 8.7 $\mathrm{g\,m^{-2}}$ is required for a gridbox to be classed as an ice-cloud gridbox. This threshold is approximately equal to the MODIS cloud mask sensitivity of an optical depth of 0.4 (Ackerman et al., 2008), following the relationship from Heymsfield et al. (2003). The shortwave forcing from these optically thin cases is assigned to underlying liquid clouds, ~~whereas the~~ assuming that the ratio of the RFaci to the forcing from $\mathscr{L}$ adjustments is the same as in the ice-cloud free regions. The longwave forcing is assumed to originate from the ice clouds, due to the emissivity of these thin clouds. The sensitivity of the decomposition to the IWP sensitivity is investigated in this work.

~~The forcing from changes in liquid cloud albedo (the “intrinsic” forcing; Chen et al., 2014) can then be further decomposed into a forcing from changes in and a change in . Using the strong dependence of cloud albedo on (Engström et al., 2015), the ERFaci due to changes can be determined by a linear regression to determine the sensitivity of liquid cloud albedo to (Eq. 10), combined with a known PI to PD change in . The forcing due to changes (the RFaci) is the residual of liquid cloud albedo forcing with the forcing removed (Eq. 9).~~

$$f_c\Delta\alpha_c = f_l\Delta\alpha_l + f_i\Delta\alpha_i$$

$$\Delta\alpha_l^{\mathscr{L}} = \left.\frac{d\alpha_l}{d\mathscr{L}}\right|_{PD} \Delta\mathscr{L}$$

$$\Delta\alpha_l^{N_d} = \Delta\alpha_l - \Delta\alpha_l^{\mathscr{L}}$$

Changes in overlying ice cloud create a change in the liquid cloud fraction (f_l), but observational estimates of the forcing from liquid cloud adjustments typically assume no change in the ice cloud fraction f_i (Gryspeerdt et al., 2016; Christensen et al., 2017). ~~By assuming that the changes in ice cloud fraction are uncorrelated to the occurrence of liquid cloud~~To get a closer agreement between models and observations, the change in liquid cloud fraction (Δf_l) is adjusted in the model output for changes in the ice cloud fraction (Δf_i) following Eq. 10, assuming that the changes in ice cloud fraction are uncorrelated to the occurrence of liquid cloud.

$$\Delta f_l \mapsto \Delta f_l + \Delta f_i \frac{f_l}{1-f_i} \tag{10}$$

2.2 Datasets

The decomposition is applied to ~~three-hourly model output from the AeroCom indirect effect experiment simulations (Zhang et al., 2016; G along with daily output output from the~~ pairs of simulations from the AeroCom and CMIP5 ~~simulations “sstClim” and “sstClimAerosol” at the native model resolution. Further detail on the models can be found in Zhang et al. (2016) and Zelinka et al. (2014) . The PI–PD difference is determined from two simulations, with~~ intercomparisons. The simulation pairs have prescribed sea surface temperatures and sea ice, differing only in their aerosol emissions. ~~The AeroCom simulations are~~ Three-hourly model output from the AeroCom indirect effect experiment simulations (Zhang et al., 2016; Ghan et al., 2016) is used, with 5 ~~years long,~~ year simulations nudged to present-day meteorology for the years 2006–2010. The CMIP5 ~~simulations are 30 year~~ models make use of the “sstClim” and “sstClimAerosol” simulations, which are thirty years long free-running simulations with ~~a climatological SST .~~climatological SST fields. Further detail on the AeroCom and CMIP5 models can be found in Zhang et al. (2016) and Zelinka et al. (2014) respectively. As a descendent of the HadGEM2-A and HadGEM3-UKCA models, UKESM1 (Sellar, subm) has also been included to provide an additional comparison between different version of the same model (futher details in Mulcahy et al., 2018). It is run in the same configuration at the AeroCom simulations.

To test the accuracy of the decomposition, two additional sets of model simulations were performed using ECHAM6–HAM2.2. The “anthsca” simulations ~~scale the~~ are the same as the base AeroCom setup, but with present-day anthropogenic aerosol emissions ~~in~~ scaled by a factor given in the simulation name. While both the aerosol distribution and the parametrisations vary between the models used in this work, the ~~present day simulation , whilst using the same pre-industrial simulation. This demonstrates~~ “anthsca” simulations demonstrate the impact of changing ~~only~~ the aerosol distribution ~~, rather than also changing the cloud parametrisation~~alone. The CND (constant N_d) simulation replaces the N_d value used in the autoconversion parametrisation with a climatological value, selected to agree with the global mean N_d in the full two-moment run. This removes any aerosol-dependent cloud adjustments, such that change ~~is~~ in liquid cloud albedo is the result of the Twomey effect alone.

3 Results

3.1 Decomposition comparisons

The total ERFaer in the ~~eight AeroCom and eight~~ AeroCom and CMIP5 models varies from -0.36 to -2.30 W m^{-2} (Tab. 1), with the majority of models having a stronger SW component that is partially offset by a smaller positive LW forcing. There is

Model			Net	Total ΔSW	Total ΔLW
AeroCom Indirect Effect Experiment					
ECHAM6-HAM2.2	●	3	−~~1.13~~1.06	−~~2.02~~1.89	~~0.89~~0.83
-CND^1	■	3	−0.41	−~~0.97~~0.94	~~0.56~~0.53
-anthsca1.5^2	▲	3	−~~1.57~~1.49	−~~2.47~~2.33	~~0.89~~0.85
-anthsca2^2	★	3	−~~1.86~~1.80	−~~2.88~~2.80	~~1.02~~1.00
-anthsca4^2	+	3	−~~2.88~~2.80	−~~4.35~~4.24	~~1.47~~1.43
CAM5.3	●	3	−~~1.49~~1.41	−~~2.12~~2.10	~~0.63~~0.69
CAM5.3-MG2	■	3	−~~1.40~~1.30	−~~1.62~~1.55	~~0.22~~0.25
CAM5.3-CLUBB	▲	3	−~~1.75~~1.73	−~~2.42~~2.44	~~0.67~~0.70
CAM5.3-CLUBB-MG2	★	3	−~~1.68~~1.65	−2.47	~~0.79~~0.82
SPRINTARS	●	3	−~~1.05~~0.99	−~~1.23~~1.18	~~0.18~~0.19
SPRINTARS-KK	■	3	−~~1.33~~1.23	−~~1.54~~1.46	~~0.21~~0.23
HadGEM3-UKCA	■	2	−2.30	−2.74	0.44
UKESM1	▲	2	−1.13	−1.35	0.22
CMIP5					
CanESM2	●	1	−0.88	−0.95	0.07
HadGEM2-A	●	2	−1.23	−1.33	0.09
IPSL-CM5A-LR	●	1	−0.74	−0.53	−0.21
MIROC5	●	3	−1.30	−1.78	0.49
MRI-CGCM3-p1	●	3	−1.11	−2.06	0.96
MRI-CGCM3-p3^3	■	3	−1.48	−2.63	1.15
MPI-ESM-LR-p1	●	0	−0.36	−0.24	−0.12
MPI-ESM-LR-p2^4	■	1	−0.63	−0.43	−0.20
Mean			−~~1.12~~1.21	−~~1.52~~1.59	~~0.40~~0.39

Table 1. The ERFaer (global mean differences between the PI and PD TOA radiation) from the AeroCom (top section) and CMIP5 (bottom) models in $\mathrm{W\,m^{-2}}$. CMIP5 physics ensemble members are shown with the "-p" suffix. The ~~second~~ third column identifies the nature of the aerosol parametrisation in the model, (0-direct effect only; 1-RFaci in liquid clouds, no adjustments; 2-with liquid cloud adjustments; 3-parametrised aerosol impacts on ice cloud) following Heyn et al. (2017). Models in italics are sensitivity studies and not included in averages. The icons are used in scatter plots and models of the same family have the same color. UKESM is not an AeroCom model, but has been run in a similar configuration. Ensemble key: ^{1}Constant climatological N_d in autoconversion; ^{2}Scaled anthropogenic emissions; ^{3}Updated cloud scheme; ^{4}Different aerosol forcing data

a significant variation in the magnitude and even the sign of the components of the forcing calculated using the method from the previous section (SI Tab. S2). However, the residual ~~for the decomposition~~ of the sum of the components of decomposition compared to the total ERFaer calculated is small (typically less than 10%), increasing confidence in the completeness of the decomposition as each term is calculated independently.

The decomposition in this work also compares well to other methods. By removing the aerosol-dependent cloud adjustments using a climatological N_d (CND), the RFaci is isolated from the adjustments and is found to be within 10% of the value calculated through the decomposition in this work, with the forcing from the cloud adjustments decreasing to close to zero as the adjustments are removed (Tab. 2). Similarly, the three components of the ERFaci in liquid clouds determined using the sophisticated partial radiative perturbation (PRP) method (Mülmenstädt et al., 2019) match the results of this work to within 15% (Tab. 2). ~~This~~ There is also a close match in the spatial patterns of the forcing from the components between the different methods (Fig. S2). Due to the variability of the cloud field, a higher threshold of 40 $\mathrm{g\,m^{-2}}$ gives very similar forcing values when using daily mean data for the AeroCom models (not shown), although only the three-hourly AeroCom data is used in this work. The similarity of the results between methods suggest that the method introduced in this work is capable of accurately identifying the individual components of the ERFaer.

The estimate of the RFaci is also found to be insensitive to the value chosen for the IWP threshold used to identify ice clouds ~~.~~ (Tab. 2). Although there is a significant change in the RFaci when a ~~0~~1 $\mathrm{g\,m^{-2}}$ threshold is introduced, this is likely due to the occurrence of “clouds” in the model that have ~~no~~ little condensed water and hence are not optically active. However, for larger values of the IWP threshold, the variations in the RFaci are within 10% of the value used in this work. Even with a very large threshold value of 100 $\mathrm{g\,m^{-2}}$, the adjustments as a percentage of the RFaci are within ~~30~~20% of the best estimate, showing that this ~~is~~ method is relatively insensitive to the choice of threshold and hence is a suitable method to account for the effect of thin ice clouds.

3.2 The RFaci

Previous observation-based studies estimating the RFaci have used a limited number of methods. A sample of these estimates using various methods and estimates of the anthropogenic aerosol fraction are included in Fig. 1a. A-Gryspeerdt et al. (2017) is representative of studies (e.g. Quaas et al., 2008) using relationships between satellite observations of aerosol and N_d along with observed cloud properties to convert this to estimate the RFaci. B-Fiedler et al. (2017) use a similar method, but incorporates the observed relationship in a climate model to calculate the RFaci (e.g. Quaas et al., 2006), and C-McCoy et al. (2017) use reanalysis aerosol instead of observed aerosol properties. D-Bellouin et al. (2013) use a model strongly constrained by satellite observations to estimate the RFaci. E-Stevens (2015) combines several lines of evidence that are distinct from the other studies. F-Hasekamp et al. (subm) uses a polarimetric retrieval of aerosol to include more size information and accounts for the lower detectibility limit in satellite retrievals of aerosol. Although other studies place an implict limit on the RFaci by constraining the total ERFaer (Cherian et al., 2014) or a combination of the RFaci and the $\mathscr{L}$ adjustments (Lebsock et al., 2008; Christensen et al., 2017), they are not included here due to the weak constraint they provide on RFaci. Together the observation-based studies suggest a central estimate for the RFaci in the range -0.2 to $-1.0\,\mathrm{W\,m^{-2}}$ (Fig. 1a).

IWP$_{min}$ (g m^{-2})	RFaci	$\mathscr{L}$	f_l	$\mathscr{L}$ (%)	f_l (%)
None	−~~0.19~~ 0.29	−~~0.50~~ 0.37	−0.29	~~263~~ 127	153
~~0 −0.41 −0.50 −0.29 121 73~~1	−0.43	−0.50	−0.29	116	67
5	−~~0.44~~ 0.43	−~~0.50~~ 0.51	−0.29	~~114~~ 119	~~66~~67
8.7 (satellite)	**−~~0.45~~0.43**	**−~~0.50~~0.51**	**−0.29**	**~~111~~119**	**~~64~~67**
10	−~~0.45~~ 0.43	−~~0.50~~ 0.51	−0.29	~~111~~ 119	~~64~~67
25	−~~0.47~~ 0.44	−~~0.50~~ 0.52	−0.29	~~106~~ 118	~~62~~66
100	−~~0.64~~ 0.53	−~~0.50~~ 0.60	−0.29	~~78~~ 113	~~45~~55
CND	−0.42	−~~0.02~~ 0.03	0.07	~~5~~ 7	-16
PRP	−0.51	−0.53	−0.31	104	61

Table 2. The impact of ice water path thresholds on the RFaci estimate~~.~~, the forcing from $\mathscr{L}$ and f_l adjustments and the $\mathscr{L}$ and f_l enhancements of the RFaci. The line in bold is the threshold value used throughout the rest of this work. The bottom rows are the liquid forcing estimates from a simulation with no parametrised cloud adjustment and determined from the standard simulation using the PRP method (Mülmenstädt et al., 2019). Values are in W m^{-2} unless specified.

All the models considered in the present study show a significant $\Delta\mathrm{SW}_c$, typically dominated by changes in liquid clouds (SI Tab. S2). ~~The forcing from the Δin liquid clouds~~ This forcing varies significantly, from $-0.06\,\mathrm{W\,m^{-2}}$ to $-1.44\,\mathrm{W\,m^{-2}}$, outside the range of plausible RFaci generated by many observational constraints (Fig. 1a - crossbars). However, when the forcing due to $\mathscr{L}$ adjustments is removed, the variability is reduced, with a lower bound of $-1.26\,\mathrm{W\,m^{-2}}$ and many of the models producing an RFaci estimate around $-0.75\,\mathrm{W\,m^{-2}}$ or smaller ~~.~~ (Fig. 1a - markers). Considering the models as a whole, there is a weak relationship between the aerosol optical depth (AOD) perturbation and the RFaci (Fig. 1a), due to the weak relationship between AOD and CCN (Stier, 2016). A stronger relationship between ΔN_d and the RFaci is seen for the individual models (Fig. 1a), with the remaining variation being due to differences in the cloud field (Zelinka et al., 2014).

Global patterns of the RFaci (Fig. 2a) show a weak RFaci over land and stronger effect over the ocean, particularly in regions with large amounts of low cloud. This is very similar to a number of observational estimates, which place the majority of the aerosol forcing over the ocean due to a high N_d sensitivity to aerosol, and f_l (e.g. Quaas et al., 2008; Gryspeerdt et al., 2017; Christensen et al., 2017).

3.3 Liquid cloud adjustments

Uncertainties in the aerosol environment (source S1), droplet activation (S2) and cloud processes (S3) all contribute to the total uncertainty in forcing from liquid cloud adjustments, making model-observation comparisons difficult. However, uncertainties from both S1 and S2 ~~are shared with the RFaci estimate~~apply to both the RFaci and the adjustments. By reporting cloud adjustments in f_l and $\mathscr{L}$ as a percentage enhancement of the RFaci (Fig. 1c), the impact of S1 and S2 on the estimate of the

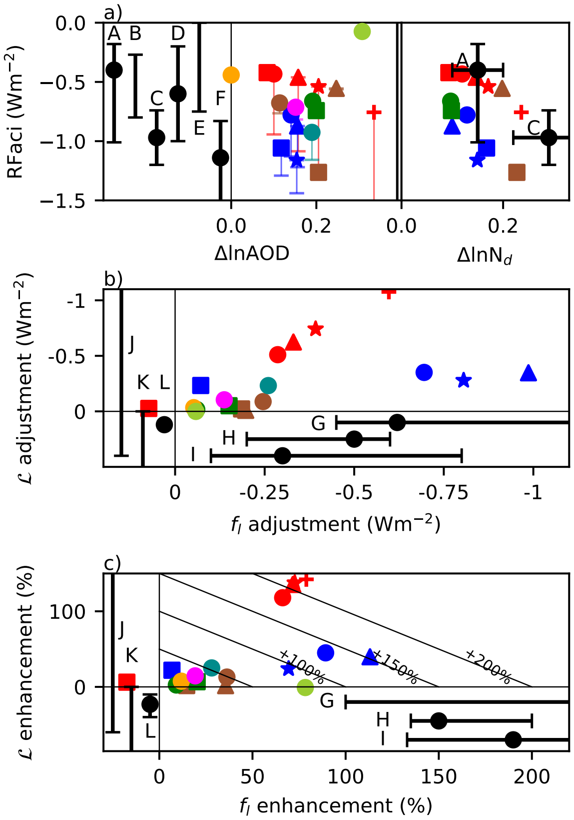

Figure 1. a) The RFaci related to the fractional change in AOD and N_d. Colours and symbols are given in Tab. 1, vertical lines link the RFaci estimates to the "intrinsic" (RFaci+LWP adjustment) forcing. The black points are the observation-based estimates from: A-Gryspeerdt et al. (2017), B-Fiedler et al. (2017), C-McCoy et al. (2017), D-Bellouin et al. (2013)~~and~~ , E-Stevens (2015) and F-Hasekamp et al. (subm). b) Forcing from adjustments in $\mathscr{L}$ and liquid f_c. Other estimates from ~~F-~~G-Andersen et al. (2017), ~~G-~~H-Gryspeerdt et al. (2016), ~~H-~~I-Christensen et al. (2017), ~~I-~~J-Gryspeerdt et al. (2019), ~~J-~~K-Sato et al. (2018), L-Toll et al. (2019) are shown. Not all studies provide a central estimate (black point). c) The percentage enhancement of the RFaci by $\mathscr{L}$ and liquid f_c changes. Diagonal lines are contours of constant total RFaci enhancement.

adjustments can be reduced. This focuses on the uncertainty in the cloud response to N_d changes (S3), simplifying comparisons between models with different anthropogenic aerosol fractions and activation schemes.

The benefit of normalisation of the adjustments by the RFaci is ~~supported~~ demonstrated by the analysis of the ECHAM6-HAM ensemble with varying aerosol emissions (ECHAM6-HAM-anthscaX, red). Although the forcing from both f_l and $\mathscr{L}$ changes in these simulations is very different (Fig. 1b), the enhancement of the RFaci by both effects is the same to within 10% (Fig. 1c). In contrast, the CAM5 microphysics ensemble (blue) has a similar aerosol environment (Fig. 1a) but very different cloud microphysics schemes for each of its members. As such, the variation in the RFaci enhancement from cloud

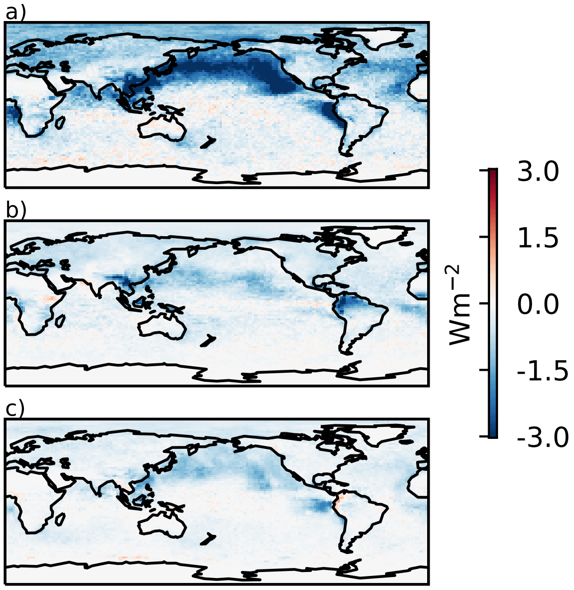

Figure 2. a) The ensemble mean shortwave RFaci. b) ERFaci contribution from f_l changes. c)ERFaci contribution from $\mathscr{L}$ changes.

adjustments is significant among members of this ensemble. This normalisation by RFaci allows the adjustments to be more closely compared with observation-based studies.

f_l **adjustments:** Three recent observational studies using different methods (Gryspeerdt et al., 2016; Andersen et al., 2017; Christensen et al., 2017) find an f_l adjustment that enhances the RFaci by around 130 to 200%. This remains the case when a different anthropogenic aerosol fraction (MACv2; Kinne, 2019) is used in the Gryspeerdt et al. (2016) estimate. The upper bound to the enhancement in Christensen et al. (2017) is unknown, as the RFaci is not reported separately from $\mathscr{L}$ adjustment. This highlights the impact the RFaci uncertainty can have in observational estimates of the enhancement when the RFaci uncertainty is large.

Many of the models, particularly those from CMIP5, have a very small f_l adjustment, producing an RFaci enhancement close to 0%. This explains the smaller mean forcing from liquid cloud adjustments in Zelinka et al. (2014), where only CMIP5 models were used. The largest model estimates of f_l adjustments are of a similar magnitude to the observational estimates, with an enhancement of around 100%. While some models are more similar to the observation-based f_l adjustment forcing (Fig. 1b) than the f_l enhancement (Fig. 1c), this is due to the model RFaci estimates typically being stronger than the average observation-based estimates (Fig. 1a). The overall pattern of the forcing from f_l changes in models (Fig. 2b) is similar to that from Gryspeerdt et al. (2016), with a stronger forcing around the edges of the stratocumulus regions, but a weaker forcing in the North Pacific. This is likely related to the mean-state f_l, as increasing the f_l is difficult if the f_l is already high.

$\mathscr{L}$ **adjustments:** Observational estimates of $\mathscr{L}$ adjustments are difficult to interpret (Neubauer et al., 2017). Several studies have found a $\mathscr{L}$ ~~decreases with increased~~ decrease with increased aerosol or N_d, suggesting a negative adjustment (Chen et al., 2014; Christensen et al., 2017; Sato et al., 2018)~~, but recent studies have~~. However, recent work has suggested that this decrease may overestimate the impact of aerosols on $\mathscr{L}$, supporting a weak $\mathscr{L}$ response to aerosol ~~(Malavelle et al., 2017; Gryspeerdt et al., 2019)~~ (Malavelle et al., 2017; Gryspeerdt et al., 2019; Toll et al., 2019). In contrast, all the models with a significant RFaci also produce a positive $\mathscr{L}$ adjustment, enhancing the ERFaci. As with the f_l adjustments, the $\mathscr{L}$ adjustments are smaller in the CMIP5 models, due to the smaller change in $\mathscr{L}$ but similar cloud radiative effects (Fig. 3). The CMIP5 models tend to have less sophisticated aerosol schemes (Tab. 1), which may explain these weaker adjustments. However, as some models with higher levels of sophistication (e.g. UKESM1, MRI-CGCM3) also have weak adjustments, model sophistication is not the only factor influencing the strength of the adjustments.

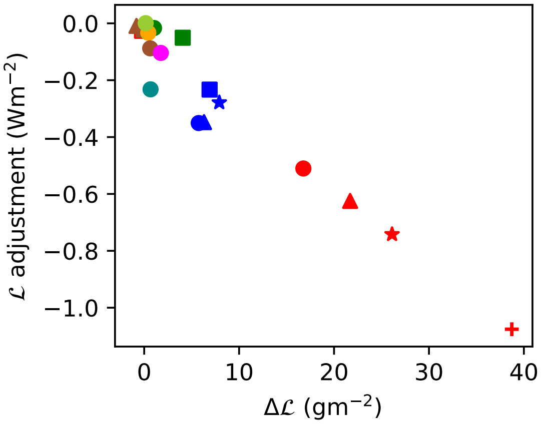

Figure 3. The relationship Δ$\mathscr{L}$ (in-cloud) and the $\mathscr{L}$ adjustment in each of the models.

In almost all of the models, the $\mathscr{L}$ and f_l adjustments have the same sign (Fig. 1b). The different sign of the f_l and $\mathscr{L}$ adjustments in the observation-based studies therefore suggests that inclusion of missing processes controlling $\mathscr{L}$, such as aerosol-dependent entrainment (Ackerman et al., 2004; Xue and Feingold, 2006), may be necessary for models to reproduce the observed relationships (e.g. Salzmann et al., 2010; Guo et al., 2011; Zhou and Penner, 2017; Mülmenstädt and Feingold, 2018).

Although the models ~~have a~~ typically have stronger $\mathscr{L}$ and weaker f_l ~~adjustment~~ enhancements to the RFaci that those from observation-based studies, the models with stronger adjustments have a similar magnitude for the total RFaci enhancement due to adjustments when compared to observations (Fig. 1c). This is an encouraging sign, but highlights the potential for models to produce the right answer for the wrong reason.

3.4 Ice cloud ERFaci

As shown in previous modelling studies (Zelinka et al., 2014; Heyn et al., 2017), the model shortwave (SW) and longwave (LW) total aerosol forcings are strongly correlated (Fig. 4a), indicating a strong role of ice clouds, which dominate the longwave

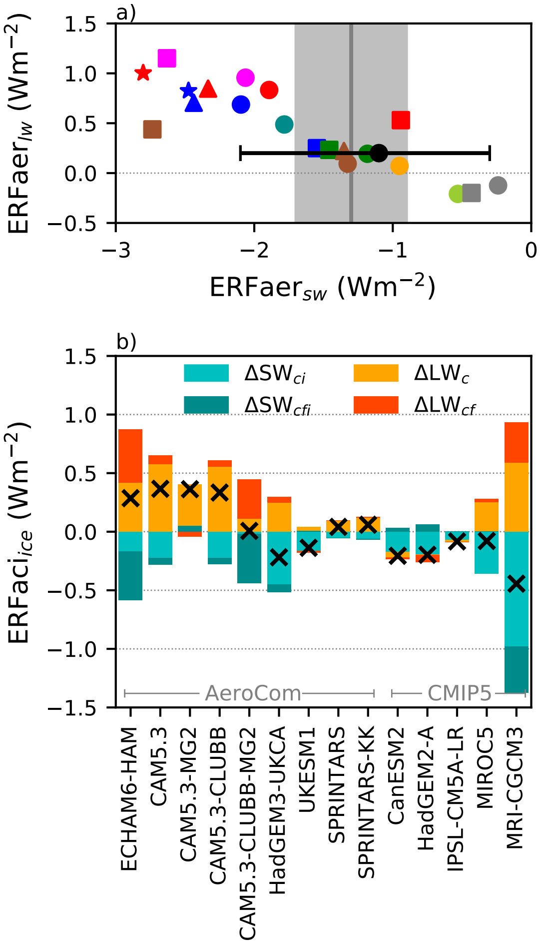

Figure 4. a) The total ERFaer in the longwave as a function of the shortwave ERFaer. The grey range is the estimate from Cherian et al. (2014) and the black circle the expert assessment from Boucher et al. (2013). b) The ERFaci due to changes in ice cloud properties. Shortwave changes from the cloud albedo ($\Delta \mathrm{SW}_{\overline{c}ci}$) and ice f_c (f_i) ($\Delta \mathrm{SW}_{\overline{cf}cfi}$) are shown in blue, including the impact of ice cloud changes masking lower level clouds. Longwave changes from changes in intrinsic cloud properties ($\Delta \mathrm{LW}_c$) and cloud fraction ($\Delta \mathrm{LW}_{cf}$) are in red and yellow. The cross is the total ERFaci from changes in ice clouds.

aerosol forcing (SI Tab. S3). The magnitude of slope of this relationship is smaller than one, such that an increased negative SW forcing is not completely cancelled by a positive LW forcing.

All of the models show an increase in the albedo of ice clouds (Fig. 4b), due to a Twomey-like effect in ice clouds. This is in agreement with current observational studies, suggesting an increase in N_i with an increased aerosol emissions (Gryspeerdt et al., 2018; Mitchell et al., 2018), although there are no current large scale observational constraints on the forcing from ice clouds. This is offset by a decrease in the outgoing longwave from clouds. These effects occur even in models with no parametrised effect of aerosol directly on convective clouds or ice processes, likely through processes such as droplet freezing.

There is a strong variation in the response of high cloud amount to aerosol between the models. The increase in ice cloud fraction exhibited by some models produces a negative shortwave forcing (~~$\Delta\mathrm{SW}_{cf}$~~$\Delta\mathrm{SW}_{cfi}$), but this is closely offset by a positive longwave forcing (~~$\Delta\mathrm{LW}_{cf}$~~$\Delta\mathrm{LW}_{cf}$), such that the net effect from f_i changes in high clouds is close to zero. ~~Note that the $\Delta\mathrm{SW}_{cf}$ is reduced by up to 0.2due to lower lying liquid cloud.~~ The balance between ~~$\Delta\mathrm{SW}_{cf}$ and $\Delta\mathrm{LW}_{cf}$~~ $\Delta\mathrm{SW}_{cfi}$ and $\Delta\mathrm{LW}_{cf}$ varies between the models. The AeroCom models tend to produce a larger longwave effect, resulting in a positive overall forcing (similar to Gettelman et al., 2012), whilst the CMIP5 models generally have an overall forcing close to zero. This may be due to the more detailed representation of clouds and aerosols in the AeroCom models (Tab.1). ~~The~~ While the AeroCom models are nudged to PD horizontal winds (compared to the free-running CMIP5 models), ~~but~~ previous studies show that this does not have a significant impact on the forcing (Zhang et al., 2014) and the negative forcing from UKESM1 (run with the AeroCom setup) further suggests that model setup does not explain this difference. The variability of the ice cloud ERFaci is in contrast to the constant adjustment of +0.2 W m^{-2} used in Boucher et al. (2013), highlighting the current uncertainty in the contribution of ice clouds to the total ERFaer.

4 Discussion

The results in this work have shown that when the individual components of the ERFaer are compared, there is an improved agreement between observations-based and global model estimates. However, there are two important caveats to these results.

The agreement between the observational uncertainty and model diversity, especially for the RFaci (Fig. 1a), is particularly surprising as the RFaci is typically not diagnosed separately from cloud adjustments. Although many models have parameters that can be used to tune the ERFaer, the weak correlation between the ERFaer and the RFaci in the models (r=0.24) further limits the impact of any tuning based on the total aerosol forcing. It should be noted that while the spread in the model RFaci is similar to the spread in the observation-based estimates, many of the models share development pathways (Knutti et al., 2013) and aerosol emissions. Agreement between the models is no guarantee of correctness.

This work also demonstrates that although there is significant variation in the model estimates of the magnitudes of the forcing from liquid cloud adjustments, this variation can be reduced by comparing the adjustments normalised by the RFaci. This accounts for estimates that use a large anthropogenic aerosol fraction (e.g. ECHAM6-HAM2.2-anthsca4), producing a metric that is more closely related to the strength of the liquid cloud adjustments. Uncertainties in observational estimates of the RFaci would introduce uncertainties into the estimate of this enhancement factor, even though uncertainties dependent on the anthropogenic aerosol fraction are significantly reduced by using the enhancement factor. Although there are clear advantages to the RFaci enhancement as a metric for comparing the magnitude of cloud adjustments between models and observation, further work is required to investigate the uncertainty characteristics.

5 Conclusions

Previous synthesis studies have found little overlap between distributions of model-based and observations-based estimates of the ERFaer (Boucher et al., 2013). By decomposing the aerosol radiative forcing from GCMs into components similar to recently developed observational estimates of the ~~aerosol radiative forcing~~ERFaer, this work shows that closer agreement between the model and observational estimates is achieved. ~~This decomposition of the ERFaer is simpler and more computationally efficient to implement than more sophisticated methods (e.g. Mülmenstädt et al., 2019). The~~ In particular, the RFaci in the models investigated is found to be ~~evenly distributed around~~ within current observation-based estimates, although there remains significant uncertainty in these observation-based estimates.

The decomposition shows a large variability in the liquid cloud adjustments. The spatial pattern varies from the RFaci pattern, due to the differing physics involved (Fig. 2), but analysing the adjustments as a function of the RFaci mitigates differences from varying aerosol perturbations and droplet activation schemes among the models. Given the large variation in forcing from liquid cloud changes in models, there is a surprising agreement between the model and observational estimates of the RFaci. However, the $\mathscr{L}$ and f_l adjustments show little similarity to current observation-based estimates. This indicates that further work on the observation-based and model estimates is required before they can be relied upon.

There are significant compensations in the longwave from aerosol-induced changes to high and deep clouds, and the sign and magnitude of the overall effect varies significantly between the models, leaving the overall magnitude of the effect uncertain. While early observational studies have indicated a possible negative albedo forcing in the shortwave from changes in the properties of high clouds (e.g. Gryspeerdt et al., 2018; Mitchell et al., 2018), more work is required in this area.

Although the observational and model estimates display a surprising degree of agreement in many cases, a large degree of uncertainty in the ERFaer remains, particularly in the anthropogenic aerosol fraction and in the sensitivity of cloud properties to aerosol. Even where estimates agree, the uncertainties in the model physics and observational estimates mean that this problem is not yet resolved. However, this decomposition provides an encouraging path forward for future studies. This decomposition of the ERFaer is simpler and more computationally efficient to implement than more sophisticated methods (e.g. Mülmenstädt et al., 2019), but closely matches their results. By showing a significant agreement between components of modelled and observational estimates of the aerosol radiative forcing, this study builds confidence in the global model estimates of the aerosol radiative forcing and shows that where model and observation-based studies can be more accurately compared, their similarities become increasingly clear.

Code availability. HadGEM3 code is available from https://code.metoffice.gov.uk/ (last access: 01 May 2019) for registered users. To register for an account, users should contact their local institutional sponsor or Scientific_Partnerships@metoffice.gov.uk.

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