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Review comment on acp-2021-862

Pasquale Sellitto (Referee)

Referee comment on "Important role of stratospheric injection height for the distribution and radiative forcing of smoke aerosol from the 2019/2020 Australian wildfires" by Bernd Heinold et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-862-RC2, 2021

Review of the manuscript "Important role of stratospheric injection height for the distribution and radiative forcing of smoke aerosol from the 2019/2020 Australian wildfires", Heinold et al.

The manuscript "Important role of stratospheric injection height for the distribution and radiative forcing of smoke aerosol from the 2019/2020 Australian wildfires", Heinold et al., presents and discusses the aerosol spatiotemporal distribution and radiative forcing (RF) of ECHAM6.3-HAM2.3 model simulations of the pyro-convective paroxysmal phase of the record-breaking Australian bushfires in the fire season 2019/2020. The importance of representing pyro-convection and UTLS injection of smoke into models is discussed based on sensitivity analyses. The manuscript provides a new estimation of the RF and radiative heating of this Australian fire event, which is quite a hot topic. It certainly falls within the scopes of ACP and is very interesting for the aerosol/climate community, and is potentially suitable for publication. By the way, I have found that there are several aspects – some of them very important – that must be clarified before I can recommend publication of the manuscript. Thus, I kindly ask the Authors to reply to the following major and specific comments and to provide an amended version of the manuscript, that I would be glad to re-read when ready.

Sincerely,

Pasquale Sellitto

Major Comments (MC#):

MC1) The temporal trend of the AOT obtained with ECHAM are contradictory with previously published observations. The trend shown in Fig. 2 (maximum AOT in January and then decreasing in February and March) is not consistent with, e.g., the stratospheric AOT from SAGE-III in Khaykin et al., 2020 (e.g. its Fig. 3) – paper which is cited in the present manuscript. In particular, Khaykin et al. show an increasing AOT from January to February, with a maximum in February. The Authors should discuss this marked difference. Is an increase in stratospheric-only AOT in February also present in ECHAM modelling (in Fig. 2 there's a total column AOT, I guess)? Is the possible coating of black carbon (BC) particles and evolving aerosol mixing state not well represented in ECHAM – which is quite a common feature in aerosol/climate models, as extensively discussed by Brown et al. (https://www.nature.com/articles/s41467-020-20482-9)?

MC2) The Authors obtain a relatively large and positive RF at top of atmosphere (TOA), which is basically in contradiction with all estimations available at the moment (Khaykin et al., Yu et al., Hirsch and Koren, papers that are cited in the manuscript). The observational (Hirsch and Koren) and hybrid observational/modelling (Khaykin et al.) estimations agree on a relatively large negative TOA RF. The Authors interpret this disagreement with respect to these previous estimations as the result of all-sky calculations and the high surface reflectivity in the present manuscript, which, I agree, can partially explain that. By the way, the RF is very sensitive to optical properties of the aerosol layer (in particular, the absorption properties of the layer and its angular distribution of scattering, see discussion in SC56 and other SCs). In addition, also Yu et al. obtain a negative TOA RF but at all-sky conditions. This should be discussed more thoroughly in the text (see suggestions in several of the following specific comments) and the different statements supporting a positive RF must be smoothed a bit.

Specific Comments (SC#):

SC1) P1 L24-25: "Global…wildfires": I would not call this "uncertainties" but rather "incomplete representation", like said in the following line, or something similar

SC2) P1 L27: Its more "observation-based input to the simulations" than "observation-based approach"

SC3) P1 L28: please add "Based on our simulations,…" before "The 2019-2020 Australian fires caused…"

SC4) P1 L32: "While at surface,…" is an awkward way to start a sentence, please rephrase.

SC5) P1 L34: "deep wildfire plumes…", "deep" is a bit too generic here: do you mean "with high altitude injection" or just "extreme"?

SC6) P2 L5: “life”: do you mean “wildlife”?

SC7) P2 L6-7: “In addition…whether”, this is an awkward sentence, please rephrase

SC8) P2 L14-17: please add SAGE-III and TROPOMI observations (as shown by Khaykin et al., 2020) into the discussion of evidences of the fire with satellites

SC9) P2 L18-19: The first estimation of the radiative forcing of Australian fires was provided by Khaykin et al., 2020, with hybrid observations/modelling approaches. Please mention this manuscript and the method.

SC10) P3 L1-3: please break this very long sentence

SC11) P3 L5-6: Radiative-heating-induced self-rising can occur in fires but this is not the norm, so please smooth this sentence

SC12) P3 L9: “effects”: Please specify which specific effects

SC13) P3 L10-11 “which is considered to be the strongest warming short-lived radiative forcing agent.”: please add a reference for this statement

SC14) P3 L11-12: “In addition…emitted”: also precursors of secondary organic aerosols can be emitted, please mention

SC15) P3 L12: “…radiative properties…”: you might mean “optical properties”

SC16) P3 L13-14: “…as well…altitude.”: not clear, what do you mean?

SC17) P3 L16: “…the recent accumulation of extreme wildfires..”: you mean “aggregated effect”?

SC18) P3 L21: please remove “more”

SC19) P3 L27: not sure that “to capture” is the right verb here

SC20) P4 L18: “height profiles”: do you mean “vertical profiles”?

SC21) P5 L10-12: which altitudes for these vertical layers in the model?

SC22) P5 L14: “47 levels”: which approximate vertical resolution?

SC23) P5 L18-19: “AOT and vertical profiles of extinction”: This looks redundant as the AOT is just the vertical integration of the aerosol extinction.

SC24) P5 L28: “Since no direct information was available on the actual pyroconvective injection heights…”: This is not completely true as Khaykin et al. give an upper bound for the injection altitude at circa 17 km using CALIOP, and it is also shown at approximately this altitude for Hirsch and Koren, 2021 (as you mention later in the text). Please correct the sentence.

SC25) P5 L37-38: The results of Hirsch and Koren (2021), seems more to show that smoke injection is injected at altitudes >16 km. Why do you say “14 km”?

SC26) P5 L38-39: “In addition…the original biomass burning injection…”: The original injection is what is described above (P5 L10-12)? Please clarify in the text.

SC27) Table 1: in the NoEmiss lines, when you say “1 April” you mean “4 January”?

SC28) More in general on the NoEmiss scenario: In the NoEmiss scenario, how are the previous emissions from Australian fires (i.e. the Australian fire season prior to 29/12/19) considered?

SC29) P6 L11: please suppress “significantly”

SC30) P6 L12: The long-term mean ("which implies…") is not shown in Fig. 1b, so please explain how it is calculated and rephrase the sentence?

SC31) P6 L13: "AOT": please mention wavelength

SC32) P6 L14: "compared…half a year": this is not clear at all, please rephrase

SC33) P6 L15: "…relative to 2019": You mean wrt the monthly mean AOT for January 2019? Please clarify in the text

SC34) Figure 2: wrt MC1, the trend (maximum AOT in January and then decreasing) is not consistent with observations, e.g. the SAOD from SAGE-III in Khaykin et al., 2020 (Fig. 3). Please explain why.

SC35) P7 L8: "The emissions…are reproduced…": The emissions are not "reproduced" by ECHAM but are "an input" to ECHAM: please rephrase

SC36) P7 L 10: "…provide an insight…due to wildfire smoke…": If NoEmis has only the smoke emissions of 29-31 December and 4 January switched off, then this comparison does not provide "the AOT distribution due to wildfire smoke" but rather "the AOT distribution due to pyro-convective events of 29-31/12 and 04/01". Please verify and possibly correct.

SC37) P7 L16: "AOT differences": Differences with respect to what?

SC38) P8 L15: "…is clearly better": Is it "clearly" better? Not to my eyes: the comparisons with different injection altitudes looks quite similar to me. This is not so surprising because the effect of the fires on the column AOT is not as strong as the one at selected UTLS altitudes (as visible in Fig. 4). Please, based on that, smooth these statements, and reconsider this discussion.

SC39) P8 L16-17: please suppress "using…above" (this is already clear from scenarios descriptions above, so is redundant)

SC40) P8 L18-19: "…indicating that the modeled effect…stratospheric smoke": This statement is not true because the solar absorption depends not only on the aerosol load but also on the optical properties of the aerosols - and then, for your estimations, on the assumptions made in the model on composition and atmospheric evolution of the smoke plume. Please smooth the statement.

SC41) P8 L19: “is larger” --> “is slightly larger”

SC42) P8 L20: “…correlation is also lower”: From Tab. 2 it looks like BASE R is quite comparable wrt TP(+-1) and the others setups.

SC43) P8 L25-26: “which is also consistent with the CALIPSO satellite lidar observations”: Which CALIPSO observations? (they're neither in Fig. 4 nor 5)

SC44) P8 L28: “reflect” is not a good choice here as a term, as it also has an optical meaning: please change term

SC45) P8 L28: “These remarkable values…”: what do you mean?

SC46) P9 L9-10: “again…apparent”: You refer to Fig. 4 I guess: please mention this in the text.

SC47) P9 L12-13: “The model results…smoke layer”: This is not peculiar of the simulations but only empirically visible in Fig. 4 and 5 (for model as well as in the lidar observations): please rephrase.

SC48) Figure 4: Please spell "coeff." and not "cf."; please use “km-1” as aerosol extinction units

SC49) Figure 5: here as well, please use km-1 as aerosol extinction units. Also, for the sake of visual clarity of the comparison, why not suppressing the pressure vertical axis and just put the height on the left, for your simulations results?

SC50) P10 L10: “For other model scenarios”: Please specify which scenarios.

SC51): Your estimation of the heating rate: The heating rate is very sensitive to the aerosol optical properties, please mention this in the text as a reason for large uncertainties in your estimations. Also: are these shortwave-only or shortwave+longwave heating rates?

SC52) Figure 6: are these “monthly averages” for January? Please mention this in the caption.

SC53) Figure 7: Would it be possible to have a altitude vertical axis as well?

SC54) P12 L6: “greenhouse forcing”: You mean “greenhouse gases forcing”? Black carbon also can produce greenhouse effect (but it's particle, so better to be more specific)

SC55) Your RF estimations: same question as for the heating rates: are these estimation for SW-only or LW+SW?

SC56) With reference to MC2: the RF of aerosols depends very strongly on the optical properties of the aerosol layer, which in turns, and this is very important for the complex smoke emissions by fires, depend on the atmospheric evolution of the plumes. In particular, the aerosol RF depends quite strongly on both absorption properties (summarised by SSA) and the angular distribution of scattering (the phase function, summarised by the asymetry parameter). Examples of such variability of RF on these two integral optical parameters (for volcanic aerosols, but it applies more in general) can be found here;

- Sellitto et al., 2020 (https://www.nature.com/articles/s41598-020-71635-1), see their Fig. 5

- Kloss et al., 2021 (https://acp.copernicus.org/articles/21/535/2021/), see their Fig. 9

The situation can be even more complex for fire emissions, where the optical properties of the emitted and secondary formed aerosols, as well as their evolution in a complex environment of high humidity, many gaseous emission and locally high temperatures. Thus, your estimation depends strongly on the somewhat arbitrary assumptions of your simulations. This must be critically discussed in the text.

SC57) P12 L13-17: Yu et al. (2021) also obtain a slightly negative TOA RF using a model and theirs are all-sky estimations. Please notice that their estimations are SW+LW (personal communication). As a matter of fact, your RF being positive is quite in contradiction with all previous observations, simulations and hybrid estimations of the TOA RF for Australian fires 2019/20, clear- and all-sky, and this must be mentioned in the text.

SC58) P12 L20-21: This is another reference that can be helpful in comparing your results with volcanic eruptions: https://www.nature.com/articles/ncomms8692?proof=t

SC59) P12 L22-24: No mention to the stratospheric vortex driven by rapid vertical transport + plume heating seen for this fire event? (Khaykin et al., 2020; Kablick et al., 2020)

SC60) P13 L7-8: "uncertainties in AOT; in particular…single scattering albedo…": This should be rephrased: the AOT representation and single scattering albedo are only in part inter-dependent. In addition, the angular scattering properties of the aerosol layer is also (or, at some conditions, even more) important for RF estimations (see Sellitto et al., 2020; Kloss et al. 2021, mentioned at SC56), and this should be mentioned.

SC61) P13 L9: "SSA lies between 0.82-0.85…". Single scattering albedo (and asymmetry parameter of soot aerosols, see SC60) may be significantly affected by their mixing states, and coating of BC. SSA can be significantly larger (up to ~0.95 at 550 nm) if BC is coated by aqueous secondary aerosols (organic or sulphate) - e.g. https://www.nature.com/articles/s41467-020-20482-9 . Also, and importantly, it looks like smoke aerosols are too absorbing in models due to a generally incomplete representation of the aerosol mixing state for biomass burning aerosols: https://www.nature.com/articles/s41467-020-20482-9 . This must be mentioned and discussed in the text, as this might be a large source of uncertainties for your RF estimations (as well as heating rates estimations and, even more at the basis, AOT fields).

SC62) P13 L11-12: "For the 2019-2020 Australian fires, first inversion results even point towards SSA values just below 0.8, as calculated using the method of Veselovskii et al. (2002).": more details are called here.

SC63) P13 L14-15: "Thus, together with the low bias of the modeled smoke AOT, we argue that our results illustrate a conservative estimate for the positive TOA forcing of this event.": This statement is definitely too strong, due to the large uncertainties discussed in my previous comments and to the fact that all previous RF estimations (Khaykin et al., Hirsch and Koren, Yu et al.) indicate rather a negative RF at TOA.

SC64) Table 3. As already said for the AOT trend, in Khaykin et al. the strongest RF is in February and here is in January. As already mentioned in previous comments, this might be linked to an insufficient representation of secondary aerosols formation in your model and mixing state. Please mention and comment in the text.

SC65) P14 L15: “While this is appropriate…pyroCb clouds”: Thus, why not using the satellite observations of the plumes themselves as proposed by Kloss et al. (2021, Fig. A1-2), see also SC56?

SC66) P14 L20-21: “Consequently, aerosol-climate models underestimate the wildfire aerosol impacts on the energy balance, as the vertical location of the smoke relative to clouds is fundamental to its radiative impact.”: This might be true for pyro-convective fires while it is demonstrated that the biomass burning RF is overestimated in general, in models, again available at the following link:
https://www.nature.com/articles/s41467-020-20482-9

SC67) P14 L29: please put references in chronological order.