Anonymous Referee #1 Received and published: 19 January 2020

General

The paper presents a modelling study on the direct aerosol effect on climate. The authors distinguish between clear and cloudy skies. The approach is probably state of the art although, however, to my opinion, a very simple one. Let me start with my general impression: We have satellite lidars delivering global 3-D aerosol distributions (profiles!) with detailed aerosol typing (in terms of optical, microphysical and even chemical composition and thus refractive index characteristics) around the globe from the surface up to stratospheric heights and also producing 3-D distributions of clouds layers, their thermodynamic phase, frequency and cloud cover. In addition, we have sophisticated passive remote sensing techniques, again, delivering very detailed information on cloud layering, cloud heights, cloud types, cloud cover, and thermodynamic phase. In view of all the available and complex global 3-D cloud and aerosol data sets, I am a bit surprized that teams of modellers still use rather simple approaches (here Eq.(1)) to investigate and estimate the role of aerosols (natural and anthropogenic ones) in the climate system with the goal to answer the very important and 'ultimate' question: What is the contribution of anthropogenic aerosols to climate change? Even if global MODIS column information on AOD (and maybe cloud occurrence and cover?) is included in the study, . . . is that sufficient to obtain a realistic picture on aerosol effects on climate? The global aerosol distribution (profiles) used in this manuscript is rather simple so that question arises: Does the modelled global aerosol climatology really reflect the real world?

Response: To the authors it is unsure whether the Reviewer has understood the advanced global modelling and approach applied in this study. It is incorrect that we use Eq 1 to investigate anthropogenic and natural aerosols, our application of Eq 1 is used for anthropogenic aerosols. However, the global modelling includes natural aerosols and simulations of aerosol vertical distribution on a high temporal scale. The Reviewer mention a large set observational data the authors are aware of, but the apparent misunderstanding by the Reviewer is that these observational datasets only provide the present aerosol abundance. E.g. we have already referred to two studies comparing the AeroCom models against CALIPSO in the discussion (Koffi et al., 2016; Koffi et al., 2012). In this study we have the aim of investigate anthropogenic aerosols and the observations cannot provide a clear distinction between anthropogenic and natural aerosols and therefore model information is required. We refer to several studies using observational studies to estimate the cloudy sky RF, and we mention they have large limitations not only on abundance, but also natural and anthropogenic aerosols have large differences in aerosol optical properties. After presenting the earlier studies using observations, we have noted the following: '_Note that the above-mentioned studies investigate the present, total aerosol abundance which consist of anthropogenic and natural aerosols, whereas in terms of RFari only the anthropogenic aerosols are considered_**'. In the beginning of the introduction we have now underscored that the estimate of RF is for anthropogenic aerosols. We disagree with the Reviewer that our approach is simple, even though Eq is simple.**

Maybe, there are meanwhile modelling groups and thus papers in which the measured global aerosol distributions and measured global cloud distributions are used to model the impact of aerosols on global climate conditions, and these authors here just want to offer an alternative way, a more simple, rather basic approach to estimate the aerosol effects on climate? Maybe that is the reason for this simple paper but at the end the main question is still: Can we believe in these results when such a simple approach is used?

Response: see above

And, are you sure that you cover the full spectrum of anthropogenically caused aerosols. What about all the dust in the atmosphere especially over Central and East Asia, is that all natural? Clearly: NO! But how to consider that in the model? Did you consider that in the simulations? Probably not! The paper is worth to be published, no doubt! The list of authors is full of well-known experts, and the paper is a valuable contribution to the climate debate, but the authors should at least try to provide some answers to my concerns. Yes, maybe I am 'naive' . . . as an experimentally working specialist for aerosol and cloud profiling, and my comments indicate that I am not familiar with the modern modelling world but I am probably not the only one who has trouble with the concept and content of this paper. Maybe, I completely missed the point and the overall message of the paper, but again, I will be probably not the only one. So, we need a more critical discussion on the paper approach itself in this paper.

Response: We agree that anthropogenic dust aerosols are not included in the models applied in this work and only a very few in current state of the art models. We have therefore added the following sentence in the discussion: 'All the global models that supplied simulations for this study treat the major anthropogenic aerosol components sulphate, organic aerosols, and black carbon, some also treat nitrate, but none include anthropogenic dust aerosols which have highly uncertain radiative effects.**'**

Details:

P2, l40: Bellouin et al. . .. this is obviously not a publication, there is no year of publication, nothing. So, that is not an acceptable statement. Please improve!

Response: This paper is now published, and we have now included a complete reference.

P2, l50: . . . biofuel BC emission inventory is much higher than used in previous global modelling . . .. Bad wording? What do you want to say?

Response: Sentence rewritten.

P2, l62: Eq (1) is the most basic (trivial) approach, right? Or is there even a more simple one? On the other hand, the atmospheric system is so complex, and modern instrumentation fill the aerosol and cloud data base since 20 years, continuously. You seem to ignore all this! You separate (anthropogenic) aerosol particles in absorbing and non-absorbing ones, nothing else. Is that sufficient? You introduce AC as cloud fraction! Obviously it doesn't matter whether we have one layer, two layers, three layers of clouds, whether we have liquid-water clouds, mixed-phase clouds, cirrus . . .or even complex cloud mixtures and layering, and it is also not essential whether the aerosol is below the lowest cloud layer, between the different cloud layers, etc. . . Just one parameter is sufficient: AC! For the entire globe! For rather different climate zones? One AC value everywhere. . .? This is quiet a surprizing and 'universal' assumption. The other way around, what did I miss here? Please clarify, other readers (not familiar with climate modelling) may think the same. . ., may have the same problem with the paper. Maybe all the referenced papers show that it is sufficient to have just AC to describe the impact of clouds on the aerosol radiative effect around the globe from the tropics to the poles.

Response: All the global models simulate the spatial variation in the vertical profiles of aerosols and clouds and their composition and optical properties. We underscore that the simulations are not only done for a layer, but the study is based on complex global aerosol modelling. See further comments in the response to the main comments. When determining the radiative forcings of

RFcloud, RFclear etc, all model grid boxes are utilized, where the vertical distribution of the clouds and different aerosols and their optical properties will affect the radiative forcing calculations.

P3, l70: aerosols above clouds, below clouds. . . Only these two scenarios, not more are need to be modelled and considered? . . . although the world is full of complex aerosol and cloud layering. . . and large areas over the oceans downwind of polluting continents in the northern hemisphere . . . are 'affected' by this complex layering?

Response: This is clearly a misunderstanding by the Reviewer, it is certainly not only aerosols above or below the cloud. The models applied in this study have between around 20 to 60 vertical layers in the atmosphere. See further comments above.

P3,l93: When using Stefan Kinne's aerosol climatology, did you at least check how good the agreement between CALIPSO aerosol profile observations (in combination with MERRA and CAMS simulations) and Kinne's aerosol climatology is? I speculate: Yes, you did that! My 'spontaneous feeling' is that this quiet simple aerosol profile climatology is not in good agreement with the real world. So, please comment on this!

Response: In the MACv2 climatology the distribution of AOD (where the monthly local statistics of AERONET/MAN corrects the multi-model median of AeroCom phase 1 maps on a regional basis) a distinction is made for AOD from coarse particles (dust, seasalt) and AOD from fine-mode particles (pollution, wildfires). To approximate the aerosol vertical distribution (via vertical scaling of the local monthly column AODc and column AODf data of the MACv2 climatology), scaling factor from 20-year averages from ECHAM5-HAM model simulations are applied. Hereby a single model (ECHAM) was chosen (over a global model median) because in tracer studies and in comparisons to CALIPSO data (paper by Sarah Guibert) this model behaved very well and was not so 'vertical transport-happy' as many other models in that comparison.There was a consideration to replace the vertical aerosol distribution of aerosol with (more observational) data from Calipso. That this has not been included (so far) as CALIPSO data put more aerosol closer to the surface even in comparison to CAMS (e.g. in dust-outflow regions). More importantly CALIPSO data cannot distinguish between fine-mode AOD and coarse-mode AOD. Note than for the study of anthropogenic aerosol only the fine-mode AOD is relevant as anthropogenic aerosol is predominantly an added fraction to the fine-mode AOD. The aerosol vertical distribution also needs to be seen in the context of the cloud altitude placement (The MACv2 climatology distinguishes between high mid and low level clouds, where low clouds are near 1km above the ground, mid-level clouds at ca 3km above the ground). Since random cloud overlap (clouds at 3 altitudes require 8 separate simulations for each permutation) is assumed the cloud-free fraction in MACv2 is on average only at 30%). In MACv2 there is a significant fraction of for optically thin high-only cloud fraction, which may explain a relatively negative forcing for cloudy skies in the comparison. The model description is updated in the manuscript to include information on the vertical profile as follows: *'The Max Planck Aerosol Climatology (MACv2) method combines aerosol column optical properties for fine-mode and coarse-mode sizes (of an AeroCom phase1 model median regionally adjusted by AERONET/MAN monthly statistics) with MODIS surface albedo data, ISCCP cloud properties and vertical scaling by size-mode from 20 years of ECHAM-HAM aerosol simulations. The anthropogenic properties is defined as a fraction of the fine-mode, where the fine-mode AOD scaling factor prescribed from AeroCom phase1 simulations.'*

I would suggest to include a figure with a sketch of your basic aerosol-cloud scenarios considered in the model. Show a cloud layer (provide information on the cloud height, then visualize AC, that means, the cloud should not cover the full sketch from left to right, and then indicate aerosols (just a mixture of black (absorbing) and yellow or white points (non absorbing particles). Scene 1: aerosol below the cloud, Scene 2: aerosol above the cloud layer, Scene 3: aerosol in the clear part of the sketch, if there are more scenes in the model, please continue with further scenes. . ..

Response: The model simulations are complex with (multiple grid boxes and) multiple vertical layers with clouds and aerosols of different properties found at different height, all of which varies with time and geographical location. Since the Reviewer has misunderstood that we've just do simulations for one cloud layer (see above and the comment below), we refrain any further response to this comment.

P5, l127: Result section: My only one question . . . throughout this section. . . was at what height is the cloud layer (for which we have a fixed, constant AC)? Obviously you only consider liquid-water clouds in the lower troposphere. A cloud layer at, e.g., 1 km height (boundary layer top) almost everywhere. . .. around the globe. Maybe it is stated somewhere and I missed it unfortunately. But what about the impact of all the midlevel cloud fields (partly glaciated. . .) and the extended subvisible cirrus fields around the globe. . . , no impact on the aerosol related radiavtive effects?

Response: Again, the model simulations contain complex treatments of clouds at all altitudes around the global and this reviewer comment is a bit off mark. We have included a sentence in the Result section making it clear that although aerosols are found to have a large effect when located above low clouds, all placement of different aerosols types in relation to cloud are treated in the models, be it above, within or below clouds, for different cloud types (low, mid and high, liquid, mixed and ice).

The rest of the paper sounds ok (consistent) . . .. for a non-modelling atmospheric scientists traveling around the globe and measuring the rather complex world of clouds and aerosols in regions with very high amounts of haze and dust (which is partly triggered by human activities) and partly complex aerosol layering up to the tropopause, . . . and, in contrast, in very pristine areas with simple cloud and aerosol layering as in your model.

My 'basic' comments may be confusing but the goal is to improve the paper, not to destroy it.

Koffi, B., Schulz, M., Bréon, F.-M., Dentener, F., Steensen, B. M., Griesfeller, J., Winker, D., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T., Bian, H., Chin, M., Diehl, T., Easter, R., Ghan, S., Hauglustaine, D. A., Iversen, T., Kirkevåg, A., Liu, X., Lohmann, U., Myhre, G., Rasch, P., Seland, Ø., Skeie, R. B., Steenrod, S. D., Stier, P., Tackett, J., Takemura, T., Tsigaridis, K., Vuolo, M. R., Yoon, J. and Zhang, K.: Evaluation of the aerosol vertical distribution in global aerosol models through comparison against CALIOP measurements: AeroCom phase II results, Journal of Geophysical Research: Atmospheres, 121(12), 7254-7283, 2016.

Koffi, B., Schulz, M., Breon, F. M., Griesfeller, J., Winker, D., Balkanski, Y., Bauer, S., Berntsen, T., Chin, M. A., Collins, W. D., Dentener, F., Diehl, T., Easter, R., Ghan, S., Ginoux, P., Gong, S. L., Horowitz, L. W., Iversen, T., Kirkevag, A., Koch, D., Krol, M., Myhre, G., Stier, P. and Takemura, T.: Application of the CALIOP layer product to evaluate the vertical distribution of aerosols estimated by global models: AeroCom phase I results, Journal of Geophysical Research-Atmospheres, 117, D10201, doi:10.1029/2011jd016858, 2012.

Anonymous Referee #2 Received and published: 2 March 2020

In this work, the authors investigate the contribution of aerosol in cloudy skies to the magnitude of the aerosol direct effect (RFari). They results from a collection of global models to show that the contribution to the RFari from cloud skies is small. They also investigate the parameters that affect this between different models, showing that the shortwave cloud radiative effect is the biggest contributor to inter-model differences. This work is within scope for ACP and would be relevant for the readers. I have some comments, particularly regarding the notation and some of the explanation, after which I believe it would be suitable for publication.

Response: we appreciate the nice evaluation and interpretation of our manuscript and constructive comments to improve the manuscript.

Major points

Has the choice for the meaning of RFariclear and RFaricloudy been made in a previous paper? If not, a change in the notation might improve readability. My understanding is that radiative forcings usually sum, such that Eq. 1 could be written as RF ariallsky = RF ariclear + RF aricloudy rather than a cloud-fraction weighted sum. This would improve readability throughout the paper, as “contribution of cloud sky to RFari” is written much more often than RFaricloudy at the moment (perhaps RF aricloudy = AC × RF aci| cloudy). Having a linear sum of terms would also match better with the approximate linear sum ERF aer = ERF ari + ERF aci. This is somewhat a matter of taste, so I understand if the authors prefer to stick with the current notation.

Response: We agree to the nice suggestion to improve the readability of the manuscript by simplify Eq 1. We change RFari$_{clear}$ from the earlier version to RFari$_{clear\text{-}total}$ and similar the cloudy contribution to continue illustrating the term in Fig 2.

Second, while I like the idea of the PCA decomposition, I found it hard to interpret and ended up mostly looking at the correlogram (Fig. 4b). Some more explanation and guidance to interpretation would be useful here.

Response: PCA is a fairly well known, yet complicated, statistical technique used in exploratory data analysis. As such there is limit to how much can be explained in this paper, without making the paper about explaining PCA. What is important to understand is that PCA is a dimension reduction technique, allowing for the visualization of all the variance between the variables in a two-dimensional plot (biplot). When we use a correlogram we only get to see the one to one correlation between variables. With the PCA we can assess the relationship between multiple variables simultaneously. This allows us to get a sense of the degree of influence the variables have on each other. This cannot be seen in a correlogram. However, because the projections of the variables in the biplot are dependent on each other, it can be hard to see the one to one relationship. There is therefore pros and cons to both types of plots, but together they help in the exploratory data analysis.

We have included more description of the method and interpretation of the results, see below on response to other comments.

Does it use only the values in Tab. 1 (the global mean values)?

Response: Yes, table 1 contains all the data used in the PCA. However, as mentioned on line 122 in the ACPD paper. We have two models, LMDZ-INCA and ECHAM-HAM, which have some missing data. In PCA, no missing data can exist in the analysis. If a single record (i.e. in this case a climate model) is missing data for one variable, then the entire record must be removed for all variables. Removing two models would be fairly detrimental to this study, as we have few records (8 climate models) to begin with. For this reason, we use the technique regularized iterative PCA to fill in the missing values with estimates. As we are only using the PCA to get an overview of the relationship among the variables, and we are not trying to create a predictive model for estimating "Cloudy", the use of this imputation technique should be valid. We have added at the top of the paragraph that global mean used in the Multivariate data analysis.

What does it mean that SW_CRF has no contribution to PC1, yet has the strongest correlation to the cloudy sky contribution to the RFari in 4b?

Response: The principle components represent new dimensions created to plot the variance. This is described on line 109-119. If all variables were correlated with PC1, then all the variables would all be correlated with each other. All PC are anticorrelated with each other, which is why PC1 and PC2 are perpendicular to each other. Such is also the case between PC1 and PC3, PC2 and PC3, etc. As plotted in figure 4a) SW_CRE is near perfectly positively correlated with PC2, hence it has to be anticorrelated with every other PC. Which is why the vector is perpendicular to the PC1 axis. This is also why in figure 4c) the length of the vector is so short, as it nether correlates with PC3 or PC1. So yes, SW_CRE is correlated with PC2, and also Cloudy is correlated with PC2. The vector is just pointing in the opposite direction, which means the two variables are negatively correlated with each other. This is also what figure 4b) suggest providing negative correlation between the two variables.

We have made the following changes:

In the second paragraph in section 2.2

1) **We have replaced the following sentence: '***Each following PC in turn has the highest variance possible assuming that it is orthogonal to the previous PC, successively explaining less of the magnitude of cloudy sky RFari***' with '***All the variables relationship to each other can be to a lesser degree explained (magnitude) with each exceeding PC. In other words, it's not exclusively to RF ari$_{cloud}$***.'**
2) **This sentence is added: '***All PCs are anticorrelated with each other, which is why PC1 and PC2 are perpendicular to each other.***'**

In the result section 3.2:

3) **The following text is added: '***SW_CRE is near perfectly positively correlated with PC2 (Figure 4a), hence it must be anticorrelated with every other PCs. Therefore, the vector in Figure 4a is perpendicular to the PC1 axis. This is also why the length of the vector is so short in Figure 4c since it nether correlates with PC3 or PC1. SW_CRE is correlated with PC2 and Cloudy is correlated with PC2. The vector is pointing in the opposite direction between SW_CRE and Cloudy, which means the two variables are negatively correlated with each other. Figure 4b show also a negative correlation between the two variables.***'**

"Cloudy, FIX2 and FIX3 are plotted but don't affect the projection of the other variables." - I am not quite sure what this means for the interpretation of their position, is this just their correlation with PC1 and PC2? What does this shown.

Response: In the 4 caption we have added: **'**This requirement is made since cloudy, FIX2scat and FIX3abs already depends on the other variable and see their correlation.**'**

Also, Fig 4c does not appear to be referenced in the text at all. Is this intentional?

Response: It is added that global mean values from Table 1 is used in PCA analysis. Reference to Fig 4c is now included. SW_CRF, FIX2 and FIX3 have been changed to SW_CRE, FIX2scat and FIX3abs, respectively as suggested by the reviewer.

We have added the following text in the result section 3.2: '*A biplot with PC1 and PC3 (Figure 4c) can explain more about a variable than PC1 and PC2. For example, CL_ALT has a slightly stronger projection in the PC1 and PC3 biplot and suggest that there is an anticorrelation with FIX2scat. However, in the PC1 and PC2 they are positively correlated with each other. This suggest that there is partial correlation and Figure 4b shows there is a weak positive correlation between these two variables.***'**

We have added this sentence in section 3.2: *'Adding PC3 this number increases to 89.2%.'*

Third, how do these value fit in with the "error in the cloud radiative forcing" calculated using the method in Ghan (ACP, 2013)? That method would suggest a contribution to the RFari from aerosol above cloud of +0.40 Wm−2 . Higher values (although not as large as this) are also found in Gryspeerdt et al. (ACP 2020), which uses essentially the same method. Finally, a few more commas would be nice to improve readability and there are a few typos which could be caught in the next round (I have identified some of them below)

Response: From Table S2 in Gryspeerdt et al. (ACP 2020) the mean SWaricloud is +0.01 W m^{-2} of 8 models. The residual among a large set of simulating the ERFaer (direct and indirect aerosol effects) is weak indicating that SWaricloud is likely weak in all 17 models from AeroCom and CMIP5. We have had the following to the discussion section:

'The simulations used in this study only include the RF of the aerosol-radiation interaction. In a recent multi-model study, a decomposition of all aerosol effect (including aerosol-cloud interactions) provides weak RFari$_{cloud}$ for all models, of magnitude and multi-model mean similar to this study (Gryspeerdt et al., 2020). A separate single-model study however found it to be substantial (Ghan, 2013).'

Minor points L28 - Why are SSA and SW_CRF called out here, when they control PC2?

Response: These two factors have been shown in this study to play a major in calculation of the RFari$_{cloud}$ and thus emphasized in the abstract.

L68 - substantially L99 - constraint L102 - FIX2scat, FIX3abs? These acronyms are used in Tab. 1, but not elsewhere. Having the "scat" and "abs" suffixes is helpful for those less familiar with the experiments.

Response: the comment is taken into account

L121 - Not quite clear what is going on here. Why is the variable for which you are trying to explain the variance added to the list of variables in the PCA?
Response: See the additional text added to the manuscript described above.

L151 - "All sky RFari" or RFariallsky? I know these are the same, but it might help keep things clear.

Response: Simplifying Eq 1 as suggested by the Reviewer make the readability of this sentence easier and sentence is therefore made much shorter.

L161 - “present-day” instead of “current” would make this clearer that it is not referring to a current estimate.

Response: Comment is taken into account.

L184 - SW_CRE vs SW_CRF - Cloud radiative effect is referred to, but the acronym suggests radiative forcing.

Response: We agree to the comment and have change SW_CRF to SW_CRE through the manuscript.

L188 - Supplementary information seems to be missing

Response: We have changed the reference to Supplementary to Fig 4c.

L189 - FIX2 and FIX3 are hardly used. Is there more that could be said here?

Response: We have added the following: ‘FIX2scat and FIX3abs are strongly dependent on the host model clouds and their radiative effect and anticorrelated to cloud fraction and SW_CRE, respectively’

L194 - “PCA finds a weak dependence”

Response: Sentence corrected to include ‘*a*’ before ‘*weak’*

L207 - “However, when analyzing multi-model simulations, additional factors become important.”

Response: Sentence changed as suggested.

Ghan, S. J.: Technical Note: Estimating aerosol effects on cloud radiative forcing, Atmospheric Chemistry and Physics, 13(19), 9971-9974, 2013.

Gryspeerdt, E., Mülmenstädt, J., Gettelman, A., Malavelle, F. F., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M. and Zhang, K.: Surprising similarities in model and observational aerosol radiative forcing estimates, Atmos. Chem. Phys., 20(1), 613-623, 2020.

Cloudy sky contributions to the direct aerosol effect

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Abstract: The radiative forcing of the aerosol-radiation interaction can be decomposed into clear sky and cloudy sky portions. Two sets of multi-model simulations within AeroCom, combined with observational methods, and the time evolution of aerosol emissions over the industrial era show that the contribution from cloudy sky regions is likely weak. A mean of the simulations considered is 0.01 ± 0.1 $\mathrm{Wm^{-2}}$. Multivariate data analysis of results from AeroCom Phase II shows that many factors influence the strength of the cloudy sky contribution to the forcing of the aerosol-radiation interaction. Overall, single scattering albedo of anthropogenic aerosols and the interaction of aerosols with the shortwave cloud radiative effects are found to be important factors. A more dedicated focus on the contribution from the cloud free and cloud covered sky fraction respectively to the aerosol-radiation interaction will benefit the quantification of the radiative forcing and its uncertainty range.

1 Introduction

The radiative forcing (RF) of ~~atmospheric~~ anthropogenic aerosols in the atmosphere due to the aerosol-radiation interaction – RFari (earlier denoted as direct aerosol effect) was assessed as -0.35 [-0.85 to +0.15] Wm^{-2} in the Fifth Assessment Report by the Intergovernmental Panel on Climate Chang (IPCC) (AR5) (Boucher et al., 2013). The AR5 uncertainty range is even slightly wider than in the Fourth Assessment Report (Forster et al., 2007). Despite major progress in the understanding of atmospheric aerosol composition, and almost two decades of multi-aerosol type model simulations, little progress had been made in reducing the large uncertainty in this number, until recently where Bellouin et al. (2020) ~~Bellouin et al.~~ estimate a range of -0.45 to -0.05 Wm^{-2}. Bellouin et al. estimate RFari from normalized clear sky radiative effect by aerosol optical depth (AOD) and multiply this by an assessment of anthropogenic AOD. No direct simulations were used to calculate the RFari in regions of clouds.

One reason for the larger uncertainty range in AR5 compared to AR4 was enhanced uncertainty and magnitude of the RFari of black carbon (BC) (Boucher et al., 2013). Bond et al. (2013) indicated that emission of BC was too low in the inventories applied in climate models and therefore scaled the RFari from models to observed Aeronet absorption aerosol optical depth (AAOD) retrievals. More recently it has been suggested that AAOD data from Aeronet may have a sampling bias due to sites being located close to emission source regions (Wang et al., 2018), but uncertain in magnitude (Schutgens, 2019) and that most global aerosol models may have a bias towards too much BC in the middle and upper troposphere (Kipling et al., 2013; Samset et al., 2014; Wang et al., 2014). Both of these factors indicate a too strong BC RFari in Bond et al. (2013). However, the most recent estimates of emission from fossil and biofuel BC ~~emission inventory~~ (Hoesly et al., 2018) is much higher than used in previous global modelling (Lamarque et al., 2010). These new findings indicate that the BC RFari may be stronger than what was given in some of the multi-model global aerosol modelling exercises (Myhre et al., 2013a; Schulz et al., 2006), but likely weaker than estimated in Bond et al. (2013). The uncertainties in the RFari are also large for other aerosol species. For nitrate (Bian et al., 2017) the abundance is particularly uncertain, and for organic aerosols uncertainties are large both due to abundance (Tsigaridis et al., 2014) and the optical properties, particularly for brown carbon (Samset et al., 2018).

A further complication when estimating RFari is the atmospheric mix of scattering and absorbing aerosols. Since RFari is dependent on aerosol optical properties and the underlying albedo (Haywood and Shine, 1997), it is therefore also very dependent on where the aerosols are located relative to clouds (Takemura et al., 2002). Absorbing aerosols above clouds have a strong positive RFari (Chand et al., 2009; Keil and Haywood, 2003) but it becomes considerably weaker if the aerosols are located below clouds (Takemura et al., 2002). Scattering aerosols above or below clouds may enhance the reflection of solar radiation in conditions of thin clouds. The all sky RFari can be separated into contributions from clear and cloudy sky portions:

RFari$_{\textbf{all sky}}$ = ~~(1 - AC) *~~ **RFari**$_{\textbf{clear}}$ + ~~AC *~~ **RFari**$_{\textbf{cloud}}$ (1)

~~AC is the cloud fraction,~~ RFari$_{\text{clear}}$ and RFari$_{\text{cloud}}$ are the clear sky and cloudy sky portion to RFari$_{\text{allsky}}$ ~~RFari~~, respectively. RFari$_{\text{clear}}$ and RFari$_{\text{cloud}}$ can further be described as RFari$_{\text{clear}}$ = (1-AC) * RFari$_{\text{total-clear}}$ and RFari$_{\text{cloud}}$ = AC * RFari$_{\text{ctotal-cloud}}$, where AC is cloud fraction, RFari$_{\text{total-clear}}$ is simulations excluding all clouds and RFari$_{\text{total-cloud}}$ is the aerosol radiative effect assuming the whole grid is covered by clouds. All three variables vary as a function of longitude, latitude and time. The RFari is the initial perturbation to top of the atmosphere (TOA) radiative fluxes (the instantaneous RF which for aerosol is very similar to RF). Rapid adjustments from shortwave absorption by aerosols, mostly BC, may alter atmospheric temperatures, water vapour and clouds. The sum of RFari and rapid adjustments is denoted effective radiative forcing (Boucher et al., 2013; Myhre et al., 2013b; Sherwood et al., 2015). The rapid adjustment of absorbing aerosols can be strong and may counteract the RFari substantially (Smith et al., 2018). Since RFari includes no rapid adjustments, AC is constant in simulations of RFari$_{\text{clear}}$ and RFari$_{\text{cloud}}$ ~~Equation 1~~. Oikawa et al. (2013); Oikawa et al. (2018) provide estimates of all sky, clear sky and cloudy sky radiative effect of aerosols in different regions based on satellite retrievals of clouds and aerosols. These studies further describe large differences resulting from whether aerosols are below or above clouds. Lacagnina et al. (2017); Zhang et al. (2016) found large regional variation in the radiative effect of aerosols above clouds. Note that the above-mentioned studies investigate the ~~current~~present, total aerosol abundance which consist of anthropogenic and natural aerosols, whereas in terms of RFari only the anthropogenic aerosols are considered. Anthropogenic aerosols are changes to the atmospheric composition since pre-industrial time. Matus et al. (2019) combined satellite derived aerosol radiative effect with model simulation of anthropogenic aerosol, to make an estimate of RFari.

The aim of the present study is to provide insight into factors determining ~~the contribution from cloudy sky regions~~ RFari$_{\text{cloud}}$ and thus the contribution from cloudy sky regions to the RFari ~~(second term on the right-hand side of equation 1)~~ from combining global models and observational based approaches. We present estimates of this quantity from several global studies and we use multivariate data analysis to provide insight on the core factors causing the diversity among models.

2 Methods

2.1 Global estimates of cloudy sky contribution to RFari

The models, experiments and RFari from AeroCom Phase I and II are documented in detail by Schulz et al. (2006) and Myhre et al. (2013a), respectively. We also analyze the historical evolution of RFari due to anthropogenic aerosols using output from a series of OsloCTM3 simulations (Lund et al., 2018) with emissions from the Community Emission Data System (CEDS) (Hoesly et al., 2018) inventory from year 1750 to 2014. The OsloCTM3 is a global 3-dimensional chemistry-transport model driven by 3-hourly meteorological forecast data.

The analysis is further supplemented by variables from Equation 1 extracted from Myhre (2009), who presented results from OsloCTM2 and an observational based method to explain the difference in all sky RFari between observational based and global aerosol model approaches. The model simulations were made to investigate several assumptions on aerosol optical properties and impacts of assumptions related to missing data and change in industrial era aerosol concentration for the observational method. The Max Planck Aerosol Climatology (MACv2) method combines aerosol column optical properties for fine-mode and coarse-mode sizes (of an AeroCom phase1 model median regionally adjusted by AERONET/MAN monthly statistics) with MODIS surface albedo data, ISCCP cloud properties and vertical scaling by size-mode from 20 years of ECHAM-HAM aerosol simulations. The anthropogenic properties is defined as a fraction of the fine-mode, where the fine-mode AOD scaling factor prescribed from AeroCom phase1 simulations. ~~The Max Planck Aerosol Climatology version 2 (MACv2) method combines aerosol optical properties from Aeronet, surface albedo and clouds from ISCCP, vertical profiles of absorbing aerosols from the ECHAM-HAM model, with multi-model mean data on anthropogenic fraction from AeroCom Phase I~~ (Kinne, 2019a; Kinne, 2019b).

The cloudy sky contribution to all sky RFari is calculated from daily or monthly diagnostics of allsky RFari, RFari$_{clear}$ and AC. RF is taken at the top of the atmosphere and all estimates are from pre-industrial to present.

2.2 Multivariate data analysis

Multivariate data analysis in this study is based on results from a subset of the models participating in AeroCom Phase II (CAM5, GOCART, HadGEM2, IMPACT, INCA, ECHAM-HAM, OsloCTM2 and SPRINTARS). These eight models participated in the AeroCom Phase II experiment (Myhre et al., 2013a) with no constrain on aerosol processes and in addition participated in the host model AeroCom exercise with fixed aerosol optical properties (Stier et al., 2013). From the latter FIX2scat and FIX3abs experiments can be used to retrieve the contributions from cloudy sky to RFari in two highly idealized aerosol radiative properties experiments. FIX2scat is a purely scattering aerosol case and FIX3abs is an absorbing aerosol case. The origin of the different variables derived from AeroCom phase II model simulations (Myhre et al., 2013a; Samset et al., 2013) can be found Table 1.

The global mean data in Table 1 is analyzed using principal component analysis (PCA). Here, the variables that may influence the ~~cloudy sky RFari contribution to all sky~~ RFari$_{cloud}$ are orthogonally transformed into linearly uncorrelated variables named principal components (PCs). The transformation is defined so that the first principal component (PC1) accounts for most of variance exhibited by the underlying variables. PCA is a dimension reduction technique, allowing for the visualization of all the variance between the variables in a two-dimensional plot (biplot). All PCs are anticorrelated with each other, which is why PC1 and PC2 are perpendicular to each other. How the variables relate to each other can to some extent be explained (magnitude) with each exceeding PC. In other words, each PC does not exclusively show its relation to RFari$_{cloud}$.

Data is normalized prior to PCA to ensure comparison of variance between variables. PCA results are usually plotted in a biplot, where only PC1 and PC2 (the second PC) are plotted on the x- and y-axis, respectively, since the two PCs explain most of the variance. In the biplot variables are projected as vectors along PC1 and PC2. The combined length and direction of the vector indicates the correlation the variable has with PC1 and PC2. Values range from -1 to 1 indicating negative to positive correlation with the PC. A value of 0 indicates no correlation with the PC. Since the projected vectors are directional it is possible to have high correlation with PC1 (values ~ -1 or ~ 1), and poor correlation with PC2 (value ~ 0), or *visa-versa*.

Variables that point in the same direction are positively correlated with each other. Variables that point in the opposite direction of each other are negatively correlated. Variables that are perpendicular to each other are not correlated with each other. The missing data (see Table 1) for two of the models (LMDZ-INCA and ECHAM-HAM) are filled-in using regularized iterative PCA. This technique estimates the missing values, based on the correlation between the variables and the principal components (Josse and Husson, 2012).

The ~~contribution of cloudy sky to~~ RFari$_{\text{cloud}}$ ~~(“Cloudy”)~~ (second term in Equation 1) is added as a supplementary variable in the PCA. This is to ensure that this dependent variable, the cloudy sky contribution, does not influence the projected correlations the independent variables have on each other. The same approach is applied to FIX2scat and FIX3abs, as these variables are not independent, as they are composed of the many of the variables used in the analysis. In addition, linear regression correlation coefficients are calculated between all the variables to assess the individual relationships.

3 Results

3.1 Estimates of cloudy sky contributions to RFari

Figure 1 shows the all sky RFari due to anthropogenic aerosols separated into clear sky (first term on right-hand side in equation 1) and cloudy sky (second term on right-hand side in equation 1) portion from AeroCom Phase II models (Myhre et al., 2013a). The uncertainty ranges given in the figure are one standard deviation among the global aerosol models in AeroCom Phase II. The figure shows two main results, that the cloudy sky RFari is weak and that the uncertainties in the contributions from cloudy sky and clear sky are substantial with the latter somewhat larger in magnitude.

Figure 2 shows an example from OsloCTM3 (Lund et al., 2018) of the spatial distribution ~~of various~~ terms ~~given~~ involved in equation 1. In the lower row of Figure 2 the annual mean AC, RFari$_{\text{clear-total}}$ and RFari$_{\text{cloud-total}}$ are shown, with strong negative RFari$_{\text{clear-total-}}$ over most land areas except over regions of high surface albedo such as deserts. The RFari$_{\text{cloud-total}}$ is particularly positive over regions of biomass burning aerosols overlying low level stratocumulus, but also over parts of high aerosol abundance over China. The second row shows the two terms on the right hand in equation 1, namely the contribution from the cloud free and cloudy regions to the all sky RFari. The contribution from the clear sky regions (RFari$_{\text{clear}}$, first term on the right-hand side of equation 1) is much weaker than RFari$_{\text{clear-total}}$ itself since cloud fraction is high in many of the regions of anthropogenic aerosols. While the influence from cloud fraction on ~~the cloudy sky contribution to all sky~~ RFari$_{\text{cloud}}$ relative to RFari$_{\text{cloud-total}}$ is weak over biomass burning regions, it weakens relative to the negative values in RFari$_{\text{cloud-total}}$ over many areas in the northern hemisphere. In the top row the RFari all sky is the sum of the contributions from clear and cloudy regions where their importance for the RFari varies regionally.

Figure 3a-d shows estimates of the contribution from cloud sky to RFari from several studies: two are multi-model studies, one combines model and observational based methods and one study investigates the time evolution using one model. The two multi-model AeroCom studies (Myhre et al., 2013a; Schulz et al., 2006) show that the sign varies among the global aerosol models and that two versions from one model changes sign between the two AeroCom phases (two versions of ECHAM-HAM, UIOCTM versus OsloCTM2, and UMI versus IMPACT). The two model versions INCA and LSCE have positive values in both AeroCom phases. SPRINTARS has the strongest positive (and overall strongest magnitude) ~~of cloudy sky~~ RFari$_{cloud}$ ~~contribution to all sky RFari~~ in both AeroCom phases. About half of the models shown in Figure 3a and 3b have provided sufficient diagnostics to extract estimates from both AeroCom phases. The AeroCom PhaseII results will be further discussed in section 3.2.

In Myhre (2009) several experiments were performed to explain that differences in RFari between observational based methods and global aerosol models arise from a relatively larger change in absorbing aerosols over the industrial era than in the current abundance of the absorbing aerosols. Whereas an observational method uses aerosol optical properties from measurements of the present time of the combined natural and anthropogenic aerosols, the models simulate a relatively larger change in the abundance of anthropogenic absorbing aerosols than assuming no change in the industrial era aerosol optical properties. Figure 3c shows the contribution of the cloudy sky to RFari from several of these experiments. The two experiments MODIS(SCREEN) and MODIS use satellite retrievals of aerosol optical depth (AOD), ~~current~~ present aerosol optical properties retrieved (single scattering albedo and asymmetry factor) from AERONET, and a model estimate of the anthropogenic AOD. The difference between these experiments is that the MODIS experiment uses model information over regions of missing AOD from the satellite retrievals, while these regions are ignored in MODIS(SCREEN). ~~The contribution of cloudy sky to all sky~~ RFari$_{cloud}$ is similar in these two experiments. On the other hand, in the experiment MODIS(Model), changes in the aerosol optical properties from pre-industrial to present causes the change in sign in the cloud sky contribution to RFari compared to MODIS and MODIS(SCREEN). The MODIS(Model) has very similar RFari, as well as ~~cloudy sky contribution to all sky~~ RFari$_{cloud}$, to the standard global aerosol model simulations (MODEL INT and MODEL EXT). The two latter model simulations differ on whether internal or external mixing of BC is taken into account~~ or not~~, respectively. The MACv2 ~~cloudy sky contribution to~~ RFari$_{cloud}$ is -0.13 Wm^{-2}. This estimate does not consider change in the aerosol optical properties over the industrial era and can thus be compared to MODIS(SCREEN) and MODIS experiments described above.

The time evolution of the contribution of cloudy sky to RFari in OsloCTM3 is shown in Figure 3d where all variations are caused by changes in the anthropogenic aerosol composition and abundance since all other factors are kept constant. Values are negative in the period 1960 to 1990 due to a strong increase in SO$_2$ emissions and thereby a domination of scattering aerosols and radiative impacts even in cloudy skies. In the period after 1990 the regional SO$_2$ emissions have changed strongly, but with a small reduction in the global emissions. Emissions of BC have on the other hand increased substantially making anthropogenic aerosols more absorbing in the OsloCTM3 causing a relatively stronger positive contribution from the cloudy sky to RFari.

3.2 Multivariate data analysis of cloudy sky contribution to the all sky RFari

Table 1 lists the AeroCom Phase II models and variables included in the multivariate data analysis for investigating ~~contributions to cloudy sky~~ RFari$_{\text{cloud}}$ denoted as “Cloudy” in the table (the second term on the right-hand side of equation 1). The results of the multivariate data analysis are plotted in a biplot and a correlogram Figure 4. The principal component analysis (PCA) found that 68.2% of the total variance is explained by the first and second principal component (PC1 and PC2), see Figure 4a. Adding PC3 this number increases to 89.2% (see Figure 4c). The analysis shows that several factors are important for the contribution of cloudy sky to RFari (“Cloudy”). Among all variables total short-wave cloud radiative effect (SW_CRE~~F~~) is the most important. SW_CRE is near perfectly positively correlated with PC2 (Figure 4a), hence it must be anticorrelated with every other PCs. Therefore, the vector in Figure 4a is perpendicular to the PC1 axis. This is also why the length of the vector is so short in Figure 4c since it neither correlates with PC3 nor PC1. SW_CRE is correlated with PC2 and Cloudy is correlated with PC2. The vector is pointing in the opposite direction between SW_CRE and Cloudy, which means the two variables are negatively correlated with each other. Figure 4b also shows a negative correlation between the two variables. A biplot with PC1 and PC3 (Figure 4c) can explain more about a variable than PC1 and PC2. For example, CL_ALT has a slightly stronger projection in the PC1 and PC3 biplot and suggest that there is an anticorrelation with FIX2scat. However, in the PC1 and PC2 they are positively correlated with each other. This suggest that there is partial correlation and Figure 4b shows there is a weak positive correlation between these two variables.

Single scattering albedo (SSA) being a crucial variable for the anthropogenic aerosols may potentially be an important factor (a higher SSA is expected to give a more negative cloudy sky forcing). However, independent correlations plotted in the correlogram (Figure 4b) suggests that the ~~cloudy sky~~ contribution to RFari$_{\text{cloud}}$ and SSA is weak (r = 0.17). In depth analysis of the linear correlation between cloudy sky to RFari and SSA suggests that the linear relationship exist only at higher PCs (see ~~supplementary~~ Figure 4c~~S1~~).

The contribution of cloudy sky to RFari shows a closer dependence on similar quantities for the idealized experiment FIX3abs than FIX2scat, where FIX2scat has purely scattering aerosols. Both FIX2scat and FIX3abs are dependent on host model properties such as ~~clouds,~~ surface albedo and radiative transfer schemes (Stier et al., 2013). They are further strongly dependent on the host model clouds and their radiative effect and anticorrelated to cloud fraction (CLD_FR) and SW_CRE, respectively. PCA finds negative correlation between ~~cloudy sky contribution to~~ RFari$_{\text{cloud}}$ and total short-wave cloud radiative flux (SW_CRE~~F~~), also supported by the linear regression. One example here is the GOCART model with the weakest SW_CRE~~F~~ and most negative cloudy sky contribution to RFari of the models included in the multivariate data analysis. At the same time the PCA finds a small dependence between the cloudy sky contribution to RFari and cloud fraction (CLD_FR) or cloud altitude (CL_ALT). The negative correlation between ~~cloudy sky contribution to~~ RFari$_{\text{cloud}}$ and SW_CRE~~F~~ can be explained by reflective clouds enhancing the underlying albedo and thus making the radiative forcing more positive with an increase in absorbing aerosols in the cloudy sky portion.

Overall FIX3abs (where models have a fixed SSA) and SW_CRE~~F~~ seem to be the main explanatory variables for the variance in the cloudy sky contribution to RFari. It is however worth noting that the correlation for cloudy sky contribution to RFari with the variables is not particular strong in any direction (indicated by the short arrow). This suggests that some of the variance may be explained along the third or fourth principal component etc.

4 Discussion and conclusions

The multivariate data analysis shows that host model characteristics (especially SW_CRE~~F~~) are important for the modelled cloudy sky contribution to RFari (RFari$_{cloud}$), but ~~also~~ further indicates that many other factors are important. Furthermore, several other studies presented here show that aerosol properties (in particular SSA) are important for this quantity. Locally and especially in regions with aerosols above clouds, as well as in single model studies the SSA is crucial for RFari$_{cloud}$~~cloudy sky contribution to RFari~~. ~~However, analyzing multi-model simulations then additional factors are becoming important.~~ The two AeroCom phases give RFari$_{cloud}$~~cloudy sky contribution to RFari~~ estimates of 0.0 ± 0.10 Wm^{-2} and 0.04 ± 0.10 Wm^{-2} and the mean of two observational based methods is -0.02 (range from -0.13 to 0.09 Wm^{-2}). Combining the numbers from these three studies, we find a RFari$_{cloud}$ of 0.01 ± 0.1 Wm^{-2} ~~for the cloudy sky contribution to all sky RFari~~. The new emission inventory from CEDS has a strong increase in BC emissions leading to an increase in RFari$_{cloud}$~~cloudy sky contribution to RFari~~ of 0.05 Wm^{-2} from 2000 to 2014 in OsloCTM3. Using OsloCTM3 simulations to investigate the importance of ~~using~~ diagnostics for every radiation time step (3 hourly) shows differences up to 0.01 W m^{-2} relative to daily mean data and up to 0.04 W m^{-2} for monthly data, but this may be model dependent (Haywood and Shine, 1997). The simulations used in this study only include the RF of the aerosol-radiation interaction. In a recent multi-model study, a decomposition of all aerosol effect (including aerosol-cloud interactions) provides weak RFari$_{cloud}$ for all models, of magnitude and multi-model mean similar to this study (Gryspeerdt et al., 2020). A separate single-model study however found RFari$_{cloud}$ to be substantial (Ghan, 2013). Determining the quantity of black carbon from instrumentation such as the SP2 has provided a new set of consistent data for assessing the performance of aerosol models (e.g. Kipling et al. (2013); Wang et al. (2014)). Knowledge of BC mass is fundamentally insufficient for determining the ambient aerosol single scattering albedo owing to additional complexities such as the degree of internal and external mixing. In the past, the aerosol modelling community has relied either on indirect remotely sensed measurements from AERONET (e.g. Chin et al. (2009)) or on imperfect in-situ measurements of aerosol scattering from nephelometers (e.g. Anderson et al. (2003)) and absorption from filter-based systems (e.g. Bond et al. (1999)). Both of these systems are relatively imprecise corrections to account for scattering and absorption artifacts (e.g. Davies et al. (2019); Massoli et al. (2009)). The single scattering albedos can be determined much more accurately using combinations of cavity ring-down measurements of extinction (e.g. Lack et al. (2006)) and photoacoustic measurements of aerosol absorption (e.g. Baynard et al. (2006)). These instruments are becoming more routine on aircraft equipped for making atmospheric measurements that can make highly accurate assessments of the aerosol single scattering albedo at above-cloud altitudes (e.g.

Davies et al. (2019); Langridge et al. (2011)). These measurements will provide an invaluable additional source of data for model evaluation. All the global models that supplied simulations –for this study treat the major anthropogenic aerosol components sulphate, organic aerosols, and black carbon, some also treat nitrate, but none include anthropogenic dust aerosols which have highly uncertain radiative effects.

Koffi et al. (2016); Koffi et al. (2012) show that global aerosol models generally tend to have an overabundance of aerosols at higher altitude compared to satellite retrievals from CALIPSO and Samset et al. (2014) show that the AeroCom models overestimate BC at mid and high tropospheric altitudes compared to aircraft measurements. Too much BC above the clouds would overestimate RFari$_{cloud}$~~the contribution of the cloudy sky to RFari~~. On the other hand, Peers et al. (2016) show that over the biomass burning region in south Africa most of the AeroCom models underestimate the AAOD over the stratocumulus layer, which would underestimate RFari$_{cloud}$~~the contribution of cloudy sky to RFari~~.

In future studies of RFari, particular attention should be put on how global aerosol models simulate the location of aerosols in relation to clouds and how aerosol optical properties change with altitude in regions with high cloud cover compared to measurements in order to further constrain the spread in the modelled cloudy sky contribution. Nowhere is this high sensitivity more clearly demonstrated than over the SE Atlantic where biomass burning aerosols over-lie (and sometimes interact with) relatively bright stratocumulus clouds (e.g. Zuidema et al. (2016)). In addition to further analysis of aerosol RF in cloudy sky regions, more emphasis~~ze~~ should be devoted to quantifying the RFari in cloud free regions and its trend (Paulot et al., 2018), where the magnitude of forcing is larger than in cloudy regions.

Acknowledgments, Samples, and Data

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References

Anderson, T. L., Masonis, S. J., Covert, D. S., Ahlquist, N. C., Howell, S. G., Clarke, A. D. and McNaughton, C. S.: Variability of aerosol optical properties derived from in situ aircraft measurements during ACE-Asia, Journal of Geophysical Research: Atmospheres, 108(D23), 2003.

Baynard, T., Garland, R. M., Ravishankara, A. R., Tolbert, M. A. and Lovejoy, E. R.: Key factors influencing the relative humidity dependence of aerosol light scattering, Geophysical Research Letters, 33(6), 2006.

Bellouin, N., Quaas, J., Gryspeerdt, E., Kinne, S., Stier, P., Watson-Parris, D., Boucher, O., Carslaw, K. S., Christensen, M., Daniau, A.-L., Dufresne, J.-L., Feingold, G., Fiedler, S., Forster, P., Gettelman, A., Haywood, J. M., Lohmann, U., Malavelle, F., Mauritsen, T., McCoy, D. T., Myhre, G., Mülmenstädt, J., Neubauer, D., Possner, A., Rugenstein, M., Sato, Y., Schulz, M., Schwartz, S. E., Sourdeval, O., Storelvmo, T., Toll, V., Winker, D. and Stevens, B.: Bounding Global Aerosol Radiative Forcing of Climate Change, Reviews of Geophysics, 58(1), e2019RG000660, 2020.

Bian, H., Chin, M., Hauglustaine, D. A., Schulz, M., Myhre, G., Bauer, S. E., Lund, M. T., Karydis, V. A., Kucsera, T. L., Pan, X., Pozzer, A., Skeie, R. B., Steenrod, S. D., Sudo, K., Tsigaridis, K., Tsimpidi, A. P. and Tsyro, S. G.: Investigation of global particulate nitrate from the AeroCom phase III experiment, Atmos. Chem. Phys., 17, 12911-12940, 2017.

Bond, T. C., Anderson, T. L. and Campbell, D.: Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols, Aerosol Science and Technology, 30(6), 582-600, 1999.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Karcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research-Atmospheres, 118(11), 5380-5552, 2013.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B. and Zhang, X.-Y., Clouds and Aerosols. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen et al. (Editors), Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 571-657, 2013.

Chand, D., Wood, R., Anderson, T. L., Satheesh, S. K. and Charlson, R. J.: Satellite-derived direct radiative effect of aerosols dependent on cloud cover, Nature Geoscience, 2(3), 181-184, 2009.

Chin, M., Diehl, T., Dubovik, O., Eck, T. F., Holben, B. N., Sinyuk, A. and Streets, D. G.: Light absorption by pollution, dust, and biomass burning aerosols: a global model study and evaluation with AERONET measurements, Annales Geophysicae, 27(9), 3439-3464, 2009.

Davies, N. W., Fox, C., Szpek, K., Cotterell, M. I., Taylor, J. W., Allan, J. D., Williams, P. I., Trembath, J., Haywood, J. M. and Langridge, J. M.: Evaluating biases in filter-based aerosol absorption measurements using photoacoustic spectroscopy, Atmos. Meas. Tech., 12(6), 3417-3434, 2019.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M. and Van Dorland, R., Changes in Atmospheric Constituents and in Radiative Forcing. In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Ghan, S. J.: Technical Note: Estimating aerosol effects on cloud radiative forcing, Atmospheric Chemistry and Physics, 13(19), 9971-9974, 2013.

Gryspeerdt, E., Mülmenstädt, J., Gettelman, A., Malavelle, F. F., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., Wang, M. and Zhang, K.: Surprising similarities in model and observational aerosol radiative forcing estimates, Atmos. Chem. Phys., 20(1), 613-623, 2020.

Haywood, J. M. and Shine, K. P.: Multi-spectral calculations of the direct radiative forcing of tropospheric sulphate and soot aerosols using a column model, Quarterly Journal of the Royal Meteorological Society, 123(543), 1907-1930, 1997.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J. I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R. and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11(1), 369-408, 2018.

Josse, J. and Husson, F.: Handling missing values in exloratory multivariate data analysis methods, Journal de la Société Fraçaise de Statistique, 153, 79–99, 2012.

Keil, A. and Haywood, J. M.: Solar radiative forcing by biomass burning aerosol particles during SAFARI 2000: A case study based on measured aerosol and cloud properties, Journal of Geophysical Research-Atmospheres, 108(D13), 8467, 2003.

Kinne, S.: Aerosol radiative effects with MACv2, Atmos. Chem. Phys., 19(16), 10919-10959, 2019a.

Kinne, S.: The MACv2 aerosol climatology, Tellus B: Chemical and Physical Meteorology, 71(1), 1-21, 2019b.

Kipling, Z., Stier, P., Schwarz, J. P., Perring, A. E., Spackman, J. R., Mann, G. W., Johnson, C. E. and Telford, P. J.: Constraints on aerosol processes in climate models from vertically-resolved aircraft observations of black carbon, Atmos. Chem. Phys., 13(12), 5969-5986, 2013.

Koffi, B., Schulz, M., Bréon, F.-M., Dentener, F., Steensen, B. M., Griesfeller, J., Winker, D., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T., Bian, H., Chin, M., Diehl, T., Easter, R., Ghan, S., Hauglustaine, D. A., Iversen, T., Kirkevåg, A., Liu, X., Lohmann, U., Myhre, G., Rasch, P., Seland, Ø., Skeie, R. B., Steenrod, S. D., Stier, P., Tackett, J., Takemura, T., Tsigaridis, K., Vuolo, M. R., Yoon, J. and Zhang, K.: Evaluation of the aerosol vertical distribution in global aerosol models through comparison against CALIOP measurements: AeroCom phase II results, Journal of Geophysical Research: Atmospheres, 121(12), 7254-7283, 2016.

Koffi, B., Schulz, M., Breon, F. M., Griesfeller, J., Winker, D., Balkanski, Y., Bauer, S., Berntsen, T., Chin, M. A., Collins, W. D., Dentener, F., Diehl, T., Easter, R., Ghan, S., Ginoux, P., Gong, S. L., Horowitz, L. W., Iversen, T., Kirkevag, A., Koch, D., Krol, M., Myhre, G., Stier, P. and Takemura, T.: Application of the CALIOP layer product to evaluate the vertical distribution of aerosols estimated by global models: AeroCom phase I results, Journal of Geophysical Research-Atmospheres, 117, D10201, doi:10.1029/2011jd016858, 2012.

Lacagnina, C., Hasekamp, O. P. and Torres, O.: Direct radiative effect of aerosols based on PARASOL and OMI satellite observations, Journal of Geophysical Research: Atmospheres, 122(4), 2366-2388, 2017.

Lack, D. A., Lovejoy, E. R., Baynard, T., Pettersson, A. and Ravishankara, A. R.: Aerosol Absorption Measurement using Photoacoustic Spectroscopy: Sensitivity, Calibration, and Uncertainty Developments, Aerosol Science and Technology, 40(9), 697-708, 2006.

Lamarque, J., Bond, T., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M., Shindell, D., Smith, S., Stehfest, E., Van Aardenne, J., Cooper, O., Kainuma, M., Mahowald, N., McConnell, J., Naik, V., Riahi, K. and van Vuuren, D.: Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmospheric Chemistry and Physics, 7017-7039, 2010.

Langridge, J. M., Richardson, M. S., Lack, D., Law, D. and Murphy, D. M.: Aircraft Instrument for Comprehensive Characterization of Aerosol Optical Properties, Part I: Wavelength-Dependent Optical Extinction and Its Relative Humidity Dependence Measured Using Cavity Ringdown Spectroscopy, Aerosol Science and Technology, 45(11), 1305-1318, 2011.

Lund, M. T., Myhre, G., Haslerud, A. S., Skeie, R. B., Griesfeller, J., Platt, S. M., Kumar, R., Myhre, C. L. and Schulz, M.: Concentrations and radiative forcing of anthropogenic aerosols from 1750 to 2014 simulated with the Oslo CTM3 and CEDS emission inventory, Geosci. Model Dev., 11(12), 4909-4931, 2018.

Massoli, P., Murphy, D. M., Lack, D. A., Baynard, T., Brock, C. A. and Lovejoy, E. R.: Uncertainty in Light Scattering Measurements by TSI Nephelometer: Results from Laboratory Studies and Implications for Ambient Measurements, Aerosol Science and Technology, 43(11), 1064-1074, 2009.

Matus, A. V., L'Ecuyer, T. S. and Henderson, D. S.: New Estimates of Aerosol Direct Radiative Effects and Forcing From A-Train Satellite Observations, Geophysical Research Letters, 46(14), 8338-8346, 2019.

Myhre, G.: Consistency between satellite-derived and modeled estimates of the direct aerosol effect, Science, 325(5937), 187-190, 2009.

Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevag, A., Lamarque, J. F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, O., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J. H., Zhang, K., Zhang, H. and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, Atmospheric Chemistry and Physics, 13(4), 1853-1877, 2013a.

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T. and Zhang, H., Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor,

S. K. Allen et al. (Editors), Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 659-740, 2013b.

Oikawa, E., Nakajima, T., Inoue, T. and Winker, D.: A study of the shortwave direct aerosol forcing using ESSP/CALIPSO observation and GCM simulation, Journal of Geophysical Research: Atmospheres, 118(9), 3687-3708, 2013.

Oikawa, E., Nakajima, T. and Winker, D.: An Evaluation of the Shortwave Direct Aerosol Radiative Forcing Using CALIOP and MODIS Observations, Journal of Geophysical Research: Atmospheres, 123(2), 1211-1233, 2018.

Paulot, F., Paynter, D., Ginoux, P., Naik, V. and Horowitz, L. W.: Changes in the aerosol direct radiative forcing from 2001 to 2015: observational constraints and regional mechanisms, Atmos. Chem. Phys., 18(17), 13265-13281, 2018.

Peers, F., Bellouin, N., Waquet, F., Ducos, F., Goloub, P., Mollard, J., Myhre, G., Skeie, R. B., Takemura, T., Tanré, D., Thieuleux, F. and Zhang, K.: Comparison of aerosol optical properties above clouds between POLDER and AeroCom models over the South East Atlantic Ocean during the fire season, Geophysical Research Letters, 43(8), 3991-4000, 2016.

Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S. M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J. F., Lin, G., Liu, X., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K. and Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, Atmos. Chem. Phys., 14(22), 12465-12477, 2014.

Samset, B. H., Myhre, G., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kinne, S., Kirkevag, A., Lamarque, J. F., Lin, G., Liu, X., Penner, J. E., Seland, O., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K. and Zhang, K.: Black carbon vertical profiles strongly affect its radiative forcing uncertainty, Atmospheric Chemistry and Physics, 13(5), 2423-2434, 2013.

Samset, B. H., Stjern, C. W., Andrews, E., Kahn, R. A., Myhre, G., Schulz, M. and Schuster, G. L.: Aerosol Absorption: Progress Towards Global and Regional Constraints, Current Climate Change Reports, 4(2), 65-83, 2018.

Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Dentener, F., Guibert, S., Isaksen, I. S. A., Iversen, T., Koch, D., Kirkevag, A., Liu, X., Montanaro, V., Myhre, G., Penner, J. E., Pitari, G., Reddy, S., Seland, O., Stier, P. and Takemura, T.: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmospheric Chemistry and Physics, 6, 5225-5246, 2006.

Schutgens, N. A. J.: Site representativity of AERONET and GAW remotely sensed AOT and AAOT observations, Atmos. Chem. Phys. Discuss., 2019, 1-29, 2019.

Sherwood, S. C., Bony, S., Boucher, O., Bretherton, C., Forster, P. M., Gregory, J. M. and Stevens, B.: Adjustments in the Forcing-Feedback Framework for Understanding Climate Change, Bulletin of the American Meteorological Society, 96(2), 217-228, 2015.

Smith, C. J., Kramer, R. J., Myhre, G., Forster, P. M., Soden, B. J., Andrews, T., Boucher, O., Faluvegi, G., Fläschner, D., Hodnebrog, Ø., Kasoar, M., Kharin, V., Kirkevåg, A., Lamarque, J.-F., Mülmenstädt, J., Olivié, D., Richardson, T., Samset, B. H., Shindell, D., Stier, P., Takemura, T., Voulgarakis, A. and Watson-Parris, D.: Understanding Rapid Adjustments to Diverse Forcing Agents, Geophysical Research Letters, 45(21), 12,023-12,031, 2018.

Stier, P., Schutgens, N. A. J., Bellouin, N., Bian, H., Boucher, O., Chin, M., Ghan, S., Huneeus, N., Kinne, S., Lin, G., Ma, X., Myhre, G., Penner, J. E., Randles, C. A., Samset, B., Schulz, M., Takemura, T., Yu, F., Yu, H. and Zhou, C.: Host model uncertainties in aerosol radiative forcing estimates: results from the AeroCom Prescribed intercomparison study, Atmospheric Chemistry and Physics, 13(6), 3245-3270, 2013.

Takemura, T., Nakajima, T., Dubovik, O., Holben, B. N. and Kinne, S.: Single-scattering albedo and radiative forcing of various aerosol species with a global three-dimensional model, Journal of Climate, 15(4), 333-352, 2002.

Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S. E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S., Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen, T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H., Lin, G., Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J. J., Müller, J. F., Myhre, G., Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M., Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou, T., Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q. and Zhang, X.: The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem. Phys., 14(19), 10845-10895, 2014.

Wang, Q. Q., Jacob, D. J., Spackman, J. R., Perring, A. E., Schwarz, J. P., Moteki, N., Marais, E. A., Ge, C., Wang, J. and Barrett, S. R. H.: Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific, Journal of Geophysical Research-Atmospheres, 119(1), 195-206, 2014.

Wang, R., Andrews, E., Balkanski, Y., Boucher, O., Myhre, G., Samset, B. H., Schulz, M., Schuster, G. L., Valari, M. and Tao, S.: Spatial Representativeness Error in the Ground-Level Observation Networks for Black Carbon Radiation Absorption, Geophysical Research Letters, 45(4), 2106-2114, 2018.

Zhang, Z., Meyer, K., Yu, H., Platnick, S., Colarco, P., Liu, Z. and Oreopoulos, L.: Shortwave direct radiative effects of above-cloud aerosols over global oceans derived from 8 years of CALIOP and MODIS observations, Atmos. Chem. Phys., 16(5), 2877-2900, 2016.

Zuidema, P., Redemann, J., Haywood, J., Wood, R., Piketh, S., Hipondoka, M. and Formenti, P.: Smoke and Clouds above the Southeast Atlantic: Upcoming Field Campaigns Probe Absorbing Aerosol’s Impact on Climate, Bulletin of the American Meteorological Society, 97(7), 1131-1135, 2016.

Tables

Table 1. Diagnostics from AeroCom Phase 2 (Myhre et al., 2013a; Samset et al., 2013; Stier et al., 2013) used in multivariate data analysis to investigate factors influencing the contribution of cloudy sky to RFari. The variable "Cloudy" is the contribution of cloudy sky to RFari (second term on the right-hand side of equation 1). FIX2scat and FIX3abs are the contributions from cloudy sky to RFari in two highly idealized aerosol radiative properties experiments in Stier et al. (2013), where FIX2scat is a purely scattering aerosol case and FIX3abs is an absorbing aerosol case. The other variables, from AeroCom phase II model simulations, are total short-wave cloud radiative effect (SW_CRE~~F~~), cloud fraction (CLD_FR), weighed cloud height (CL_ALT), weighted anthropogenic aerosol height (AER_ALT), single scattering albedo (SSA) of anthropogenic aerosols and fraction of anthropogenic BC mass above 5 km (BC_mass_5km). The variables Cloudy, FIX2scat, FIX3abs and SW_CRE~~F~~ are given in Wm^{-2}, CLD_FR and BC mass>5km in percent, with SSA unitless. AER_ALT and CL_ALT are given in hPa where the pressure levels are weighted by aerosol extinction and cloud fractions, respectively.

		Host model dependences					Aerosol properties		
Models	Cloudy	FIX2scat	FIX3abs	SW_ CRE~~F~~	CLD FR	CL ALT	AER ALT	SSA	BC mass >5km
	W m-2	W m-2	W m-2	W m-2	%	hPa	hPa	1	%
CAM5	0.121	-1.8	1.8	-48.4	64	592	908	0.901	18.1
GOCART	-0.114	-1.6	1.2	-21.8	58	520	867	0.937	27.1
HadGEM2	0.0554	-1.1	1.5	-53.1	55	638	921	0.947	33.6
IMPACT	0.114	-1.5	2.1	-68.6	66	554	850	0.973	5.8
LMDZ-INCA	0.0756	-0.8	2.5	-53.1	47	585	NA	0.968	28.9
ECHAM-HAM	-0.0242	-1.7	1.1	NA	63	NA	NA	0.936	10.8
OsloCTM2	0.0934	-1.4	1.4	-49.3	62	616	885	0.939	30.1
SPRINTARS	0.155	-1.5	1.7	-47.4	60	525	913	0.958	30.3

Figures

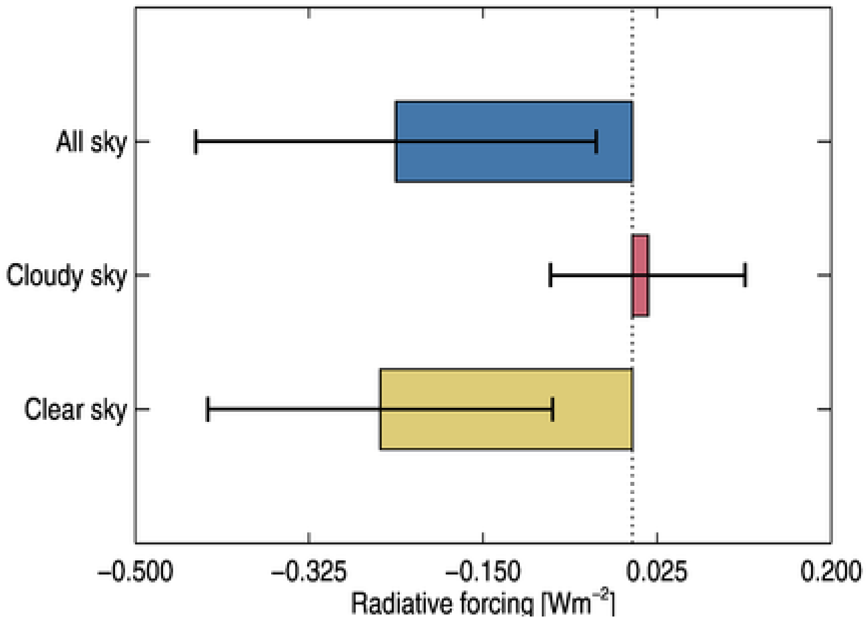

Figure 1: All sky anthropogenic RFari, and its decomposition into contributions from cloudy sky and clear sky areas (second and first term on the right hand of equation 1, respectively), based on AeroCom Phase II simulations (Myhre et al., 2013a).

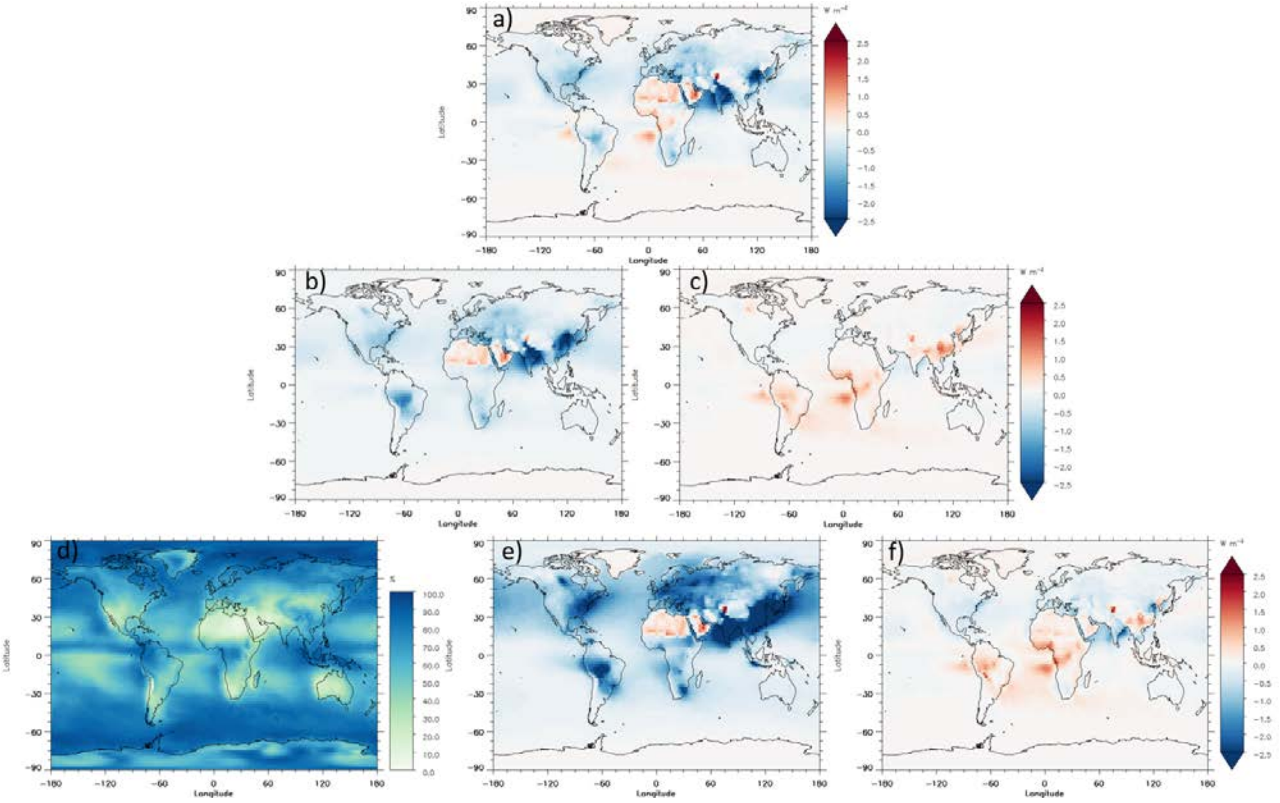

Figure 2. Annual mean all sky anthropogenic RFari and various terms from clear and cloudy skies simulated with OsloCTM3 (Lund et al., 2018). The panel in the top row show~~is~~ the all sky RFari a), the second row shows the contributions from clear (RFari$_{clear}$) and cloudy sky (RFari$_{cloud}$) (first b) and second term c) on right hand side on equation 1, respectively). The third row shows cloud fraction (AC) d), RFari$_{total-clear}$ e) and RFari$_{total-cloud}$, f) respectively~~ (see equation 1)~~. Panel d) on cloud fraction is showed in percent and the other panels in W m^{-2}.

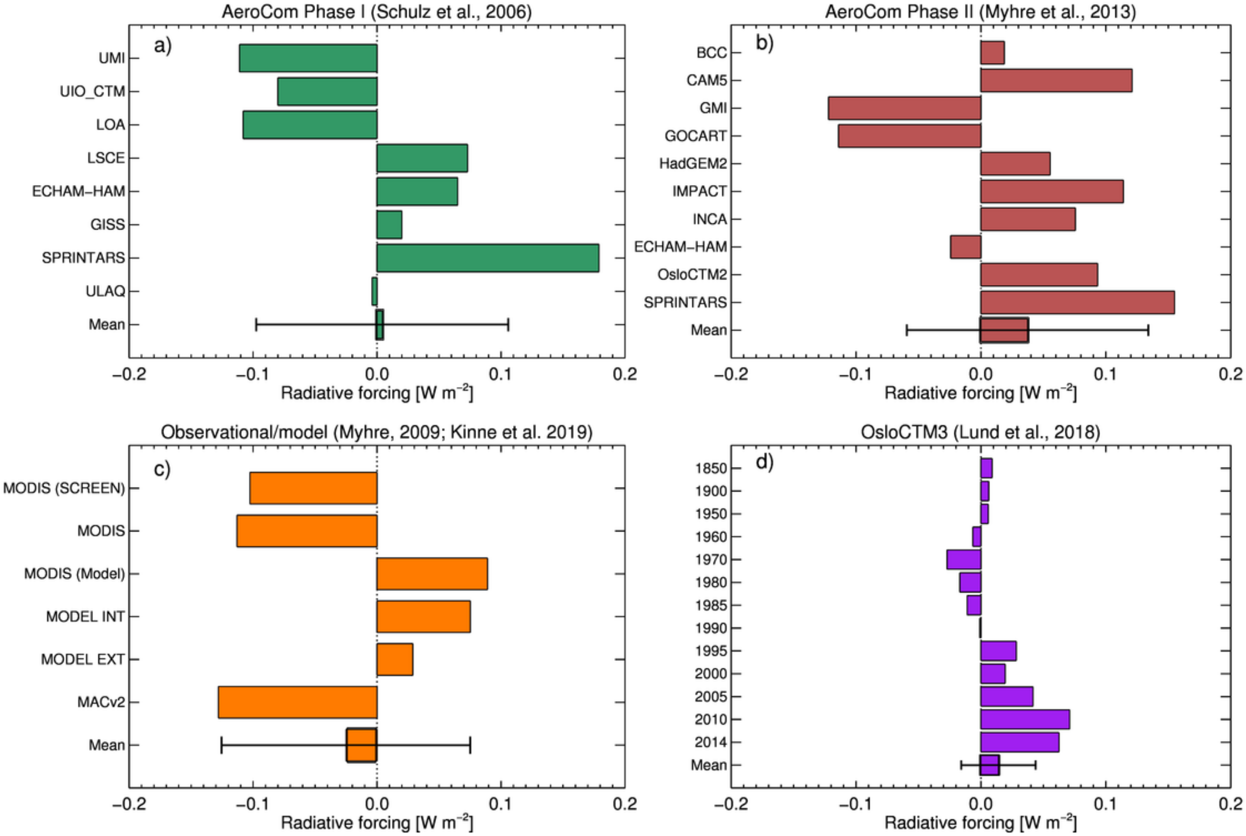

Figure 3. The contribution of cloudy sky to RFari from AeroCom Phase I (Schulz et al., 2006) ~~(~~a), AeroCom Phase II (Myhre et al., 2013a) ~~(~~b), combination of observational based and model simulation (Kinne, 2019a; Myhre, 2009), where mean and standard deviation are based on all methods used in the panel ~~(~~c), and time evolution from OsloCTM3 (Lund et al., 2018) ~~(~~d).The multi-model mean is shown by the bars and the one standard uncertainty range of the models is given by whiskers.

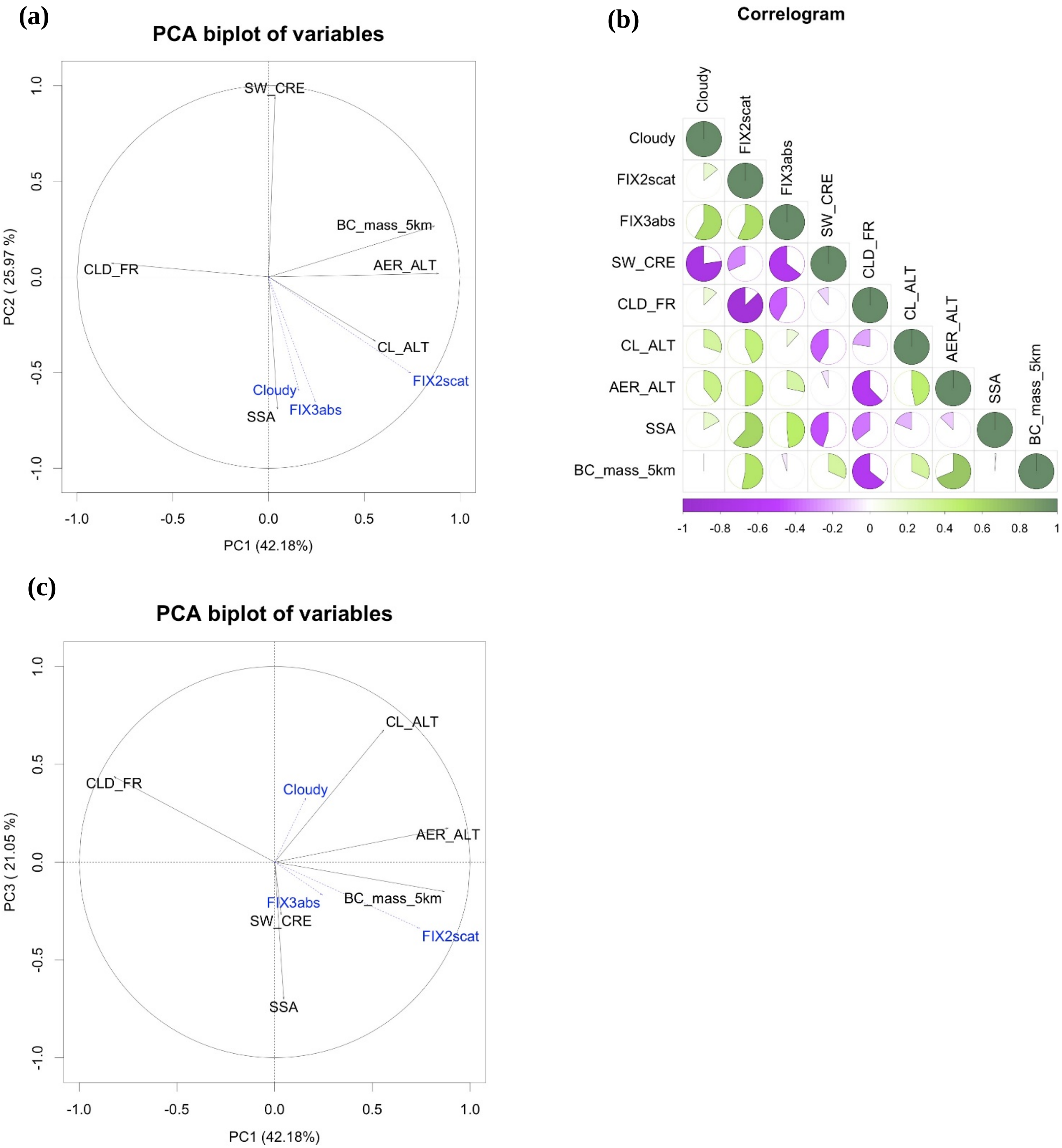
(a)
PCA biplot of variables
SW_CRE
BC_mass_5km
AER_ALT
CLD_FR
CL_ALT
FIX2scat
Cloudy
FIX3abs
SSA
PC2 (25.97 %)
PC1 (42.18%)
1.0
0.5
0.0
-0.5
-1.0
(b)
Correlogram
Cloudy
FIX2scat
FIX3abs
SW_CRE
CLD_FR
CL_ALT
AER_ALT
SSA
BC_mass_5km
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1
(c)
PCA biplot of variables
CL_ALT
CLD_FR
Cloudy
AER_ALT
BC_mass_5km
FIX3abs
SW_CRE
FIX2scat
SSA
PC3 (21.05 %)
PC1 (42.18%)

Figure 4. Multivariate data analysis of eight AeroCom Phase II models (Myhre et al., 2013a) using diagnostics shown in Table S1. Principal Component Analysis biplot of the variables ~~(~~a). The length of the arrows indicates the strength of the correlation each variable has in relation to PC1 and PC2, representing 42.2% and 26.0% of the variance respectively. Variables clustered together indicate positive correlation with each other. Variable opposing each other indicate negative correlation. Cloudy, FIX3abs and FIX2scat (in blue) are added as supplementary variables, and do not influence the projection of the other variables. This requirement is made since cloudy, FIX2scat and FIX3abs already depends on the other variable and see their correlation. FIX2scat and FIX3abs have fixed SSA globally and for all models, where the former experiment has pure scattering aerosol and FIX3abs has relatively low SSA (and thus high aerosol absorption). In (b), the correlogram shows the one to one linear regression correlation each variable has to each other. The correlation coefficients (r) are presented on a color scale from -1 (purple) to 0 (white) to +1 (green). The strength of the correlation is additionally presented as pie charts filling clockwise in green for positive correlations between two variables, and counter clockwise in purple for negative correlation, where they can range from empty and full pie charts, indicating an absolute correlation respectively from 0 to 1. Panel (c) is same as for (a), but for PC1 and PC3.