Authors' Response to Reviewers' Comments

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Title: Important role of stratospheric injection height for the distribution and radiative forcing of smoke aerosol from the 2019/2020 Australian wildfires

Authors: Bernd Heinold, Holger Baars, Boris Barja, Matthew Christensen, Anne Kubin, Kevin Ohneiser, Kerstin Schepanski, Nick Schutgens, Fabian Senf, Roland Schrödner, Diego Villanueva, and Ina Tegen

Anonymous Referee #1 (RC1), 2nd round

Compared to the first version the paper improved a lot. But there are still some minor issues to be corrected.

We sincerely thank the anonymous reviewer for his time and constructive comments also in the second round. We hope that we have responded satisfactorily to all the questions and that any remaining unclear points could be clarified.

Specific comments

(The line numbers refer to the version with change tracking.)

Page 4, line 18: I wonder why there is no routine similar to the one for the lidars (page 6) for output sampling at the AERONET stations. It is not necessary to repeat calculations.
The standard output of ECHAM-HAM are AOD values at 550 nm, 865 nm and 55,555 nm. 550 nm is a common wavelength for satellite products, but also some AERONET stations provide values for this wavelength. The lidar simulator, on the other hand, is a relatively new implementation, specifically for CALIOP comparisons, and therefore directly provides the optical parameters at 532 nm. Since AERONET observations are often used for evaluation, it is certainly useful to extend the default output in a future model version. Nevertheless, the conversion using an Angstrom parameter is a suitable as well as common method.

Page 5, line 22ff: Is this also the case for the lower stratosphere? The statement appears to be not consistent to some results shown in Figure 7. Do the 44% refer to the total column or some layers?
Yes, indeed, this statement also applies to the lower stratosphere. Thank you for pointing this out, we have added it accordingly and now also include a reference to Liu et al (2019) who evaluated the performance of the CALIOP V4 CAD algorithm in the troposphere and stratosphere: "While the V4 CAD can distinguish aerosols and clouds for stratospheric layers, uncertainties tend to increase as the altitude increases. This increasing uncertainty derives from the fact that the very low aerosol occurrence frequency at high altitudes does not provide a statistically significant sample size to constrain the [probability density functions] PDFs [...]".
However, we agree that in this particular case the discrepancy between modeled extinction coefficients and those observed by CALIOP decreases again in the uppermost part of the

profile (>23 km). Although it seems that there the CALIOP profiles for all years converge to some background value.
Here, "globally" is meant literally. The 44% value is a vertical mean across the entire globe (Watson-Parris et al., 2018).

Additional reference:
Liu, Z., Kar, J., Zeng, S., Tackett, J., Vaughan, M., Avery, M., Pelon, J., Getzewich, B., Lee, K.-P., Magill, B., Omar, A., Lucker, P., Trepte, C., and Winker, D.: Discriminating between clouds and aerosols in the CALIOP version 4.1 data products, Atmos. Meas. Tech., 12, 703–734, https://doi.org/10.5194/amt-12-703-2019, 2019.

Section 2.4: The dual call of the radiation code for calculation of forcing and heating is not mentioned here in contrast to the reply to the referees. At least it is mentioned on page 13.
That's right. Eventually, we decided that the description is best placed where the instantaneous forcing is actually discussed. However, additional details on the radiation scheme in ECHAM-HAM, including the dual call for diagnosing the instantaneous aerosol radiative forcing, are now given in Section 2.4.

Page 6, line 30, abstract and introduction: Is the agreement of mass by chance or are the data taken from the references?
No. As we describe in this line and several times in the text, the strength of the biomass burning emissions in the model is based exclusively on the GFAS product and were not further adjusted for the 2019/2020 Australian fires. An adjustment was only applied with respect to the injection height as described in Section 2.4.2. This shows the quality of the GFAS emission inventory. For clarification we add: "These values agree well with the previously mentioned estimates by Peterson et al. (2021) and are kept unchanged throughout the sensitivity experiments."

Page 6, line 36f and Table 1: Is there a gap between the layers with smoke in the scenarios TP1_8020 and TP1_5050 or do you mean the layer containing the tropopause instead the one below the tropopause? More precise please. I suppose you mean "all wildfire smoke" in TP+1, if yes please include in Table 1.
There is of course no gap between layers with smoke in the scenarios TP1_8020 and TP1_5050. To be more precise the sentence was modified as follows "TP1_8020: as TP+1 but only 80% of the emitted smoke injected above the tropopause and 20% distributed between tropopause level and surface."
In the sentence above we clearly say that only the emission heights over Southeastern Australia were modified.

Page 10, line 23: Also scenario NoEmiss, right?
That's true. Here, the misrepresentation of the stratospheric injection height has the same effect as if the fire emissions of the pyroCb days had not been considered at all. We add: "…just like in the NoEmiss case."

Page 11, line 2: Mention Fig. 6.
Done.

Figure 6: "DRF" in the caption is not defined, please spell out somewhere.

Done.

Page 12, line 15ff: It might be good to mention a typical range of the lidar ratio used by the simulator to create Fig.7.
In order to produce Fig. 7, which shows extinction profiles, no assumptions about the lidar ratio are necessary on the model side (lidar simulator). As a matter of interest and for evaluation of the smoke optical properties in the model, Section 3.2 now includes a statement as follows: "Accordingly, Ohneiser et al. (2020, 2022) report lidar ratios at 532 nm in the range of 75 – 112 sr (average 97 sr) for their Punta Arenas observations, also indicating strongly absorbing aerosol. The lidar simulator of the model, in comparison, provides slightly lower lidar ratios at 532 nm between 70 and 100 sr for the stratospheric smoke layer."

Page 14, line 14: Do you see similar features in your simulations? You should include a sentence on that even if it is modified by nudging.
Yes, such a vortex is also seen in the 50-hPa wind fields of the ECHAM6.3-HAM2.3 simulations. However, the analysis of the atmospheric dynamic effects is the subject of a follow-up study. A sentence to this effect was added.

Technical corrections

Page 3, line 5: Correct grammar.
Done.

Page 4, line 15, line 30 and Page 6, line 16f: Are the calculations of optical properties really done for the three wavelengths 532, 550 and 553nm or extrapolated like stated on page 4?
The wavelength of 553 nm for the lidar simulator is a typo. The calculation of the 550-nm optical thickness using the Angstrom parameter is only done for the AERONET observations. The lidar simulator provides aerosol extinction profiles at 532 nm that can be used directly for comparisons with CALIOP and the ground-based lidar at Punta Arenas.
Thank you for pointing this out.

Page 6, line 41: "by" instead of "due to"?
"By" does not express the explanation that is meant here. Still replaced by "because of".

Table 1: I suppose you mean "the layer around 14km".
Corrected.

Figure 2: Please check the color code. I don't see the high values stated in the text in the figure.
Thank you for bringing this inconsistency to our attention again. There is a misunderstanding here that we overlooked in the last round of reviews. In the text, monthly means of the AOT of the respective scenarios (BASE, TP+1) are given in absolute values. In contrast, Figure 2 shows the differences in mean AOT for January to March 2020 between the model scenarios BASE (2a-c) and TP+1 (2d-f) against the NoEmiss results, respectively. Therefore, they exclusively show the contribution of the smoke-only AOT for the case where no smoke injection by pyroconvection is prescribed in the model (Fig. 2a-c) or for smoke injection into the model layer above the tropopause on the AOT for pyroCb days 29-31 December 2019 and 4 January 2020 in southeastern Australia (Fig. 2d-f).

In order to clarify, the sentence was modified as follows: "While the monthly mean smoke AOT is simulated in absolute values as high as 0.26 and 0.22 for January just downwind of the fire region in Southeast Australia for the BASE and TP+1 experiments, respectively,…"

Page 20, line 9: You should include a doi for that online reference.
We are not sure which of two references is meant, but both Heinold et al. and Hersbach et al. include the doi in a format suggested by Zenodo and RMetSoc, respectively.

Anonymous Referee #2 (RC2), 2nd round

I would like to sincerely thank the Authors for their work and revisions of their very interesting manuscript. I can see that most of my Specific Comments have been tackled and I'm very satisfied of the way this was addressed by the Authors. By the way, my main concerns have been mostly skipped, namely the model's representation of secondary aerosols (Major Comment 1 and a few Specific Comments) and the role of evolving optical properties on the radiative impacts (Major Comment 2 and a few Specific Comments). To be clear, I honestly think that the paper should be published soon, as it deals with an important topic, but I would in any case require that these two points are better addressed before publication. This basically means: 1) smoothing many very strong statements (e.g. about the "certainly positive radiative forcing of the plume" or the "perfect optical properties simulated by the model" or the "secondary aerosols which are surely not formed") and 2) adding a deeper and comprehensive discussion on the two issues. I strongly suggest the Authors to make this effort. A few more details are in the following.

Thank you for the interesting work and discussion,
Pasquale Sellitto

We also thank you for your availability for the second round of review and the critical discussion and suggestions that helped to further improve this work.

As outlined in the response to the comments below, we have tried to address your comments as best as we could. Specifically, we have included secondarily formed particles as a possible reason for the underestimation of the modeled smoke aerosol. In general, we point more strongly to the uncertainties in the optical properties in our model results. And we discuss the possibility that dilution impacts on smoke aging could have led to temporal and spatial variability in smoke optical properties, which cause further uncertainties in the model estimate of the direct radiative forcing of the 2019-2020 Australian wildfire smoke.

Major comments (#MC)

MC1) In Khaykin et al., 2020, it was supposed that one reason for the increasing trend of SAOD could be saturation of the OMPS-LP detector. This was the proposed reason at the time of publication of that paper (which I personally co-authored). By the way, following reflections and analyses since that publication, while keeping this as a possible explanation of this time evolution of the SAOD, led to other possible explanations:

1) the formation of secondary aerosol and aerosol mixing, (which, on the other hand is a known issue in terms of representation of biomass burning plumes in models like the one used in this work (Brown et al., 2020)) and 2) plume's progressive lofting due to in-plume radiative heating. These aspects are further addressed and discussed in a recent preprint publication (https://egusphere.copernicus.org/preprints/egusphere-2022-42/) that I suggest checking. As a matter of fact, these two aspects cannot be excluded, and this certainly needs further discussion in the paper. Why are you so deterministic in this statement "Secondary aerosol formation appears unlikely to be the explanation considering the required amount of smoke."? The progressively less absorbing aerosol properties seem to actually point at a progressive secondary aerosol formation and brown-carbonification of black carbon emissions.

Thank you for bringing this follow-up paper to our attention. In particular, the sensitivity study is very interesting with respect to the effects of plume aging and mixing on the evolution of smoke optical properties and ultimately the radiative forcing. We picked this up now, where it fit.
In reply to your comment, we would first like to emphasize that we already pointed out several times in the last paper version the overestimation of smoke aerosol absorption in this model and climate models in general, as noted by Brown et al. (2020), resulting from an inadequate representation of the plume aging and particle mixing state. And we already discussed (page 10, line 17/18 of the new change-tracked version) that "The underestimation of the fire aerosol loading in all configurations […] is partly due to missing secondary aerosol formed in the plume, which is not considered by the model." Just as you say, the heating-induced lofting affects the radiative influence of the smoke plume. This effect is also reproduced by the model as we show in Section 3.1.
So, we are in fact aware of the uncertainties due to secondary aerosol formation and the evolution of fire aerosol properties. On the other hand, we cannot ignore the good agreement of the model results with the lidar measurements and retrievals of optical particle properties from southern Chile, which apply very well at least for this particular pyroconvective case. These lidar data are the best constraints of aerosol optical properties available for this Southern Hemisphere wildfire event.
In contrast, the problem of the delayed SAOD peak in February, which could be due to saturation of the OMPS LP detector, does not seem to be resolved in your study (https://egusphere.copernicus.org/preprints/egusphere-2022-42/): „… we cannot exclude any of the above hypotheses, we are inclined to consider the aging of the plume as an important factor at play...". One of the EGUsphere reviewers also points out this possible weakness of the instrument. We cannot judge this, but tend to believe the measurements from AERONET and the lidar observations at Punta Arenas, which point to a peak already in January 2020.
Nevertheless, we think you raise an important point here regarding the temporal and spatial evolution of the plume optical properties, especially with respect to secondary aerosol formation and mixing. So, we mention secondarily formed particles as a possible reason for the model underestimation of fire smoke more often in the text, and now also refer more strongly to the uncertainties in the optical properties due to secondary fire aerosol and aging that might have played a more important role on the larger scale in Section 3.2 (further details below).

The LiDAR SSA inversions (by the way, please discuss briefly the inversion methodology in the Data and Methods section) that are now presented in the manuscript cannot actually demonstrate the fact that there is no secondary aerosol formation, and then progressive larger SSA and lesser absorption from plume's aerosol, because: 1) if I got it right, these are measurements for January 2020 only, too early to have a marked secondary aerosol formed and a clear signature in the plume's aerosols optical properties; 2) LiDAR inversions of optical properties have usually significant uncertainties and SSA variability is small (from 0.75-0.80, for black carbon; 0.85-0.90, for brown carbon, only 10-15% increase on SSA but sufficient to switch the RF sign from positive to negative). Also, the statement, P15 L3-4: "Ohneiser et al. (2022) show an SSA of 0.79 for the rotating smoke disk on 26 January above Punta Arenas in Chile, which is also representative for other smoke measurements" is not true: the vortex plume is an isolated patch of fresh smoke aerosols, isolated from the environment and absolutely not representative, in terms of optical properties, of the overall large-scale plume: please correct. It is necessary that you add a substantial discussion on these aspects in your manuscript and be more cautious in this respect in the Abstract and Conclusions.

It was only in January that the smoke AOT was high enough for Ohneiser et al. (2022) to perform the multi-wavelength inversion to derive the single scattering albedo from the Polly lidar measurements in Punta Arenas/Chile. At this time, the SSA was about 0.80 with an error of about 0.05. As Ohneiser et al. (2022) show (see their Figure 8b), the 532 nm lidar ratio remained high well beyond the end of January, indicating strongly absorbing aerosol, which is why low SSA values can be assumed to continue occurring in February and March. Regarding the formation of secondary aerosol and aerosol mixing, we think that the condensation of gases onto smoke particles usually lasts on the order of 2 days in the troposphere, while it will certainly continue for longer durations in the stratosphere. The observed decline in the lidar depolarization ratio at Punta Arenas marks the completion of the aging of the smoke aerosol by mid-February at the latest (see Fig. 8c in Ohneiser et al., 2022), with low depolarization values indicating aged round particles after this time. After the condensation phase, only coagulation is considered to change the smoke size distribution, and coagulation is not as effective in the stratosphere as in tropospheric layers.

The statement that the smoke optical properties obtained for January 2020 are also representative for other mid and high latitudes in the Southern Hemisphere is actually a conclusion by Ohneiser et al. (2022) that we refer to. We agree that the vortex structure most likely preserved the enclosed smoke plume from being rapidly diluted within the environment. However, there is probably no reason why the smoke trapped in the vortex should not be subject to aging or SOA formation, with volatile precursors being co-emitted with BC and coagulation also taking place. On the other hand, it cannot be excluded that dilution impacts on the aging of the plume may have led to a different evolution of the smoke optical properties between the vortex and larger scale environment, which were not shown by the lidar measurements.

Accordingly, the discussion in Section 3.2 was revised and extended, including more references to the findings by Ohneiser et al. (2022). Furthermore, we included secondarily formed particles as a possible reason for the underestimation of the modeled smoke aerosol, for example in Section 3.1. We also point more strongly to potential uncertainties in the optical properties in our model results. In addition, the possibility is discussed, that dilution *effects at the plume edges in contrast to the core could have influenced the spatio-temporal evolution and variability of the smoke optical properties, which may not have been captured by the local lidar observations but may still be a source of uncertainty in model estimates of radiative forcing.*

The lidar inversions of single scattering albedo (SSA) were not performed in this study but are actually part of the work by Ohneiser et al. (2022), which we refer to in the discussion in Section 3.2. Going into details would be beyond the scope of this paper. Nevertheless, it is certainly a good idea to refer to the inversion method by Veselovskii et al. (2002) and the study by Ohneiser et al. (2022) already in the Data and Methods section.

MC2) First, please accept my apologies for my mistake: Yu et al. (2021) is also clear-sky RF estimations and not full-sky as I stated in the previous review round. By the way, it is undoubtedly true that optical properties of the aerosol layer have dramatic impacts on the radiative forcing of a given aerosol layer, which is even more important for biomass burning highly evolving plumes. The LiDAR observations and all discussion in the revised manuscript only deal with the young plume (in January), while the optical properties of biomass burning aerosols should evolve (e.g. SSA and g) at longer timescales and mostly visible, in case, starting from February-March. Thus, it cannot be accepted what you state: “This analysis, however, further supports that the optical properties of the fire aerosol are reasonably realistic for this case, and thus the positive instantaneous solar radiative forcing at TOA”. Again, yours is a valuable work and should be published but the limits of the model assumptions must be discussed, and the fact that the magnitude and sign of the radiative forcing depend on the modelled aerosol optical properties must be clearly stated. The strict certainty of a positive radiative forcing, that you suggest, should be avoided throughout the whole text. In the meanwhile, a preprint with sensitivity analyses of radiative forcing for this event to optical properties has been published (see MC1); please exploit, in your paper, these sensitivity analyses in the discussion of this aspect.

As described above, the Ohneiser et al. (2022) observations support the assumption that the aging process of the Australian fire plume was completed by the end of January, but that the smoke particles continued to be absorbing as indicated by persistently high lidar ratios. Since the previous paper version apparently gave the wrong impression that we were referring only to the lidar observations in January, we expanded Section 3.2 for clarification, as already replied to MC#1.

We were happy to include your study in the introduction and discussion. And we point more strongly to the uncertainties in the optical properties in our model results, as well as the possibility that this secondary fire aerosol and aging have had a larger effect on the larger scale. Please note in this regard our response to MC# 1 and, in particular, the modifications to the text in Section 3.2 and Section 4 ‘Implications and perspectives’.