Response to referee comments on Radiative effects of inter-annually varying versus inter-annually invariant aerosol emissions from fires

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We thank the two referees for taking the time to evaluate the manuscript and to provide comments. Please find the referee comments (shown in blue) and our responses (shown in black) below. Quotes from the revised manuscript are shown in red.

1 Response to Referee #1 (D. S. Ward)

Grandey et al. compute the global radiative effects of fire aerosol emissions in a series of simulations with CAM5. In this experiment they compare the radiative effects that result when a monthly climatology is used to drive the emissions, to the radiative effects that result when emissions include interannual variability. The magnitude of the forcing is greater when the monthly climatology is used, suggesting a saturation of the first indirect aerosol effect, and shedding light on previous estimates of fire aerosol radiative forcings which typically use the climatology approach. There is no similarly large difference in precipitation between the two emissions schemes. Results are broken down by aerosol species and by region which highlights the greater interannual variability of fires in boreal regions compared to the tropics and subtropics.

The design of the experiment is thoughtful and well suited to answer the questions posed in the introduction. The main result, a 20% decrease in RF magnitude when using the interannually varying fire emissions in this model setup, has implications beyond fire modeling and for Earth System modeling since many major ESM projects use decadal mean fire emissions. I have some comments, mostly minor, and suggestions for discussing the saturation effect differently.

Thank you for taking the time to thoughtfully read the manuscript and for providing helpful suggestions. We respond to your comments below. Incidentally, we think your use of "monthly climatology" is a clear way to describe our FMEAN simulation. Hence, we have added the following sentence to the description of FMEAN in the Method section: "In other words, the FMEAN emissions consist of a monthly climatology." We have also incorporated "monthly climatology" elsewhere, e.g. in the Abstract ("Conventionally, many climate modelling studies have used an inter-annually invariant monthly climatology of emissions of fire aerosols.") and at the start of the Results section ("In most global climate modelling studies, a monthly climatology of aerosol emissions from fires is used and hence the inter-annual variability of the emissions is ignored.").

General comments:

1. The argument in the Discussion section (Pg 11 Lines 3-6) for saturation of the aerosol radiative effect on clouds for the high fire emission years and locations (as opposed to the steady mean emissions) seems right, but the test of this hypothesis, illustrated with Figure 9, is not as convincing. Figure 9 shows how the sublinear response of the indirect effect to increases in aerosols is evident for a few grid points in a region, but does not give reason to think that the emissions vs. RFP relationship at these three grid points should apply generally. Especially give that two of the five points selected do not show the same relationship. And since the five points in the figure were already chosen specially because they had the largest FMEAN-F{yyyy} difference (among the other criteria), the figure does not show that the saturation effect matters here necessarily.

What if you were to plot timeseries (maybe monthly?) of Boreal Asia region average OC emissions, CCN (I think there is a standard CCN 0.2%SS variable output in CAM5?) and RFP, or some metric of forcing efficiency, with each timeseries standardized so they could be plotted together. Just one F{yyyy} could be plotted with FMEAN I think. This could show whether peaks and valleys in the interannual varying OC/CCN timeseries are matched by similar increases and decreases in RFP or if this saturates at the high extremes as would be expected. I am not sure if this effect would be clearly visible on a regional average but this might be something to try.

I also suggest including a standard reference for the saturation effect (maybe at Pg 11, Line 4) such as Boucher and Pham (2002). And since the first indirect effect is not isolated from all aerosol/cloud effects in this study, i.e. lifetime effect, semi-direct effect, it would be helpful to include discussion of how the additional aerosol effects factor in to estimates of the saturation effect. Or how they cloud the conclusions that can be drawn.

Thank you for your suggestions, including that of plotting a timeseries. After considering this suggestion, we have decided not to include a monthly timeseries since this would be complicated by the fact that the RFP is highly dependent on solar insolation which exhibits a strong seasonal cycle over Boreal Asia. In order to address the question of generality, we have instead opted to plot a further figure of cloud shortwave RFP versus surface organic carbon aerosol concentration averaged across the whole Boreal Asia region for July and August. We have included this as a new Fig. S13 to complement Fig. 9, and have updated the text as follows:

"It is worth noting that the results presented in Fig. 9 have two obvious limitations. First, as pointed out above, surface organic carbon aerosol concentration may not always be a good proxy for the cloud condensation nuclei available to clouds. Second, these results may not apply generally to the Boreal Asia region because they represent only a relatively small sample of location-month combinations, selected because they have large "conventional" – "revised" differences.

"In order to partially address the second of these limitations, corresponding results showing cloud shortwave RFP versus surface organic carbon aerosol concentration averaged across the whole Boreal Asia region during the months of July and August are shown in Fig. S13. The first two observations about Fig. 9 listed above also apply to the Boreal Asia regional averages shown in Fig. S13: the surface organic carbon aerosol concentration is very similar between the "conventional" and "revised" approaches; and a logarithmic fit works well, with the cloud shortwave RFP scaling approximately linearly with the logarithm of the surface organic carbon aerosol concentration. However, the "conventional" approach still produces stronger cloud shortwave RFPs than would be expected from the regional average organic carbon aerosol concentrations, although this may be primarily due to the fact that the use of regional averages obscures differing spatial inhomogeneities in surface organic carbon aerosol concentration.

"Fig. 9 (especially Fig. 9c) and Fig. S13 provide some evidence in support of our hypothesis that the overestimation of cloud shortwave RFP strength occurs due to the non-linear influence of aerosols on clouds. In order to test this hypothesis more conclusively, it would be advantageous to perform idealised simulations designed to isolate the contribution of different aerosol sources, aerosol species, and aerosol effects. Such further analysis is outside the scope of the present study."

Thank you for suggesting that we include a reference for the saturation effect. We have added an additional sentence before the one in which we suggest that the indirect effects may saturate: "For example, there is evidence of a "sublinear dependence of cloud droplet concentrations on aerosol number" (Stevens and Feingold, 2009)."

2. Related to this, I think it would be helpful to include more details about how aerosols interact with the radiative budget in this model setup, probably in the Methods section 2.1. For example, mentioning the different effects that are included in the RFP such as the direct effect, semi-direct effect, etc., explaining how aerosols can perturb the radiative flux in this model setup. This is also important to explain for the hydrological response which is discussed in Section 3.2 and shown in Figure 8. Aerosol impacts on both stratiform and convective clouds are included even though aerosols in the model do not directly interact with the convective scheme (Section 2.1). So here is another instance where explaining how the aerosols indirectly impact convection in the methods would be helpful. We have adopted your suggestion, and have modified the paragraph in question to include the following: "Aerosol direct and semi-direct effects are included via coupling between the aerosols and the radiation. Aerosol indirect effects on stratiform clouds are included via coupling between the aerosols and the stratiform cloud microphysics (Morrison and Gettelman, 2008; Gettelman et al., 2010). As a result of these indirect effects (Ghan et al., 2012), CESM1-CAM5 produces a relatively strong total aerosol radiative effect compared to many other global climate models (Table 7 of Shindell et al., 2013). CAM5 does not include a representation of aerosol indirect effects on convective cloud microphysics. However, the aerosols may indirectly impact convection via interaction with the radiation."

Minor comments:

Pg 5, Lines 15-17: There are not many estimates of fire RF but you could include Chuang et al. (2002), -1.16 Wm-2 (first indirect effect only), and Ward et al. (2012), -1.38 Wm-2 (also CAM5), as additional points of comparison.

Thank you for pointing out these two studies. We have now included them: "The global mean net RFP of -1.3 W m^{-2} is comparable to that found by Clark et al. (2015, their "interpolation method" simulation) who also used CAM5. This value, which is dominated by the shortwave (see below), is also comparable to two previous estimates of the shortwave forcing associated with fire aerosols: an estimate of -1.4 W m^{-2} by Ward et al. (2012, their Table 5) who also used CAM5; and an estimate of -1.2 W m^{-2} by Chuang et al. (2002) who used CCM1/GRANTOUR to quantify the cloud albedo effect only."

Pg 6, Line 9: The foundation for the rest of the results section is developed very nicely in this section. For this line, please include how you define statistical significance here.

We have now added mention of the method used to test statistical significance, and refer readers to the table caption for details: "A "conventional" – "revised" difference of $-0.24 \,\mathrm{W}\,\mathrm{m}^{-2}$ is found (Table 1), primarily due to the cloud shortwave RFP component (Fig. 3a). Welch's *t*-test reveals that this difference is statistically significant (see Table 1 caption). These results indicate that the conventional approach of using inter-annually invariant fire aerosol emissions leads to a 23% overestimation of the negative RFP exerted by fire aerosols on the climate system (Fig. 5a)."

Pg 6, Lines 16-17: It is mentioned here and shown in Figure 5 that fire aerosol impacts on forcing extend over ocean but it seems like only land area is considered in the regional estimates? In Ward et al. (2012) we also found large forcings (< -5 Wm-2) over ocean for fire aerosols, in fact, globally the largest indirect aerosol forcings were for the marine stratocumulus decks off the coasts of western Africa and South America.

If forcings over ocean are not considered here, can you discuss how the answers might be different if these areas were included?

We have now modified the paragraph in question: "Although the discussion below focuses on land regions, readers should note that the radiative effects of the fire aerosols are not limited to land regions but extend over ocean regions downwind of fire sources. Net RFPs stronger than $-5 \,\mathrm{W}\,\mathrm{m}^{-2}$ occur over parts of the North Pacific, Tropical Pacific, and Tropical Atlantic oceans, including the stratocumulus decks off the coasts of western South America and western Africa (Fig. 6a,b). Ward et al. (2012) similarly identified strong cloud forcing associated with these stratocumulus decks. The presence of such highly inhomogeneous forcing over ocean may lead to perturbed surface temperature gradients which in turn may lead to large-scale circulation changes and precipitation impacts (Wang, 2015). However, changes in SST gradients are outside the scope of the present study which analyses results from prescribed-SST simulations."

Pg 7, Lines 26-27: I wonder if you can speculate why the BC emissions have a higher cloud sw forcing efficiency in these regions, and why the BC sw cloud forcing does not have a consistent sign in general when looking at all global regions. Perhaps this is a result of semi-direct effects (e.g. Sakaeda et al., 2011; Koch and Del Genio, 2010).

Following your comment, we have modified the sentences in question: "The cloud shortwave RFP is primarily driven by the organic carbon emissions (Fig. 3d). The black carbon emissions also drive a negative cloud shortwave RFP, possibly as a result of semi-direct effects leading to increased cloud water path (Fig. S14d) – Koch and Del Genio (2010) have previously highlighted that there are "several mechanisms by which absorbing aerosols may either increase or decrease cloud cover"."

Pg 8, Lines 4-6: This summary point for the South American region is clear and nicely explained. I think it is important to note again here that aerosols are only microphysically active for stratiform clouds. This could be especially important in the tropical regions.

Thank you. We have added a sentence following your suggestion: "However, if the model were to include a representation of aerosol indirect effects on convective cloud microphysics, the results might be different for these convectively active tropical regions."

Pg 9, Line 1: Why is EQAS divided into two sub-regions for this study?

We chose not to use the EQAS region due to the difficulty of distinguishing land from ocean in the Maritime Continent at the model resolution. We have modified the Fig. 1 caption to include: "Unlike the land-only GFED4.0s basisregions, the two Maritime Continent regions both include ocean as well as land, due to the difficulty of distinguishing land from ocean at a resolution of $1.9^{\circ} \times$ 2.5° over the Maritime Continent." We look at the Western Maritime Continent and the Eastern Maritime Continent separately due to our interest in these regions, especially the Western Maritime Continent.

Pg 10, Lines 9-12: This sentence left me wondering why this is the case ? that is that the fire aerosols lead to a precipitation increase. I am not sure it is feasible to tease out the reasons for this given the scope of this study, but it might be worth looking at what other changes are occurring such as changes in cloud fraction, cloud depths, CDNC, etc. that might provide some insight. Or maybe this has to do with differences in snow cover due to fire aerosols.

We have modified this paragraph to clarify that total precipitation is still

suppressed, even over the Boreal regions: "The exceptions are the two boreal regions where, although total precipitation is suppressed, large-scale precipitation is actually enhanced slightly by the fire aerosols (Fig. 7b–c). This enhancement of large-scale precipitation over the Boreal regions partially offsets the much stronger suppression of convective precipitation."

Pg 10, Lines 15-16: This is a good point, and might be a good place to note that Tosca et al. (2013) and Clark et al. (2015) do show a dynamical response in precipitation to fire aerosols using a slab ocean, although with non-interannually varying emissions. Another place these could be cited would be on/near Pg 9, Line 24.

Thank you for the suggestion. However, we think that discussing these references in this section would reduce clarity. At the end of the Discussion section, however, we do mention that "if SST feedbacks were to be included, the "conventional"—"revised" RFP differences would likely impact surface temperature gradients. Changes in surface temperature gradients are known to impact precipitation patterns (Wang, 2015)."

Pg 13, Line 8: It is great that the data from this study are made available and so easily accessible.

Thank you for the encouragement!

References:

Boucher, O. and M. Pham, History of sulfate aerosol radiative forcings, Geophys. Res. Lett., 29(9), doi:10.1029/2001GL014048, 2002.

Chuang, C. C., Penner, J. E., Prospero, J. M., Grant, K. E., Rau, G. H., Kawamoto, K.: Cloud susceptibility and the first aerosol indirect forcing: Sensitivity to black carbon and aerosol concentrations, J. Geophys. Res., 107, 4564, doi:10.1029/2000JD000215, 2002.

Clark, S. K., Ward, D. S., and Mahowald, N. M.: The sensitivity of global climate to the episodicity of fire aerosol emissions, Journal of Geophysical Research: Atmospheres, 120, 11,589-11,607, doi:10.1002/2015JD024068, 2015.

Koch, D. and Del Genio, A. D.: Black carbon semi-direct effects on cloud cover: review and synthesis, Atmos. Chem. Phys., 10, 7685-7696, doi:10.5194/acp-10-7685-2010, 2010.

Sakaeda, N., Wood, R., and Rasch, P. J.: Direct and semidirect aerosol effects of southern African biomass burning aerosol, J. Geophys. Res.-Atmos., 116, D12205, doi:10.1029/2010JD015540, 2011.

Tosca, M., J. Randerson, and C. Zender (2013), Global impact of contemporary smoke aerosols from landscape fires on climate and the hadley circulation, Atmos. Chem. Phys., 12, 28,069-28,108.

Ward, D., S. Kloster, N. Mahowald, B. Rogers, J. Randerson, and P. Hess (2012), The changing radiative forcing of fires: Global model estimates for past, present and future, Atmos. Chem. Phys., 12, 10,857-10,886.

Thank you once again for all your insightful comments.

2 Response to Referee #2 (A. Veira)

Aerosol emissions from open-burning fires show a distinct inter-annual variability. This manuscript by Grandey et al. quantifies the impact of the inter-annual fire emission variability on global and regional radiative effects. The authors present results from simulations with the Community Atmosphere Model version 5 (CAM5) which compare top-of-atmosphere radiative flux pertubations for inter-annually varying and inter-annually invariant emission inventories. The application of inter-annually varying fire emissions reduces the mean net radiative aerosol effect by about 23% globally and up to 58% regionally. The simulated global changes in precipitation, however, are not significant.

The scientific topics addressed by this manuscript are within the scope of ACP and the study design is adequate for investigation of the given research questions. Overall, the paper is well structured and well written. The results are presented in a clear and understandable way, supported by neat figures and tables. The detailed discussion of the individual regional impacts appears slightly long-winded in some parts, but it provides substantial information for the overall conclusions of the study and might as well be beneficial for subsequent future studies. The reference list is comprehensive and largely reasonable. Due to the large differences in the overall fire aerosol radiative forcing between CAM5 and other global aerosol models, slightly more detailed descriptions and discussions of the used emission inventory uncertainties, the applied microphysical aerosol properties and the cloud shortwave radiative flux pertubations in the model, are desirable. In addition, I have some minor comments and suggestions.

I recommend publication in ACP if the authors sufficiently address the following comments in their revised manuscript.

Thank you for your recommendation and for your suggestions, including your comment that "slightly more detailed descriptions and discussions of the used emission inventory uncertainties, the applied microphysical aerosol properties and the cloud shortwave radiative flux perturbations in the model, are desirable" – we deal with this overarching comment in the context of your general comments and specific comments below.

General comments:

1. The fire emission data set GFED applied in this study is a well-established and frequently used fire emission data set and there are no objections to use it for the purpose of this study. However, fire emission estimates based on other retrieval methods (e.g. the Global Fire Assimilation System GFAS, Kaiser et al., 2012) provide emission estimates which show substantial regional emission flux differences compared to GFED (e.g. Kaiser et al., 2012 and Zhang et al., 2014). Therefore I suggest at least to mention the emission estimate uncertainties in more detail in Section 2.2 and in the discussion of the results.

Thank you for highlighting this source of uncertainty. Following your suggestion, we have added two sentences about uncertainties associated with fire emissions in the Method section: "Several fire emission inventories are available (van der Werf et al., 2010; Wiedinmyer et al., 2011; Kaiser et al., 2012). There is uncertainty associated with these fire emission inventories – for example, Lee et al. (2016) found that differences between two of these inventories contributes to uncertainty in modelled aerosol concentration over Southeast Asia."

We have also added a sentence in the second paragraph of the Results section: "In addition to large uncertainty associated with the parameterisation of indirect effects, there is also uncertainty associated with emissions of fire aerosols."

2. Globally, the radiative flux perturbation (RFP) of more than 1 Wm-2 of both, the 'conventional' and the 'revised' fire emission approaches presented in Section 3.1.1 are surprisingly large compared to most other studies which provide top-of-atmosphere radiative forcings of fire emissions which range between -0.3 Wm-2 and +0.1 Wm-2 (for a list of references see Veira et al., 2015, Section 5, page 7188). Can you provide a more detailed explanation, why this is the case? The comparatively large values of the anthropogenic / total RFP in CAM5.1 provided by Shindell et al., 2013 (cited on P3, L16 and P5, L18 in this manuscript) do not necessarily explain the very large radiative effect of the fire aerosol emissions in particular. Might the black carbon (BC) and organic carbon (OC) atmospheric lifetime largely differ in the other models? Which refractive indices for BC and OC are used in CAM5 (for a comparison to ECHAM-HAM2, see Zhang et al., 2012)? What specific implementation in the cloud microphysics is included in CAM5, but not in the other models?

Thank you for encouraging us to consider this in more detail and for pointing out the further references. We now reference the studies by Jones et al. (2007) and Unger et al. (2010) in the Introduction: "Chuang et al. (2002), Jones et al. (2007), Unger et al. (2010), Ward et al. (2012), Clark et al. (2015) and Veira et al. (2015) all appreciated the importance of modeling the indirect effects of fire aerosols."

We considered including the results of Jones et al. (2007) in the discussion of our global mean net RFP at the start of the Results section. However, we decided not to in the end due to the fact that Jones et al. (2007) used year-1860 fire emissions as their reference rather than zero fire emissions, hence their year-2000–year-1860 net RFP of $-0.3 \,\mathrm{W}\,\mathrm{m}^{-2}$ cannot be directly compared with our FMEAN–F0 net RFP – the difference in reference scenario will likely make a large difference.

Following you comment, and a comment from Reviewer #1, we have adapted the paragraph in question: "The "conventional" global mean net RFP associated with fire aerosols is -1.3 W m^{-2} (Table 1). Note that this RFP is relative to simulation F0 which has zero emissions of fire aerosols. If a different reference, for example year-1850 emissions of fire aerosols, were to be chosen, the global mean net RFP would be smaller in magnitude. The global mean net RFP of -1.3 W m^{-2} is comparable to that found by Clark et al. (2015, their "interpolation method" simulation) who also used CAM5. This value, which is dominated by the shortwave (see below), is also comparable to two previous estimates of the shortwave forcing associated with fire aerosols: an estimate of -1.4 W m^{-2} by Ward et al. (2012, their Table 5) who also used CAM5; and an estimate of -1.2 W m^{-2} by Chuang et al. (2002) who used CCM1/GRANTOUR to quantify the cloud albedo effect only. However, a value of $-1.3 \,\mathrm{W}\,\mathrm{m}^{-2}$ is much larger than ECHAM6-HAM2's fire aerosol net RFP of $-0.2 \,\mathrm{W}\,\mathrm{m}^{-2}$ (Veira et al., 2015). This discrepancy appears to be primarily due to differences in the parameterisation of indirect effects, especially the sensitivity of stratiform clouds to organic carbon aerosol emissions (see below). CESM-CAM5, which produces an anthropogenic aerosol year-2000-year-1850 net RFP of $-1.5 \,\mathrm{W}\,\mathrm{m}^{-2}$ (Ghan et al., 2012), is known to produce a stronger net RFP than many other global climate models (Shindell et al., 2013). In addition to large uncertainty associated with the parameterisation of indirect effects, there is also uncertainty associated with the emissions of fire aerosols."

Since the net RFP is dominated by the cloud shortwave RFP, the refractive indices of BC and OC are not directly relevant here, especially since the semi-direct effect is relatively small in CAM5. Similarly, we think that any perturbation to the BC and OC lifetimes, which are approximately 4 days in CAM5 (Liu et al., 2012; Clark et al., 2015), would be at most a second-order effect. The references for the cloud microphysics (Morrison and Gettelman, 2008; Gettelman et al., 2010) are provided in the Method section.

3. Cloud shortwave RFP is identified as major driver of the total net RFP differences between the simulations with inter-annually invariant and interannually varying fire emissions for many regions in the results section. Can you provide any numbers on high and low cloud cover changes in these regions? Do you see distinct differences in vertical cloud profiles? Some numbers would be helpful to further explain the non-linear saturation effects which are nicely presented in the discussion section.

Thank you for your suggestion. Rather than including cloud cover changes, we have now decided to include a figure showing changes in grid-box average cloud water path in the supplemental material (Fig. S14). We refer to this new figure a handful of times in the Results section, including in the "Global distribution" section: "For the regions discussed below, negative cloud short-wave RFPs are generally associated with increases in grid-box average liquid water path and total water path (Fig. S14). Negative cloud longwave RFPs are generally associated with decreases in ice water path (Figs. S11 and S14)."

Specific Comments:

Abstract:

P1, L14-15: '[...], we need to more accurately quantify the effects of aerosols [...]'. This sub-clause is a very general statement and does not provide specific outcomes of this study. Therefore the last sentence of the abstract can be shortened by pointing directly to the specific importance of the inter-annual variability of fire aerosols quantified in this study.

Following your suggestion, we have re-written the final sentence of the abstract: "In order to improve understanding of the climate system, we need to take into account the inter-annual variability of aerosol emissions."

Introduction:

P1, L21-22: The references referring to peat fires in Indonesia are slightly misleading in this context. Although the sulfur content of the biomass varies substantially, all fire categories named in line 16-17 include emissions of sulfur dioxide as the emission factor inventories by Andreae and Merlet, 2001 and Akagi et al., 2011 demonstrate.

Thank you for highlighting that this sentence is potentially misleading. We have now dropped this sentence, and have mentioned sulphate in the preceding sentence instead (see response to comment below).

P1, L22-23: Does the statement 'These aerosols have a negative impact on [. . .]' refer to sulfur dioxide emissions from fire? It should be clarified that Lelieveld et al., 2015 quantified the overall impact of biomass burning aerosols (including black carbon and organic carbon) on human health, not the human health impact of sulfur emissions from fires in particular.

Following our response to your previous comment (see above), this ambiguity has now been resolved: "Other pollutants include aerosols, containing organic carbon, black carbon, and sulphate. These aerosols have a negative impact on air quality and human health (Lelieveld et al., 2015)."

P2, L1: 'Aerosols scatter or absorb incoming sunlight'. The conjunction 'or' is misleading as both processes, scattering and absorption, apply to most atmospheric aerosol species.

We have re-written this: "Aerosols can scatter and absorb incoming sunlight".

P2, L7-8: '[. . .] many other aerosol effects on clouds have also been proposed.' Please provide examples of these 'other' effects.

Following your suggestion, we now mention two further examples: "In addition to these two indirect effects, other aerosol effects on clouds have also been proposed, including a glaciation indirect effect (Lohmann, 2002) and the convective invigoration hypothesis (Andreae et al., 2004; Rosenfeld et al., 2008). For further discussion of possible aerosol effects on clouds, readers are referred to the review papers written by Lohmann and Feichter (2005), Tao et al. (2012), and Rosenfeld et al. (2014)."

P2, L11 and L14: In the reference list, there are two references given for Wang et al., 2009. Therefore it is unclear, which of the two papers is referred to in line 11 and line 14, respectively.

The first of these contains an "et al." (Wang et al., 2009), whereas the second is a single-author paper (Wang, 2009). We appreciate that this may be confusing. However, we do not know how to make the distinction clearer while still remaining within the standard ACP reference style guidelines.

Methods:

P4, L14-18: In this paragraph, it should be explicitly stated that the black carbon and organic carbon fire emission estimates from GFED applied in this study represent an important source of uncertainty (see general comment 1).

We have adopted your suggestion. Please see our response to your first general comment.

Results:

P5, L26: Can you provide numbers for the increase in snow cover? It would be interesting to see the quantitative relationship between snow cover increase and surface albedo change.

Thank you for the suggestion to look at snow cover. In the section of the Results focusing on the Boreal regions, we now write that "during the summer months of June–August, the fraction of the ground covered by snow increases from 4.0% in F0 to 5.3% in FMEAN."

P9, L19: Comparing Figure S1 to Table 1, it is interesting to see that a statistically significant overestimation of the net RFP strength is found for Central America and South-east Asia. In contrast, the Eastern Maritime Continent and Temperate North America show temporal OC emission patterns of similiar magnitude like those of Central America and Southeast Asia, respectively (Figure S1), but nevertheless no statistically significant overestimation of the net RFP is simulated for the conventional approach in these regions. Might these findings indicate that the influence of the meteorology varies significantly in these regions or is this due to the saturation effects?

This is an interesting observation. Differences in meteorology likely play a major role.

P10, L1-2: Why does black carbon suppress large-scale precipitation in such a way? If available, an analysis of vertical temperature profiles / atmospheric stability changes might be helpful for a better understanding although the discussion of these relations does of course not represent the focus of this study.

We have added a sentence stating that "It is difficult to interpret the mechanisms behind these global mean features, due to spatial inhomogeneity in the regional response (see below)."

Discussion:

P11, L1: 'Rather, across much of the globe, the "conventional" approach leads to a systematic overestimation of net RFP [. . .]'. Table 1 and Fig. 6c show that the statistically significant overestimation is largely limited to some specific regions. Therefore the use of the statement 'much of the globe' is inappropriate in this context.

Following your suggestion, we have replaced "across much of the globe" with "for some regions".

P12, L12: What 'other factors' do you think of? Might interaction with other, non-fire related aerosol species play a role?

Thank you for encouraging us to think further about what "other factors" might include. After considering this comment, and also Referee #1's first general comment (see above), we have decided to drop the references to "other factors".

Conclusions:

P12, L28-30: This paragraph represents a nice description of those regions, where ignoring the inter-annual variability leads to an overestimation of the net radiative effect of fire aerosols. However, as shown in Table 1, there are many other regions, where no significant differences between the two scenarios are found and this should also be stated here.

Following your comment, we have included "for some regions": "Regionally, for some regions, the "conventional" approach leads to an even larger overestimation of the strength of the net radiative effect of the fire aerosols".

P13, L3-5: At this point I suggest to provide some implications and ideas for subsequent future studies, which could be initiated by the results and conclusions presented in this paper, e.g. the quantification of daily vs. monthly fire emissions or a deeper analysis of the cloud micro-physical processes which are responsible for the found saturation effects.

Following your comment, we have added a paragraph recommending avenues for further research: "Following the findings presented in this paper, we suggest three avenues for further research. First, similar simulations could be performed using other aerosol-climate models in order to test whether the conclusions of this study apply more generally if different parameterisations of aerosol indirect effects are used. Second, idealised simulations could be performed to improve understanding of the saturation of aerosol indirect effects. Third, coupled atmosphere–ocean simulations could be performed to investigate the impact of inter-annually varying emissions on the hydrological slow response and other components of the climate system, including modes of climate variability."

All figures presented in the manuscript represent elaborated and easily readable visualizations of high quality with detailed figure captions. Figure 6 and Figure 8 are neat and self-explanatory figures, but in the final manuscript the figure size should be increased in order to improve the readability of the stippling.

Thank you! We hope to increase the figure size in the final ACP manuscript.

References:

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039-4072, doi:10.5194/acp-11-4039-2011, 2011.

Andreae, M. O., and P. Merlet (2001), Emission of trace gases and aerosols from biomass burning, Global Biogeochem. Cycles, 15(4), 955-966, doi:10.1029/2000GB001382.

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Radiative effects of inter-annually varying versus inter-annually invariant aerosol emissions from fires

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Abstract. Open-burning fires play an important role in the Earth's climate system. In addition to contributing a substantial fraction of global emissions of carbon dioxide, they are also a major source of atmospheric aerosols such as containing organic carbon, black carbon, and sulphate. These "fire aerosols" can influence the climate via both direct and indirect radiative effects. In this study, we investigate these radiative effects and the hydrological fast response using the Community Atmosphere Model

- 5 version 5 (CAM5). Emissions of fire aerosols exert a global mean net radiative effect of -1.0 W m^{-2} , dominated by the cloud shortwave response to organic carbon aerosol. The net radiative effect is particularly strong over boreal regions. By Conventionally, many climate modelling studies have used an inter-annually invariant monthly climatology of emissions of fire aerosols. However, by comparing simulations using inter-annually varying emissions versus inter-annually invariant emissions, we find that ignoring the inter-annual variability of the emissions can lead to systematic overestimation of the strength of the
- 10 net radiative effect of the fire aerosols. Globally, the overestimation is $+23 \% (-0.2 \text{ W m}^{-2})$. Regionally, the overestimation can be substantially larger. For example, over Australia and New Zealand the overestimation is $+58 \% (-1.2 \text{ W m}^{-2})$, while over Boreal Asia the overestimation is $+43 \% (-1.9 \text{ W m}^{-2})$. The systematic overestimation of the net radiative effect of the fire aerosols is likely due to the non-linear influence of aerosols on clouds. However, ignoring inter-annual variability in the emissions does not appear to significantly impact the hydrological fast response. In order to improve understanding of the
- 15 climate system, we need to more accurately quantify the effects of aerosols, taking into account important characteristics such as take into account the inter-annual variability of aerosol emissions.

1 Introduction

There are many types of open-burning fires, caused by both natural and human influences. Broad categories include agricultural waste burning, grassland fire, peat fire, and various types of forest fire (van der Werf et al., 2010). In addition

20 to producing heat, these fires emit pollutants into the atmosphere. One such pollutant is carbon dioxide, contributing to climate change – Page et al. (2002) estimate that the large-scale burning of peat and forests in 1997 in Indonesia alone emitted the equivalent of 13–40% of global annual emissions of carbon dioxide from fossil fuels. Other pollutants include aerosols, containing organic carbonand black carbon. Some fires, including burning of sulphur-containing peat in Indonesia

(Gras et al., 1999; Balasubramanian et al., 1999; Langmann, 2003), also emit sulphur dioxide, leading to the formation of sulphateaerosol,

black carbon, and sulphate. These aerosols have a negative impact on air quality and human health (Lelieveld et al., 2015).

Aerosols also affect climate both regionally and globally. Aerosols scatter or can scatter and absorb incoming sunlight, cooling the earth's surface, an effect known as the "direct aerosol effect" (Haywood and Boucher, 2000). Absorbing aerosols, such as black carbon, warm the atmosphere, potentially affecting cloud formation

- (the "semi-direct effect; Ackerman, 2000)(the "semi-direct effect"; Ackerman, 2000). Hygroscopic aerosols, such as sulphate, also play an important role in cloud formation, acting as the cloud condensation nuclei on which cloud droplets condense. Hence changing the availability of sulphate aerosol hygroscopic aerosols may affect the droplet size and number in clouds, potentially changing cloud reflectivity (the "cloud albedo effect" or "first indirect effect"; Twomey, 1974, 1977), or and cloud
- 10 lifetime (the "cloud lifetime effect" or "second indirect effect"; Albrecht, 1989). In addition to these two indirect effects, many other aerosol effects on clouds have also been proposed(<u>Tao et al., 2012; Rosenfeld et al., 2014</u>)..., including a glaciation indirect effect (Lohmann, 2002) and the convective invigoration hypothesis (Andreae et al., 2004; Rosenfeld et al., 2008). For further discussion of possible aerosol effects on clouds, readers are referred to the review papers written by Lohmann and Feichter (2005), Tao et al. (2012), and Rosenfeld et al. (2014).
- Surface cooling and atmospheric heating perturb regional temperature gradients, potentially affecting large-scale circulations such as monsoon systems. For example, many studies have suggested that aerosols impact the South Asian monsoon (e.g. Chung et al., 2002; Lau et al., 2006; Wang et al., 2009; Bollasina et al., 2011; Lee et al., 2013; Lee and Wang, 2015). Remote impacts are also possible. For example, we have recently demonstrated that anthropogenic emissions of aerosols from Asia may affect rainfall in remote locations such as Australia and the Sahel (Grandey et al., 2016). Remote impacts have also been

20 explored by Menon (2002), Wang (2007, 2009), and Teng et al. (2012).

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Most studies investigating the climate impacts of aerosols have focused on anthropogenic aerosols emitted by industry or power plants. Fewer studies have focused on the radiative and climate impacts of aerosols emitted from open-burning fires, which we will refer to as "fire aerosols", the focus of our current study.

Clark et al. (2015) recently investigated the radiative and climate effects of sub-monthly variability in fire emissions. They
found that the "monthly-mean emissions approximation holds roughly in the tropics, where fires are more frequent and less
episodic; however it does not perform as well in the boreal regions" (Clark et al., 2015). Jeong and Wang (2010) demonstrated
the importance of seasonal variation in the emissions of fire aerosols. They found that the "seasonality of biomass burning
emissions uniquely affects the global distributions of convective clouds and precipitation" (Jeong and Wang, 2010). In light of
the findings of Jeong and Wang (2010), we use seasonally varying emissions in our study. Beyond seasonal variability, Feng

30 and Christopher (2014) and Sena and Artaxo (2015) showed that the direct radiative effects of fire aerosols also exhibit interannual variability, something that is not taken into account in most climate modelling studies. We complement the sub-monthly focus of Clark et al. (2015) and the seasonal focus of Jeong and Wang (2010) by investigating inter-annually varying emissions.

Three of the above-mentioned studies (Jeong and Wang, 2010; Feng and Christopher, 2014; Sena and Artaxo, 2015) considered the direct radiative effects of fire aerosols but did not consider other radiative effects. However, as pointed out by Jacobson

35 (2014), the radiative effects of fire aerosols are not limited to the direct effect. Jiang et al. (2016) found that the indirect effects

of fire aerosols may be substantially larger than the direct effect. <u>Chuang et al. (2002)</u>, Jones et al. (2007), Unger et al. (2010), Ward et al. (2012), Clark et al. (2015), and Veira et al. (2015) all appreciated the importance of modelling the indirect effects of fire aerosols. In addition to representing the direct and semi-direct effects, the aerosol–climate model used in this study also includes a representation of indirect effects on stratiform clouds. However, some other possible effects, such as cloud

5 absorption effects (Jacobson, 2014), are not represented.

Using the ECHAM6-HAM2 aerosol-climate model, Veira et al. (2015) recently investigated the influence of fire emission height. Although they found that emission height can influence the radiative effects of fire aerosols, they concluded that "Significant improvements in aerosol wildfire modeling modelling likely depend on better emission inventories and aerosol process modeling modelling rather than on improved emission height parametrizations" (Veira et al., 2015). In this study, we prescribe

10 emission heights according to fire type, as described in the Method section.

Building on these previous studies, we explore the radiative effects associated with inter-annually varying emissions of fire aerosols. We use Global Fire Emissions Database (GFED) aerosol emissions to drive a global aerosol–climate model. We quantify the radiative effects for the globe and several different regions (Fig. 1). We also discuss the hydrological fast response. In the Discussion section, we focus on one primary research question: how do the effects of inter-annually varying emissions

15 differ from those of inter-annually invariant <u>climatological</u> emissions? In the context of this research question, we consider the non-linear influence of aerosols on clouds.

2 Method

2.1 Model configuration

The Community Earth System Model (CESM) version 1.2.2, which includes the Community Atmosphere Model version 5
(CAM5) (Neale and Coauthors, 2012), is used. CAM5 contains a modal aerosol model with three lognormal modes (MAM3) (Liu et al., 2012). Aerosol direct and semi-direct effects are included via coupling between the aerosols and the radiation. Aerosol indirect effects on stratiform clouds are represented included via coupling between the aerosols and the stratiform cloud microphysics (Morrison and Gettelman, 2008; Gettelman et al., 2010). As a result of these indirect effects (Ghan et al., 2012), CESM1-CAM5 produces a relatively strong total aerosol radiative effect compared to many other global climate models

25 (Table 7 of Shindell et al., 2013). CAM5 does not include a representation of aerosol indirect effects on convective cloud microphysics. However, the aerosols may indirectly impact convection via interaction with the radiation.

A finite volume grid with a horizontal resolution of approximately $1.9^{\circ} \times 2.5^{\circ}$ and 30 levels is used for CAM5. The land model is run at the same horizontal resolution. Greenhouse gas concentrations, sea-surface temperatures (SSTs), and sea-ice are prescribed using year-2000 climatological values.

30 The "prescribed-SST" approach is suitable for diagnosing radiative flux perturbations (RFPs) (Haywood et al., 2009) and the hydrological "fast response" (Bala et al., 2010) associated with aerosols. RFPs are diagnosed by calculating the difference in the top-of-atmosphere radiative flux between two prescribed-SST simulations forced with different aerosol emissions. In contrast to a strict definition of "radiative forcing", the RFP approach allows clouds and precipitation to adjust to the aerosol forcing via fast feedback processes. Hence RFPs allow quantification of the indirect effects of aerosols on clouds. To facilitate decomposition of the RFPs (Ghan, 2013), the radiation scheme is called twice at each radiation time step. The first radiation call includes all aerosol species. The second radiation call, which is purely diagnostic, excludes all aerosol species, allowing diagnosis of "clean-sky" fluxes.

5 2.2 Emissions

Emissions of organic carbon, black carbon, sulphur dioxide, primary sulphate, dimethyl sulphide, and secondary organic aerosol precursors mostly follow the default MAM3 emissions for year-2000. The exception is that fire emissions of organic carbon, black carbon, sulphur dioxide, and primary sulphate are modified.

As in the "interpolation method" simulation of Clark et al. (2015), monthly fire emissions are used in this study. The
Several fire emission inventories are available (van der Werf et al., 2010; Wiedinmyer et al., 2011; Kaiser et al., 2012). There is uncertainty associated with these fire emission inventories – for example, Lee et al. (2016) found that differences between two of these inventories contributes to uncertainty in modelled aerosol concentration over Southeast Asia. We have chosen to use monthly fire emissions are obtained from the Global Fire Emission Database version 4.0 with small fires included (GFED4.0s) (revised version of van der Werf et al., 2010), which has data available from 1997 onwards. The recommended

15 GFED4.0s emission factors are used to convert partitioned dry matter emissions to emissions of organic carbon, black carbon, and sulphur dioxide. These emissions are then conservatively remapped from the GFED4.0s grid $(0.25^{\circ} \times 0.25^{\circ})$ to the grid used in the MAM3 emissions files (approximately $1.9^{\circ} \times 2.5^{\circ}$).

Following Liu et al. (2012), emission height profiles are based on Table 4 of Dentener et al. (2006). For boreal forest fires, the "Boreal (Eurasia)" emission height profile is used instead of the "Boreal (Canada)" profile. The Dentener et al. (2006)
20 height profiles are linearly interpolated to the higher resolution vertical levels used in the MAM3 emission files.

The GFED4.0s fire categories are subsequently mapped to the MAM3 emission sector categories. The GFED4.0s "agricultural waste burning" category corresponds to the MAM3 agricultural waste burning category; the GFED4.0s "savanna, grassland, and shrubland fires" category corresponds to the MAM3 grass fire category; and the GFED4.0s "boreal forest fires", "temperate forest fires", "tropical forest fires (deforestation and degradation)", and "peat fires" categories are combined into

25 the MAM3 forest fire category. The resulting global and regional organic carbon, black carbon, and sulphur dioxide annual emissions for the different simulations (described below) are shown in Fig. 2 and supplemental material Figs. S1–S5. Globally, averaged across 1997–2006, fire emissions are responsible for 60% (18 Tg yr⁻¹) of the total organic carbon emissions of 30 Tg yr⁻¹, 28% (2 Tg yr⁻¹) of the total black carbon emissions of 7 Tg yr⁻¹, and 2% (2.6 Tg yr⁻¹) of the total sulphur dioxide emissions of 131 Tg yr⁻¹—it. It is worth noting that the total black carbon emissions of 7 Tg yr⁻¹ used in this study

30 may represent an underestimate - Cohen and Wang (2014) estimate that global BC emissions are 17.8 ± 5.6 Tg yr⁻¹.

It is assumed that 2.5% of the sulphur dioxide is emitted as primary sulphate (Dentener et al., 2006; Liu et al., 2012). Size distributions of emitted organic carbon, black carbon, and primary sulphate follow the size distributions described in the supplementary material of Liu et al. (2012).

2.3 Simulations

Fifteen simulations are performed. The first of these, F0, is a control simulation. In F0, fire emissions of organic carbon, black carbon, sulphur dioxide, and primary sulphate are set to zero. In this paper, RFPs are calculated with respect to simulation F0, rather than a pre-industrial control.

- 5 Ten simulations use fire emissions for the ten different years between 1997 and 2006. For example, F1997 uses year-1997 fire emissions, F1998 uses year-1998 fire emissions etc. Together, this ensemble of ten simulations is referred to as {Fyyyy}. Comparison of {Fyyyy} with F0 reveals the radiative effect of inter-annually varying emissions of fire aerosols. We refer to this as the "revised" approach, in contrast to the "conventional" approach described in the next paragraph.
- In contrast to the {Fyyyy} ensemble, the "FMEAN" simulation uses mean emissions averaged across 1997–2006 for each 10 month. The seasonal cycle is retained, but inter-annual variability is removed. In other words, the FMEAN emissions consist of a monthly climatology. We refer to this as the "conventional" approach. Comparison of FMEAN ("conventional") with {Fyyyy} ("revised") reveals the influence of ignoring inter-annual variability.

"OMEAN" combines the organic carbon emissions of FMEAN with the black carbon, sulphur dioxide, and primary sulphate emissions of F0 (i.e. zero emissions of black carbon and sulphur from fires). Similarly, "BMEAN" combines the black carbon

15 emissions of FMEAN with the organic carbon, sulphur dioxide, and primary sulphate emissions of F0; and "SMEAN" combines the sulphur dioxide and primary sulphate emissions of FMEAN with the black carbon and organic carbon emissions of F0. Comparison of OMEAN, BMEAN, and SMEAN with FMEAN reveals the relative contributions of organic carbon, black carbon, and sulphur dioxide.

The first two years of each simulation are treated as spin-up. Simulations F0 and FMEAN are each run for 44 years, providing
an analysis period of 42 years. Simulations OMEAN, BMEAN, SMEAN, and each member of the {Fyyyy} ensemble (i.e. F1997 etc.) are each run for fourteen years, providing an analysis period of twelve years. The total analysis period for {Fyyyy} is 120 years, because {Fyyyy} consists of ten separate simulations. When deciding the simulation lengths, we sought a balance between the improved statistical power of larger sample sizes and the computational expense of the simulations.

3 Results

25 **3.1 Radiative effects of fire aerosols**

3.1.1 Global mean

In most global climate modelling studies, the inter-annual variability a monthly climatology of aerosol emissions from fires is used and hence the inter-annual variability of the emissions is ignored. Here, we also start with this "conventional" approach of ignoring inter-annual variability.

30 The "conventional" global mean net RFP associated with fire aerosols is -1.3 W m^{-2} (Table 1),—. Note that this RFP is relative to simulation F0 which has zero emissions of fire aerosols. If a different reference, for example year-1850 emissions

of fire aerosols, were to be chosen, the global mean net RFP would be smaller in magnitude. The global mean net RFP of -1.3 W m^{-2} is comparable to that found by Clark et al. (2015, their "interpolation method" simulation). This is smaller than CESM-CAM5's anthropogenic aerosol year-2000 – year-1850 net RFP of -1.5 who also used CAM5. This value, which is dominated by the shortwave (see below), is also comparable to two previous estimates of the shortwave forcing associated

- 5 with fire aerosols: an estimate of -1.4 W m^{-2} (Ghan et al., 2012), but by Ward et al. (2012, their Table 5) who also used CAM5; and an estimate of -1.2 W m^{-2} by Chuang et al. (2002) who used CCM1/GRANTOUR to quantify the cloud albedo effect only. However, a value of -1.3 W m^{-2} is much larger than ECHAM6-HAM2's fire aerosol net RFP of -0.2 W m^{-2} (Veira et al., 2015). This discrepancy appears to be primarily due to differences in the parameterisation of indirect effects, especially the sensitivity of stratiform clouds to organic carbon aerosol emissions (see below). CESM-CAM5, which produces
- 10 an anthropogenic aerosol year-2000-year-1850 net RFP of -1.5 W m⁻² (Ghan et al., 2012), is known to produce a strong net RFP compared to stronger net RFP than many other global climate models (Shindell et al., 2013). In addition to large uncertainty associated with the parameterisation of indirect effects, there is also uncertainty associated with the emissions of fire aerosols.

This global mean net RFP of -1.3 W m^{-2} is dominated by the cloud shortwave RFP of -1.1 W m^{-2} (Fig. 3a, Table 2), primarily driven by the organic carbon emissions (Fig. 3a), in general agreement with the findings of Jiang et al. (2016). The

organic carbon emissions also drive a corresponding increase in global mean grid-box average liquid water path (Fig. S14a).

The cloud longwave RFP of -0.1 W m^{-2} (Fig. 3a, Table S1) is also driven by the organic carbon emissions (Fig. 3a). Interestingly, again in agreement with Jiang et al. (2016), the cloud longwave RFP associated with the fire aerosols is negative, in contrast to the results of Ghan et al. (2012) who found a positive cloud longwave RFP associated with anthropogenic aerosols (see also Gettelman et al., 2012). The occurrence of negative cloud longwave RFP values corresponds to decreasing fractional

cover of high clouds and decreasing ice water path, especially over equatorial land regions (Fig. S11).

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The organic carbon emissions drive a small surface albedo RFP of -0.1 W m^{-2} (Fig. 4a, Table S2), due to increasing snow cover in Boreal North America (see below) and other parts of the world, likely in response to the cloud shortwave RFP. The organic carbon emissions also drive a small direct effect RFP , but this is more than offset by the positive direct effect RFP of the black carbon emissions (Fig. 4a), resulting in a small positive direct effect RFP overall (Table S3).

When the emissions for individual years are used to drive separate simulations in the ten-member {Fyyyy} ensemble, the inter-annual variability of the RFP components is revealed. Most of the variability in global mean net RFP is dominated by the

cloud shortwave RFP component (Fig. 3a). The global mean net RFP ranges from -0.8 W m⁻² in F2000, which has the lowest organic carbon emissions, to -1.3 W m⁻² in F1998, which has the second highest organic carbon emissions (Figs. 2aand, 3a). Interestingly, this maximum net RFP of -1.3 W m⁻² in F1998 is comparable to the "conventional" global mean net RFP, suggesting that the global impact of the fire aerosols may saturate (see Discussion). The simulation with the highest organic carbon emissions (Fig. 2a), F1997, has a comparatively weak global mean net RFP of -0.9 W m⁻² (Fig. 3a), demonstrating that the global mean net RFP response is not a monotonic function of global annual organic carbon emissions (Fig. 5a). This suggests that the location and timing of emissions play an important role in determining the RFP response.

In contrast to the "conventional" approach of using inter-annually invariant emissions, comparison of {Fyyyy} with F0 reveals the mean effect of inter-annually varying emissions of fire aerosols. The "revised" global mean net RFP is -1.0 W m^{-2} (Table 1). A statistically significant "conventional"—"revised" difference of -0.24 W m^{-2} is found (Table 1), primarily due to the cloud shortwave RFP component (Fig. 3a). This shows that Welch's *t*-test reveals that this difference is statistically

5 significant (see Table 1 caption). These results indicate that the conventional approach of using inter-annually invariant fire aerosol emissions leads to a 23 % overestimation of the negative RFP exerted by fire aerosols on the climate system (Fig. 5a). Further discussion of the effect of ignoring inter-annual variability can be found in the Discussion section.

3.1.2 Global distribution

The global distribution of the radiative effects associated with fire aerosols is highly inhomogeneous (Fig. 6a,b). Particularly
strong net RFPs occur over high latitude boreal regions, and parts of South America, Southern Hemisphere Africa, Australia, and the Maritime Continent. These regions will be discussed below. The-

Although the discussion below focuses on land regions, readers should note that the radiative effects of the fire aerosols are not limited to land regions, but extend over ocean regions downwind of fire sources. Net RFPs stronger than -5 W m^{-2} occur over parts of the North Pacific, Tropical Pacific, and Tropical Atlantic oceans, including the stratocumulus decks off the coasts

- 15 of western South America and western Africa (Fig. 6a,b). Ward et al. (2012) similarly identified strong cloud forcing associated with these stratocumulus decks. The presence of such highly inhomogeneous forcing over ocean may lead to perturbed surface temperature gradients which in turn may lead to large-scale circulation changes and precipitation impacts (Wang, 2015). However, changes in SST gradients are outside the scope of the present study which analyses results from prescribed-SST simulations.
- 20 For the regions discussed below, negative cloud shortwave RFPs are generally associated with increases in grid-box average liquid water path and total water path (Fig. S14). Negative cloud longwave RFPs are generally associated with decreases in ice water path (Figs. S11, S14).

Organic carbon emissions are almost entirely responsible for the negative net RFPs associated with fire aerosols (Fig. S9a). By comparison, the contributions of black carbon and sulphur dioxide are much smaller (Fig. S9b,c).

25 3.1.3 Boreal North America and Boreal Asia

Forest fires dominate the fire aerosol emissions from both Boreal North America (Fig. 2b) and Boreal Asia (Fig. 2c). These boreal regions are prone to the strongest net RFPs of any of the land regions (Table 1) and are therefore discussed first.

Boreal North America has a "conventional" net RFP of -6.5 W m^{-2} (Table 1). This net RFP is dominated by the cloud shortwave RFP of -5.9 W m^{-2} (Table 2), driven by the organic carbon emissions (Fig. 3b). As pointed out by Jiang et al.

30 (2016), "the large cloud liquid water path over land areas of the Arctic favors the strong fire aerosol indirect effect" (Jiang et al., 2016). The next largest component is the surface albedo RFP of -0.6 W m^{-2} (Table S2), driven by both organic carbon and sulphur emissions (Fig. 4b), reflecting an increase in snow cover – during the summer months of June–August, the fraction

of the ground covered by snow increases from 4.0% in F0 to 5.3% in FMEAN. In comparison to the surface albedo RFP, the cloud longwave RFP (Fig. 3b, Table S1) and direct effect RFP (Fig. 4b, Table S3) are much smaller.

Compared to Boreal North America, Boreal Asia has a slightly weaker "conventional" net RFP of -6.2 W m^{-2} (Table 1). As was the case for Boreal North America, the net RFP for Boreal Asia is dominated the cloud shortwave RFP of -5.8 W m^{-2}

5 (Table 2), driven by the organic carbon emissions (Fig. 3c). The surface albedo RFP of -0.3 W m^{-2} (Table S2) and cloud longwave RFP of -0.2 W m^{-2} (Table S1) are also driven by the organic carbon emissions (Figs. 4cand, 3c). The direct effect RFP is much smaller (Table S3).

For both boreal regions, the large inter-annual variability of organic carbon emissions contributes to large variability in net RFP (Fig. 5b,c). Over Boreal North America, a strong correlation of -0.83 between the annual organic carbon emissions and

- 10 the net RFP demonstrates that the annual total organic carbon emissions can explain 69% of the variance in net RFP (assuming a linear relationship). F2001, which has the lowest emissions, has a net RFP of -2.5 W m^{-2} ; while F2004, which has the highest emissions, has a net RFP of -6.9 W m^{-2} (Figs. 3b and 2b 3b 5b). This variability in the net RFP is dominated by variability in the cloud shortwave RFP (Fig. 3b).
- Over Boreal Asia, a correlation of -0.70 demonstrates that the variability in the annual total organic carbon emissions can explain 49 % of the variance in net RFP (Fig. 5c). F2004, which has the lowest emissions, has a net RFP of -2.7 W m⁻²; while F2003, which has the highest emissions, has a net RFP of -5.1 W m⁻² (Figs. 3c and 2c, 3c, 5c). F1998, which has much lower emissions than F2003, has an even stronger net RFP of -6.0 W m⁻², indicating that the regional annual total emissions are not the sole determining factor of net RFP strength. This suggests that variability in timing and the specific location of the fires also plays a role, possibly due to the non-linear influence of aerosols on clouds (see Discussion).
- 20 When the inter-annually variability of emissions is taken into account, the "revised" net RFPs are significantly weaker than the "conventional" net RFPs over both boreal regions. Over Boreal North America, the "conventional" approach of ignoring inter-annual variability leads to a 32 % overestimation of net RFP strength (Fig. 5b, Table 1) and a 33 % overestimation of cloud shortwave RFP strength (Table 2). Over Boreal Asia, the "conventional" approach leads to a 43 % overestimation of net RFP strength (Fig. 5c, Table 1) and a 47 % overestimation of cloud shortwave RFP strength (Table 2).

25 3.1.4 Northern Hemisphere South America and Southern Hemisphere South America

Both grass fires and forest fires substantially contribute to the fire aerosol emissions from both South American regions (Fig. 2d,h). After the boreal regions, Northern Hemisphere South America has the next largest "conventional" net RFP (Table 1). In contrast to the boreal regions, the net RFP of -3.8 W m^{-2} is dominated by the cloud longwave RFP of -2.6 W m^{-2} (Fig. 3d, Table S1), driven by the organic carbon emissions (Fig. 3d). The negative cloud longwave RFP is associated with a decrease in high cloud fraction and a decrease in ice water path (Fig. S11).

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The cloud shortwave RFP of -1.1 W m^{-2} (Table 2) also contributes to the net RFP . Interestingly, the black carbon emissions cause a substantial negative over Northern Hemisphere South America. The cloud shortwave RFP , although this cloud shortwave RFP is weaker than that of is primarily driven by the organic carbon emissions (Fig. 3d). The direct effect RFP and surface albedo RFP are much smaller. However, the black carbon emissions also drive a negative cloud shortwave RFP.

(Fig. 4d, Tables S2 and S3) 3d), possibly as a result of semi-direct effects leading to increased cloud water path (Fig. S14d) – Koch and Del Genio (2010) have previously highlighted that there are "several mechanisms by which absorbing aerosols may either increase or decrease cloud cover".

In contrast to Northern Hemisphere South America, Southern Hemisphere South America has a weaker "conventional" net

5 RFP is -1.8 W m^{-2} (Table 1), with approximately equal contributions of -0.8 W m^{-2} coming from the cloud shortwave RFP (Table 2) and the cloud longwave RFP (Fig. 3h, Tables 2 and Table S1). Both the cloud shortwave RFP and the cloud longwave RFP are primarily driven by the organic carbon emissions (Fig. 3h).

Net RFP exhibits inter-annual variability over both South American regions. For example, over Northern Hemisphere South America, F2006, which has the lowest emissions, has a net RFP of -2.7 W m^{-2} ; while F2003, which has the highest emissions,

10 has a net RFP of -4.7 W m⁻² (Figs. 3d and 2d, 3d, 5d). Variability in the annual total organic carbon emissions can explain 59 % of the variance in net RFP over Northern Hemisphere South America (Fig. 5d) and 74 % of the variance in net RFP over Southern Hemisphere South America (Fig. 5h).

For both South American regions, the "revised" net RFP is comparable to the "conventional" net RFP (Table 1). In fact, the "conventional"—"revised" net RFP differences are statistically insignificant, indicating that the "conventional" approach

15 of ignoring inter-annual variability does not significantly impact net RFP over either South American region. However, if the model were to include a representation of aerosol indirect effects on convective cloud microphysics, the results might be different for these convectively active tropical regions.

3.1.5 Australia and New Zealand

As was the case for the South American regions, both grass fires and forest fires contribute to the fire aerosol emissions from 20 the "Australia and New Zealand" region (Fig. 2e). The "conventional" net RFP is -3.3 W m^{-2} (Table 1). As was the case over the Boreal regions, the net RFP over Australia and New Zealand is dominated the cloud shortwave RFP (Table 2), driven by the organic carbon emissions (Fig. 3e). The cloud longwave RFP, surface albedo effect RFP, and direct effect RFP are all statistically insignificant (Tables S1–S3).

Only a weak correlation exists between annual total organic <u>carbon</u> emissions and net RFP over Australia and New Zealand (Fig. 5e). Of the {Fyyyy} ensemble members, F2003 has the highest organic carbon emissions (Fig. 2e) yet has the weakest net RFP of -0.9 W m^{-2} (Figs. 3eand, 5e). F2002, which has comparable organic carbon emissions, has a much larger net RFP of -4.1 W m^{-2} . It is clear that the annual total emission of organic carbon is not the primary driver of inter-annual variability in net RFP over Australia and New Zealand.

The "revised" net RFP of -2.1 W m^{-2} is significantly weaker that the "conventional" net RFP of -3.3 W m^{-2} (Table 1).

30 The "conventional" approach overestimates the strength of net RFP by 58 % (Fig. 5e), the largest percentage difference found for any of the regions discussed in this section.

3.1.6 Southern Hemisphere Africa

In contrast to all the regions discussed above, the fire aerosol emissions from Southern Hemisphere Africa are almost completely driven by grass fires (Fig. 2f). The "conventional" net RFP is -3.3 W m^{-2} (Table 1). The cloud shortwave RFP of -2.0 W m^{-2} (Table 2) and the cloud longwave RFP of -1.4 W m^{-2} (Table S1) are both driven by the organic carbon emissions (Fig. 3f).

Net RFP ranges from -2.2 W m^{-2} in F1999 to -3.7 W m^{-2} in F1997, F2000, and F2005 (Fig. 3f). The correlation between annual total organic carbon emissions and net RFP is very weak (Fig. 5e). Variability in the annual total organic carbon emissions can explain less than 3% of the variance in net RFP. Other factors, such as the specific timing and location of emissions, must drive the inter-annual variability in net RFP.

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The "revised" net RFP of -3.2 W m^{-2} is comparable to the "conventional" net RFP of -3.3 W m^{-2} (Table 1). The difference is statistically insignificant. This indicates that the "conventional" approach does not significantly impact net RFP over Southern Hemisphere Africa.

3.1.7 Western Maritime Continent

Forest fires dominate the fire aerosol emissions from the Western Maritime Continent (Fig. 2g). The "conventional" net RFP is -2.5 W m^{-2} (Table 1). In contrast to the regions discussed above, with the exception of Northern Hemisphere South America, the net RFP is dominated by the cloud longwave RFP of -1.7 W m^{-2} (Fig. 3g, Table S1), driven by the organic carbon emissions (Fig. 3g). The cloud shortwave RFP is actually statistically insignificant (Table 2).

A relatively strong correlation exists between annual organic carbon emissions and net RFP, with variability in annual organic carbon emissions able to explain 53% of the variance in net RFP (Fig. 5g). One outlier likely contributes disproportionately

to the correlation: F1997 has by far the highest fire aerosol emissions (Fig. 2g) also has the largest net RFP of -3.0 W m^{-2} (Fig. 3g). Among the other {Fyyyy} ensemble members, the net RFP varies between -0.4 W m^{-2} and -2.2 W m^{-2} . The relationship between organic carbon emissions and net RFP appears to be non-linear (Fig. 5g).

The "revised" net RFP of -1.6 W m^{-2} is significantly weaker that the "conventional" net RFP of -2.5 W m^{-2} (Table 1). The "conventional" approach overestimates the strength of net RFP by 49 % (Fig. 5g). Interestingly, although the net RFP is dominated by the cloud longwave RFP, the "conventional"–"revised" difference in cloud longwave RFP is relatively small (-0.2 W m^{-2}) and statistically insignificant (Table S1). The "conventional"–"revised" difference in cloud shortwave RFP is larger (-0.6 W m^{-2}) , although this is also statistically insignificant (Table 2). These statistically insignificant differences in cloud longwave RFP and cloud shortwave RFP combine to form a statistically significant difference in net RFP.

3.1.8 Other land regions

Six of the other eight land regions have statistically significant "conventional" net RFPs ranging from -0.6 W m^{-2} to -1.6 W m^{-2} (Table 1). The "conventional" approach leads to a statistically significant overestimation of the strength of the net RFP over only two of these six regions: Central America and Southeast Asia (Table 1).

3.2 The hydrological fast response

In addition to facilitating the calculation of RFPs, the prescribed-SST simulations analysed in this paper facilitate investigation of the hydrological fast response (Bala et al., 2010). The hydrological fast response to the fire aerosols is discussed here; investigation of the hydrological slow response, which depends on the ocean response, may be investigated in future work.

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5 CAM5, in common with most global climate models, diagnoses two categories of precipitation: the component that is diagnosed by the convection scheme is referred to as "convective precipitation"; while the component that is diagnosed by the large-scale stratiform cloud scheme is referred to as "large-scale precipitation". We refer to the sum of these two components as the "total precipitation".

Globally, the fire aerosols suppress total precipitation by 2 mm/year on average (Table 3). The convective and large-scale components are both suppressed by approximately 1 mm/year (Fig. 7a). Although black carbon and sulphur dioxide were found to make only a small contribution to the net RFP of the fire aerosols, these two species play a major role in the global hydrological fast response to fire aerosols: black carbon and sulphur dioxide, rather than organic carbon, contribute to the suppression of convective precipitation; and black carbon exerts the strongest suppression of large-scale precipitation (Fig. 7a). It is difficult to interpret the mechanisms behind these global mean features, due to spatial inhomogeneity in the regional

15 response (see below).

Regionally, the strongest suppression of total precipitation by black carbon occurs over the western tropical Pacific Ocean (Fig. S10b), with the caveat that statistical significance is absent. In contrast, organic carbon often plays a more important role in the hydrological fast response over many land regions (Fig. S10a, Fig. 7b–h). For example, our results suggest that a strong suppression of precipitation over Southern Hemisphere Africa is primarily driven by organic carbon emissions (Fig. 7f), in contrast to the conclusions of Hodnebrog et al. (2016) who suggest that black carbon also plays an important role.

For all seven regions shown in Fig. 7, the organic carbon emissions suppress both total precipitation and convective precipitation. The organic carbon emissions also suppress large-scale precipitation over five of the regions (Fig. 7d–h). The exceptions are the two Boreal regions , whereboreal regions where, although total precipitation is suppressed, large-scale precipitation is actually enhanced slightly by the fire aerosols : over Boreal North America, the enhancement is driven by the organic carbon

25 emissions (Fig. 7b); whereas over Boreal Asia, organic carbon, black carbon, and sulphur dioxide all appear to contribute (Fig. 7c). b–c). This enhancement of large-scale precipitation over the Boreal regions partially offsets the much stronger suppression of convective precipitation.

Using inter-annually invariant fire emissions does not significantly affect the hydrological fast response of total precipitation either globally or over any of the regions, as shown by the lack of statistically significant "conventional"—"revised" differences

30 in Table 3 and Fig. 8c. However, if an interactive ocean model were to be used, the "conventional"—"revised" RFP differences will-would likely impact SSTs which in turn will-would likely impact the hydrological slow response.

4 Discussion: how do the effects of inter-annually varying emissions differ from those of inter-annually invariant climatological emissions?

Most global climate modelling studies use inter-annually invariant emissions of fire aerosols. In this study, the FMEAN simulation also followed this "conventional" approach of ignoring inter-annual variability. In contrast, the {Fyyyy} ensemble

5 facilitates calculation of "revised" RFPs that take into account the inter-annual variability of the emissions. Consideration of "conventional"—"revised" RFP differences reveals the impact of the "conventional" approach of ignoring inter-annual variability of emissions.

Globally, the "conventional" approach causes net RFP strength to be overestimated by 23% (Table 1, Fig. 5a). Over four six of the fifteen regions (Fig. 1), the "conventional" approach also causes net RFP strength to be statistically significantly

10 overestimated (Table 1). Over Australia and New Zealand, the overestimation is as large as 58 % (Fig. 5e). Over Boreal Asia, an overestimation of 43 % (Fig. 5c) corresponds to a "conventional"—"revised" RFP difference of -1.9 W m^{-2} (Table 1).

When the net "conventional"—"revised" net RFP differences are calculated at the model output resolution of $1.9^{\circ} \times 2.5^{\circ}$, statistically significant negative differences occur across 5.2% of the globe by area after controlling the False Discovery Rate (Benjamini and Hochberg, 1995; Wilks, 2016) (Fig. 6c). In contrast, statistically significant positive differences occur across only 0.5% of the globe. There is no clear signal of positive "conventional"—"revised" differences. Hence, there is no clear

evidence that the "conventional" approach of ignoring inter-annual variability ever leads to an underestimation of the net RFP strength. Rather, across much of the globe for some regions, the "conventional" approach leads to a systematic overestimation of net RFP strength (Fig. 6c, Table 1), dominated by an overestimation of the cloud shortwave RFP strength (Table 2). We hypothesise that the overestimation of cloud shortwave RFP strength occurs due to the non-linear influence of aerosols

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20 on clouds. For example, there is evidence of a "sublinear dependence of cloud droplet concentrations on aerosol number" (Stevens and Feingold, 2009). At high concentration levels, the aerosol indirect effects may saturate. Therefore, compared to using inter-annually varying emissions, spreading out the aerosol emissions in time by averaging across different years will likely lead to a stronger indirect effect on average.

In order to test this hypothesis, we have produced scatterplots of cloud shortwave RFP versus surface organic carbon aerosol concentration for different months and locations within the Boreal Asia region (Fig. 9). Boreal Asia has been chosen because it is the region with the largest "conventional"—"revised" difference in cloud shortwave RFP (Table 2). The first location—month combination (Fig. 9a) has been selected because it is the location—month combination with the largest "conventional"—"revised" cloud shortwave RFP difference. The second location—month combination (Fig. 9b) is the combination with the next largest difference, with the additional criterion that the location is at least 10° (either longitude or latitude)

30 from the previously selected location (Fig. 9a) so as to sample more widely geographically. Similarly, the third, fourth and fifth combinations (Fig. 9c–e) are those with the next largest differences at locations at least 10° from any of the previously selected locations. The method for selecting these five location–month combinations is shown in Fig. S12.

Looking at the scatterplots of cloud shortwave RFP versus surface organic carbon <u>aerosol</u> concentration (Fig. 9), the following observations can be made: For all five location-month combinations, the surface organic carbon <u>aerosol</u> concentration is very similar between the "conventional" and "revised" approaches. On a larger scale, no significant "conventional"—"revised" difference in surface organic carbon <u>aerosol</u> concentration is found for either the globe or any of the fifteen regions (Table S4). This suggests that "conventional"—"revised" differences in cloud shortwave RFP are not driven by differences in aerosol

lifetime. (For further discussion of how fire episodicity may impact the aerosol lifetime, see Clark et al., 2015)

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(For further discussion of how fire episodicity may impact aerosol lifetime, see Clark et al., 2015).

- 2. For three of the location-month combinations (Fig. 9a-c), a non-linear relationship between surface organic carbon <u>aerosol</u> concentration and cloud shortwave RFP is evident. A logarithmic fit works well, with the cloud shortwave RFP scaling approximately linearly with the logarithm of the surface organic carbon <u>aerosol</u> concentration. The nature of the logarithmic relationship causes the mean of the {Fyyyy} ensemble to have a weaker cloud shortwave RFP than an individual {Fyyyy} ensemble member with a similar surface organic carbon <u>aerosol</u> concentration, an effect that is particularly evident in Fig. 9a,c. The remaining two location-month combinations (Fig. 9d,e) do not exhibit a clear relationship between surface organic carbon <u>aerosol</u> concentration and cloud shortwave RFP. For these two location-month combinations, the surface organic carbon <u>aerosol</u> concentration is not a good proxy for the cloud condensation nuclei available to clouds.
- 3. For one of the location-month combinations (Fig. 9c), the cloud shortwave RFP for the FMEAN simulation is very similar to that of the {Fyyyy} ensemble member that has a similar surface organic carbon aerosol concentration. For this location-month combination, it appears that the non-linear relationship between surface organic carbon aerosol concentration and cloud shortwave RFP can explain most of the large "conventional"—"revised" difference in cloud shortwave RFP. This supports our hypothesis that the "conventional" overestimation of cloud shortwave RFP strength occurs due to the non-linear influence of aerosols on clouds. The interpretation is less clear in Fig. 9a,b, where the FMEAN cloud shortwave RFP is stronger than would be expected for the given surface organic carbon concentration. Although the non-linear relationship between surface organic carbon concentration and cloud shortwave RFP can explain part of the "conventional"—"revised" difference in cloud shortwave RFP, it would appear that other factors also play a role. aerosol concentration.

It is worth noting that the results presented in Fig. 9 have two obvious limitations. First, they represent a relatively small sample of location-month combinations. Second, as pointed out above, surface organic carbon aerosol concentration may not always be a good proxy for the cloud condensation nuclei available to clouds. Second, these results may not apply generally to

30 the Boreal Asia region because they represent only a relatively small sample of location-month combinations, selected because they have large "conventional"-"revised" differences.

Notwithstanding these two limitations, In order to partially address the second of these limitations, corresponding results showing cloud shortwave RFP versus surface organic carbon aerosol concentration averaged across the whole Boreal Asia region during the months of July and August are shown in Fig. 9-S13. The first two observations about Fig. 9 listed above also

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apply to the Boreal Asia regional averages shown in Fig. S13: the surface organic carbon aerosol concentration is very similar between the "conventional" and "revised" approaches; and a logarithmic fit works well, with the cloud shortwave RFP scaling approximately linearly with the logarithm of the surface organic carbon aerosol concentration. However, the "conventional" approach still produces stronger cloud shortwave RFPs than would be expected from the regional average organic carbon

5 aerosol concentrations, although this may be primarily due to the fact that the use of regional averages obscures differing spatial inhomogeneities in surface organic carbon aerosol concentration.

Fig. 9 (especially Fig. 9c) provides and Fig. S13 provide some evidence in support of our hypothesis that the overestimation of cloud shortwave RFP strength occurs due to the non-linear influence of aerosols on clouds. However, other factors might also play a role. In order to test this hypothesis more conclusively, it would be advantageous to perform idealised simulations

10 designed to isolate the contribution of different aerosol sources, aerosol species, and aerosol effects. Such further analysis is outside the scope of the present study.

Despite the impact on net RFP, using inter-annually invariant emissions does not significantly impact the hydrological fast response, as shown by the absence of statistically significant "conventional"—"revised" differences in Table 3 and Fig. 8c. The prescribed-SST simulations analysed here do not facilitate analysis of the hydrological slow response. However, if SST feed-

15 backs were to be included, the "conventional"—"revised" RFP differences would likely impact surface temperature gradients. Changes in surface temperature gradients are known to impact precipitation patterns (Wang, 2015). Hence it is foreseeable that the "conventional" approach of ignoring inter-annual variability might influence the hydrological slow response.

5 Conclusions

In this study, we have investigated the radiative effects of inter-annually varying emissions of fire aerosols. Our prescribed-SST 20 CAM5 simulation results suggest that fire aerosols exert a net radiative effect of -1.0 W m^{-2} on the climate system. This net radiative effect is dominated by the cloud shortwave response to organic carbon emissions. Boreal regions are especially susceptible. For example, over Boreal North America, the net radiative effect of the fire aerosols is -4.9 W m^{-2} on averageand can be as strong as , ranging from -2.5 W m^{-2} to -6.9 W m^{-2} in some different years.

The Conventionally, a monthly climatology of emissions of fire aerosols has often been used to drive global climate models. 25 This "conventional" approach of ignoring the inter-annual variability of the emissions can lead to a significant overestimation of the net radiative effect of the fire aerosols. Compared to our "revised" approach of using inter-annually varying emissions, the "conventional" approach of ignoring inter-annual variability can lead to a +23 % overestimation of the global mean net radiative effect. Regionally, for some regions, the "conventional" approach ean lead leads to an even larger overestimation of the strength of the net radiative effect of the fire aerosols: +32 % (-1.6 W m⁻²) over Boreal North America, +43 % (-1.9 W m⁻²)

30 over Boreal Asia, +58 % (-1.2 W m⁻²) over Australia and New Zealand, and +49 % (-0.8 W m⁻²) over the Western Maritime Continent.

There is evidence to suggest that the overestimation associated with the "conventional" approach arises due to the non-linear influence of aerosols on clouds. Compared to using inter-annually varying emissions (our "revised" approach), spreading out

the aerosol emissions in time by averaging across different years (the "conventional" approach) will likely lead to a stronger indirect effect on average.

Following the findings presented in this paper, we suggest three avenues for further research. First, similar simulations could be performed using other aerosol–climate models in order to test whether the conclusions of this study apply more generally

5 if different parameterisations of aerosol indirect effects are used. Second, idealised simulations could be performed to improve understanding of the saturation of aerosol indirect effects. Third, coupled atmosphere–ocean simulations could be performed to investigate the impact of inter-annually varying emissions on the hydrological slow response and other components of the climate system, including modes of climate variability.

Fire aerosols play an important role in the climate system, exerting large radiative effects regionally. In order to improve un-

10 derstanding of the climate system, we need to more accurately quantify these radiative effects, including critical characteristics such as seasonality and inter-annual variability.

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Data availability. The CESM-CAM5 data analysed in this manuscript are available via Figshare: http://dx.doi.org/10.6084/m9.figshare.3497705.

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Figure 1. (a) The GFED4s GFED4.0s basis-regions, re-gridded to the model resolution of approximately $1.9^{\circ} \times 2.5^{\circ}$ using nearest-neighbour interpolation: Boreal North America (BONA), Temperate North America (TENA), Central America (CEAM), Northern Hemisphere South America (NHSA), Southern Hemisphere South America (SHSA), Southern Hemisphere Africa (SHAF), Northern Hemisphere Africa (NHAF), the Middle East (MIDE), Europe (EURO), Boreal Asia (BOAS), Central Asia (CEAS), Southeast Asia (SEAS), Australia and New Zealand (AUST), and Equatorial Asia (EQAS). (b) The two regions used instead of EQAS in this study: the Western Maritime Continent (MCW), and the Eastern Maritime Continent (MCE). MCW and MCE-Unlike the land-only GFED4.0s basis-regions, the two Maritime Continent regions both include ocean as well as land, unlike due to the land-only GFED4s basis-regionsdifficulty of distinguishing land from ocean at a resolution of $1.9^{\circ} \times 2.5^{\circ}$ over the Maritime Continent. In the other figures and tables, "Northern Hemisphere" is often abbreviated to "NH", while "Southern Hemisphere" is abbreviated to "SH".



Figure 2. Organic carbon (OC) <u>aerosol</u> annual emissions for (a) the globe and (b)–(h) seven regions (see Fig. 1). The seven regions included here are those with the largest FMEAN–F0 net radiative flux perturbation differences (Table 1). "Other" refers to year-2000 emissions from non-fire sectors, such as domestic emissions and fossil fuel emissions from industry. Organic carbon emissions for another eight regions are shown in Fig. S1. Black carbon and sulphur dioxide emissions are shown in Figs. S2–S5.



Figure 3. Cloud shortwave, cloud longwave, and net (all components, shortwave plus longwave) top-of-atmosphere radiative flux perturbations (RFPs) for the globe and seven regions. Results for another eight regions are shown in Fig. S6. The RFPs are relative to simulation F0 and are area-weighted. Error bars represent combined standard error, using annual regional mean data as the input – e.g. for the F1997 net RFP error bars, the combined standard error equals $\sqrt{\frac{s_{F0}^2}{N_{F0}} + \frac{s_{F1997}^2}{N_{F1997}}}$, where s_{F0} and s_{F1997} are the corrected sample standard deviations, and $N_{F0} = 42$ and $N_{F1997} = 12$ are the sample sizes.



Figure 4. Surface albedo, aerosol direct effect, and net (all components, shortwave plus longwave) top-of-atmosphere radiative flux perturbations (RFPs) for the globe and seven regions. Results for another eight regions are shown in Fig. S7. The RFPs are relative to simulation F0. Error bars represent combined standard error (see Fig. 3 caption).



Figure 5. Net radiative flux perturbation (RFP) versus organic carbon (OC) emissions for the globe and seven different regions. The RFPs are relative to simulation F0. Each point represents the results for a different simulation or group of simulations (in the case of "Mean of {Fyyyy}"). Error bars represent combined standard error (see Fig. 3 caption). The Pearson's product-moment correlation coefficient (r), calculated from the ten-member {Fyyyy} ensemble (blue points), is shown in blue text at the top right of each panel. For regions where the FMEAN–{Fyyyy} difference is statistically significant at two-tailed p < 0.05 (tested using Welch's *t*-test, using annual mean data as the input), the FMEAN–{Fyyyy} percentage difference (relative to {Fyyyy}) is shown in red text.



Figure 6. Net (shortwave plus longwave) top-of-atmosphere radiative flux perturbation perturbations (RFPRFPs) for (a) simulation FMEAN relative to simulation F0, (b) the {Fyyyy} ensemble relative to simulation F0, and (c) the FMEAN-{Fyyyy} difference. Stippling indicates differences that are statistically significant at a significance level of $\alpha_{fdr} = 0.05$ after controlling the False Discovery Rate (FDR; Benjamini and Hochberg, 1995; Wilks, 2016). The two-tailed *p*-values are generated by Welch's *t*-test, using annual mean data as the input. The approximate *p*-value threshold, p_{fdr} , is written in purple underneath each map. The percentage of the globe (area-weighted) over which negative ("-ve") statistically significant differences occur is written in red text.



Figure 7. Large-scale, convective, and total (large-scale plus convective) annual precipitation differences, relative to simulation F0, for the globe and seven regions. Error bars represent combined standard error. Results for another eight regions are shown in Fig. S8.



-120-100-80 -60 -40 -20 0 20 40 60 80 100 120 Total precipitation difference, mm/yr

Figure 8. Total (large-scale plus convection) annual precipitation differences for (a) simulation FMEAN relative to simulation F0, (b) the {Fyyyy} ensemble relative to simulation F0, and (c) the FMEAN-{Fyyyy} difference. Stippling indicates differences that are statistically significant at a significance level of $\alpha_{fdr} = 0.05$ after controlling the False Discovery Rate (FDR; Benjamini and Hochberg, 1995; Wilks, 2016). The two-tailed *p*-values are generated by Welch's *t*-test, using annual mean data as the input. The approximate *p*-value threshold, p_{fdr} , is written in purple underneath each map. The percentage of the globe (area-weighted) over which negative ("-ve") statistically significant differences occur is written in brown text underneath each map, while the percentage of the globe over which positive ("+ve") statistically significant differences occur is written in green text.



Figure 9. Cloud shortwave top-of-atmosphere radiative flux perturbation (RFP) versus surface organic carbon (OC) accoss) concentration for different months and locations within the Boreal Asia region. The five location–month combinations have been selected according to the procedure described in Fig. S12, based on the following three criteria: (1) within the Boreal Asia region, select the location–month combinations with (2) the largest FMEAN–{Fyyyy} cloud shortwave RFP differences, (3) at least 10° (either longitude or latitude) from one another. The RFPs are relative to simulation F0. The organic carbon across concentrations are given in parts-per-billion by mass (1 ppbm = 10^{-9} kg/kg). Each point represents the results for a different simulation or group of simulations (in the case of "Mean of {Fyyyy}"). Error bars represent combined standard error (see Fig. 3 caption). In (a)–(c), the blue line shows the results of a linear regression fit to $y = a + b \ln x$, calculated from the ten-member {Fyyyy} ensemble (blue points). In both (d) and (e), the regression fit was found to be statistically insignificant at the $\alpha = 0.05$ significance level and is therefore not shown. The corresponding Pearson's product-moment correlation coefficient (r) for $y = a + b \ln x$, calculated from the ten-member {Fyyyy} ensemble, is shown in blue text at the top right of each panel. For regions where the FMEAN–{Fyyyy} difference is statistically significant at two-tailed p < 0.05 (tested using Welch's *t*-test, using annual mean data as the input), the FMEAN–{Fyyyy} percentage difference (relative to {Fyyyy}) is shown in red text.

Table 1. Net (shortwave plus longwave) top-of-atmosphere radiative flux perturbation (RFP) differences for different simulation combinations and regions (see Fig. 1). Area-weighting has been applied. Combined standard errors have been calculated using corrected sample standard deviations, using annual regional mean data as the input – e.g. the standard error for the global {Fyyyy}–F0 difference was calculated using $\sqrt{\frac{s_{Fyyyy}^2}{N_{Fyyyy}}} + \frac{s_{F0}^2}{N_{F0}}}$, where s_{Fyyyy} and s_{F0} are the corrected sample standard deviations, and $N_{\{Fyyyy\}} = 120$ and $N_{F0} = 42$ are the sample sizes. Significance has been tested using Welch's *t*-test, using annual regional mean data as the input: '*' indicates differences that are statistically significant at two-tailed p < 0.05; and '**' indicates differences that are statistically significant at two-tailed p < 0.01. The regions have been ranked according the FMEAN–F0 "conventional" net RFP strength. A horizontal line is used to indicate the seven regions that receive more focus in the Results section.

	Net radiative flux perturbation (RFP), W m $^{-2}$		
Region	FMEAN-F0	{Fyyyy}}-F0	FMEAN-{Fyyyy}
	"conventional"	"revised"	difference
Global (land and ocean)	$-1.26 \pm 0.03^{**}$	$-1.02 \pm 0.03^{**}$	$-0.24 \pm 0.03^{**}$
Boreal North America	$-6.52 \pm 0.18^{**}$	$-4.92 \pm 0.21^{**}$	$-1.60 \pm 0.21^{**}$
Boreal Asia	$-6.23 \pm 0.28^{**}$	$-4.35 \pm 0.17^{**}$	$-1.88 \pm 0.27^{**}$
NH South America	$-3.83 \pm 0.21^{**}$	$-3.67 \pm 0.18^{**}$	-0.16 ± 0.18
Australia and New Zealand	$-3.30 \pm 0.41^{**}$	$-2.10 \pm 0.31^{**}$	$-1.21 \pm 0.36^{**}$
SH Africa	$-3.30 \pm 0.31^{**}$	$-3.21 \pm 0.27 ^{**}$	-0.10 ± 0.26
Western Maritime Continent	$-2.46 \pm 0.24^{**}$	$-1.65 \pm 0.19^{**}$	$-0.81 \pm 0.22^{**}$
SH South America	$-1.78 \pm 0.17^{**}$	$-1.71 \pm 0.16^{**}$	-0.07 ± 0.13
Central America	$-1.60 \pm 0.29 ^{**}$	$-1.14 \pm 0.26 **$	$-0.46 \pm 0.22*$
Eastern Maritime Continent	$-1.45 \pm 0.20^{**}$	$-1.22\pm0.18^{**}$	-0.24 ± 0.17
Temperate North America	$-0.98 \pm 0.32^{**}$	$-1.25 \pm 0.26^{**}$	$+0.27\pm0.28$
Central Asia	$-0.92 \pm 0.28^{**}$	$-0.73 \pm 0.22^{**}$	-0.19 ± 0.22
NH Africa	$-0.79 \pm 0.17^{**}$	$-0.73 \pm 0.13^{**}$	-0.06 ± 0.16
Southeast Asia	$-0.61 \pm 0.23^{**}$	-0.23 ± 0.20	$-0.39 \pm 0.19 *$
Europe	-0.27 ± 0.33	-0.14 ± 0.24	-0.13 ± 0.28
Middle East	-0.14 ± 0.12	-0.08 ± 0.10	-0.07 ± 0.10

	Cloud shortwave radiative flux perturbation (RFP), $W m^{-2}$			
Region	FMEAN-F0	{Fyyyy}}-F0	FMEAN-{Fyyyy}	
	"conventional"	"revised"	difference	
Global (land and ocean)	$-1.11 \pm 0.03 **$	$-0.87 \pm 0.03^{**}$	$-0.24 \pm 0.03^{**}$	
Boreal North America	$-5.88 \pm 0.18^{**}$	$-4.40 \pm 0.18^{**}$	$-1.47 \pm 0.20^{**}$	
Boreal Asia	$-5.76 \pm 0.21 ^{**}$	$-3.91 \pm 0.17^{**}$	$-1.85 \pm 0.21^{**}$	
NH South America	$-1.08 \pm 0.41^{**}$	$-1.16 \pm 0.32^{**}$	$+0.08\pm0.33$	
Australia and New Zealand	$-2.92 \pm 0.67 ^{**}$	$-1.44 \pm 0.49^{**}$	$-1.48 \pm 0.59 *$	
SH Africa	$-2.01 \pm 0.46^{**}$	$-1.81 \pm 0.40^{**}$	-0.20 ± 0.39	
Western Maritime Continent	-0.62 ± 0.43	-0.00 ± 0.36	-0.62 ± 0.36	
SH South America	$-0.82 \pm 0.24^{**}$	$-0.84 \pm 0.21^{**}$	$+0.02\pm0.18$	
Central America	$-1.75 \pm 0.47 ^{**}$	$-1.08 \pm 0.41 *$	-0.68 ± 0.37	
Eastern Maritime Continent	-0.62 ± 0.37	$-0.72 \pm 0.33 *$	$+0.09\pm0.30$	
Temperate North America	$-1.05 \pm 0.37^{**}$	$-1.30 \pm 0.31^{**}$	$+0.26\pm0.31$	
Central Asia	-0.30 ± 0.28	-0.22 ± 0.23	-0.08 ± 0.23	
NH Africa	-0.28 ± 0.35	-0.17 ± 0.28	-0.11 ± 0.28	
Southeast Asia	-0.20 ± 0.44	$+0.41\pm0.37$	-0.61 ± 0.37	
Europe	$+0.48\pm0.41$	$+0.10\pm0.30$	$+0.38\pm0.36$	
Middle East	-0.37 ± 0.30	-0.12 ± 0.21	-0.25 ± 0.25	

Table 2. Cloud shortwave top-of-atmosphere radiative flux perturbation (RFP) differences for different simulation combinations and regions. Combined standard errors have been calculated. Significance has been tested using Welch's *t*-test: '*' indicates differences that are statistically significant at two-tailed p < 0.05; and '**' indicates differences that are statistically significant at two-tailed p < 0.05;

	Total precipitation difference, mm yr ^{-1}		
Region	FMEAN-F0	{Fyyyy}}-F0	FMEAN-{Fyyyy}
	"conventional"	"revised"	difference
Global (land and ocean)	$-2.1 \pm 0.4^{**}$	$-2.0 \pm 0.4^{**}$	-0.1 ± 0.4
Boreal North America	$-11.8 \pm 3.6^{**}$	$-14.1 \pm 3.4^{**}$	$+2.3\pm2.7$
Boreal Asia	$-22.7 \pm 3.5^{**}$	$-18.5 \pm 2.9 {**}$	-4.2 ± 3.0
NH South America	$-102.1 \pm 15.1 ^{**}$	$-85.0 \pm 12.1 ^{**}$	-17.1 ± 12.5
Australia and New Zealand	$-34.8\pm15.1*$	$-49.2 \pm 11.3^{**}$	$+14.4\pm13.2$
SH Africa	$-38.5 \pm 8.2^{**}$	$-50.5 \pm 6.5^{**}$	$+12.0\pm7.2$
Western Maritime Continent	-39.1 ± 21.7	$-58.5 \pm 18.4^{**}$	$+19.4\pm18.2$
SH South America	$-33.9 \pm 5.2^{**}$	$-26.5 \pm 4.8^{**}$	-7.4 ± 4.1
Central America	$+19.5\pm10.6$	$+12.0\pm9.1$	$+7.5\pm8.4$
Eastern Maritime Continent	-6.5 ± 16.4	-2.6 ± 14.6	-3.9 ± 13.0
Temperate North America	-2.5 ± 8.7	$+7.7\pm7.7$	-10.1 ± 6.5
Central Asia	-6.3 ± 5.1	-2.9 ± 4.2	-3.4 ± 4.1
NH Africa	$-31.9 \pm 10.0^{**}$	$-33.1 \pm 7.9^{**}$	$+1.2\pm8.0$
Southeast Asia	-12.5 ± 14.0	-18.3 ± 10.9	$+5.8\pm11.9$
Europe	-6.5 ± 5.7	-5.0 ± 4.4	-1.5 ± 4.7
Middle East	$+8.4\pm5.5$	$+3.0\pm4.0$	$+5.3\pm4.6$

Table 3. Total (large-scale plus convective) annual precipitation differences for different simulation combinations and regions. Combined standard errors have been calculated. Significance has been tested using Welch's *t*-test: '*' indicates differences that are statistically significant at two-tailed p < 0.05; and '**' indicates differences that are statistically significant at two-tailed p < 0.01.