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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Implementing Microscopic Charcoal Particles Into a Global Aerosol-Climate Model" *by* Anina Gilgen et al.

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We thank the reviewer for his helpful and valuable comments. Please note that we found a bug in the charcoal code, which affected the results. The following main points have changed:

- For the calibration data set, the Pearson correlations coefficients show now nearly no difference for different parameter sets (before: ranging between 0.21 and 0.32; now: ranging between 0.21 and 0.23).
- We chose a different parameter set with the new simulations, which has the highest variability (old parameter set: $remi2.5, rthr4.9, dens0.6$; new parameter set: $remi5, rthr4.9, dens0.6$).

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- The emission factor that is in best agreement with the observations is now 250 (before: 40). This is both due to the error in the code and due to the different parameter set that we chose. We mention now in the text the effect of the parameter set on the estimated emission factor.

Furthermore, based on your comments, we decided to create a Supplementary Material, which now includes part of the manuscript and the Appendix. In the following, the reviewer's comments are shown in italic font and our answers in bold font.

The authors present the technical implementation of charcoal particles into a global climate model, calibrate emissions using a test dataset, and then evaluate their initial model performance against a global set of observations. Understanding microscopic charcoal emissions, transport and deposition is clearly a relevant topic, both for atmospheric modeling and for climate studies, and the con- struction of models capable of dealing with this class of particle is very welcome. The paper is thorough and well written, and well suitable for publication in ACP. I do, how- ever, have some comments on the evaluation the authors perform of their model, and the conclusions they draw. Also, as with many technical papers, quite a number of statements and sentences are difficult to understand for a broader audience. Hence, I recommend major revisions before final publication.

Major comments: My main concern with the paper is about the emission scaling and validation against observations. The authors state very clearly that their initial implementation of charcoal particles fails to capture the full range of variability in the observations. This is quite understandable, and improving this correspondence should a fruitful and important line of research in coming years. Looking at Figures 1 and 2, however, it's clear that both the scaling factor used for emissions, and the parameters chosen for density and mean/threshold radius, don't really affect the correspondence much. I understand that the parameters are guided by observations (section 3.1.2), but both the early figures and Table 1 show that varying them don't really change the

correlation. I believe that the reason for this is that there are physical processes, both in transport and charcoal retrieval, that are not represented in the model - as the authors also comment on. Hence, the whole discussion of parameters and emission factors could be toned down quite a bit. It's relevant as a sensitivity test, and should be mentioned, but as the paper stands it seems to indicate that some conclusion about the "best" parameter set can be drawn - and I don't think the numbers support that. Further, the emission scaling is confusing. The numbers 34 and 40 are used interchangeably through the manuscript - probably indicating that the lack of wide range correlation with observations precludes a more precise estimate. I would think that sensitivity study of emission scaling factors would be as, or more, important than the microscopic parameters discussed - so here, I would encourage the authors to add a little bit more information on how the scaling was chosen. (Especially since the abstract states that a factor of approx. 40 matches the calibration dataset "best".) In conclusion, I recommend harmonizing the detail level in the discussions of micro- scopic parameters and emission factor, and admitting more clearly that the lack of variability in the model results precludes drawing firm conclusions about either. The implementation itself is important enough to warrant publication. **We perfectly agree with the reviewer. In principal, we cannot derive from our simulations which parameter set is the most realistic one because of the underestimated variability and the weak Pearson correlation. Since the scaling factor depends on the parameter set, there is consequently also uncertainty concerning the scaling factor. We changed the following to account for the reviewer's comments:**

- **We changed the sentence in the abstract to: "We found that scaling black carbon fire emissions from the Global Fire Assimilation System (a satellite-based emission inventory) by approximately two orders of magnitude matches the calibration dataset best."**
- **We shortened the text about the realistic range of the parameters and partly moved it to the (new) supplementary material.**

- **The scaling factor of 34 was used as an initial estimate. The scaling factor was then adapted until it was in best agreement with the observations. In the old paper version, this was 40. We rewrote the Section "Calibration of emission":**

  "We conducted test simulations and compared the result to the European observations from Adolf et al. (2018). Three measures were used for the comparison: i) the Pearson correlation, which is a measure for linear correlation; ii) the Spearman rank correlation, which assesses monotonic relationships; iii) the quartile coefficient of dispersion, which is a normalised and robust variability measure ($\frac{Q_3-Q_1}{Q_3+Q_1}$, where Q_1 and Q_3 are the first and third quartiles, respectively). Table 1 shows some parameter combinations with positive correlation coefficients. In all test simulations, the correlation coefficients are very similar. While the Pearson correlation coefficients are low (0.21-0.23) and statistically insignificant, the Spearman rank correlation coefficients are much higher (0.67-0.69) and statistically significant. One reason for that are some observations with clearly larger charcoal fluxes than the simulated values ("outliers") because the Pearson correlation coefficients are much more sensitive to outliers than the Spearman rank correlation coefficients. These outliers can nicely be seen in Supplementary Fig. 4 for the example of *remi2.5,rthr3.9,dens0.5*." ... "The quartile coefficients of dispersion (Table 1) show that the variability differs between the test simulations. The simulation with the highest variability (*remi5,rthr4.9,dens0.6*; still having a lower variability than the observations, though) has only slightly lower correlation coefficients than the other simulations. Therefore, we choose this parameter set as the "best". However, we are aware that choosing the parameter set with the highest variability might compensate for errors not related to the parameters (e.g. the model resolution) that are responsible for an underestimated variability. Furthermore, none of the parameter sets has a statistically significant Pearson

correlation. Therefore, we cannot conclude from our simulations which parameter set is the most realistic one.

For the chosen parameter set (*remi5,rthr4.9,dens0.6*), we conducted simulations with different scaling factors (see Supplementary Fig. 5). The correlation coefficients and the quartile coefficients of dispersion do hardly depend on the scaling factor because charcoal particles do not coagulate with each other. We did not use the root mean squared error as a measure for the best scaling factor because the charcoal observations span several orders of magnitudes and the absolute deviations would be biased by the highest absolute charcoal fluxes (including the outliers). Instead, we consider the scaling factor for which approximately the same number of observations lies above and below the 1:1 line to be in best accordance with the observations. This is the case for a scaling factor on the order of $SF = 250$ (see Supplementary Fig. 5c), which has furthermore the smallest mean absolute error. However, note that the scaling factor depends on the chosen parameter set. Considering all parameter sets listed in Table 1, the best scaling factors range between $SF \approx 50$ and $SF \approx 250$."

Minor comments:

- *Section 3.1.4: Interactions with radiation and clouds is a whole other topic, which is insufficiently covered by this section. E.g. the assumption about spherical particles will have large implications for radiative transfer. I recommend removing this discussion and taking it up more thoroughly in a later publication.*
- **We shortened this section considerably and say now that we do not expect that the radiation and the interactions with cloud microphysics will have a large impact on our results.**
- *p3,l1: Should be DM<10 micrometers for microscopic?*

- **No, it is indeed $D_M > 10\ \mu m$ for microscopic and $D_M > 100\ \mu m$ for macroscopic charcoal.**

- *p3,l7-8: "homogenised the variance of individual records with a Box-Cox transforma- tion, rescaled the transformed data to the range (0, 1), and standardised it." This is a good example of a line that is too technical for its context. I recommend making the introduction more accessible to a broader audience.*

- **We changed the text to: "To circumvent the problem of inhomogeneous data, global synthesis studies such as Power et al. (2008) and Marlon et al. (2008) homogenised, rescaled, and standardised the data."**

- *p6,l27: "The right-skewed histogram of Clark and Hussey"... is another example. Please explain, so the uninformed reader doesn't have to look though the references.*

- **We deleted the word "right-skewed". It is not really necessary since the implication of the right-skewedness is mentioned in the text.**

- *p10,l1: Here, the authors describe an ageing process of charcoal particles, which likely influences wet deposition rates. What is the ageing timescale? How sensitive are the results (and the variability) to this parameter?*

- **In ECHAM6-HAM2, no ageing timescale is calculated. The ageing – i.e., coagulation and coating with sulphate – is explicitly calculated while the model is running. We can therefore not easily adapt the ageing timescale. However, if the particles aged faster, then their lifetime would be lower. This is not only due to increased wet deposition, but also due to faster sedimentation, since the aged charcoal particles contain more material and are larger.**

- *p10,l21: "...nobody has measured..." Out of curiosity, could FireLab (https://www.firelab.org/) have performed some relevant experiments here? I saw something at a conference a while back, but can't quite recall the details.*
- **Thank you very much for this hint. Unfortunately, we could not find any measurement from firelab which covers the relevant size range for microscopic charcoal particles.**
- *p11,l12: "...and some uncertain parameters..." Which ones? Please be specific. (It's described below, so it's just a matter of wording.)*
- **We rewrote the text.**
- *p11,l29: "Pearson correlation coefficients larger than 0.2." Is this the requirement for significance for your number of degrees of freedom? Please discuss the significance criteria in a bit more detail. (It's mentioned in a few places that the correlation becomes significant at the 5% level; according to what test? Which numbers are not significant?) This is related to my main comment above.*
- **As mentioned above, we rewrote the Section "Calibration of emissions" and make it now clear whether the results are statistically significant or not. Moreover, we added the following sentence to the methodology: "For comparing the simulations with the observations (e.g. calculating correlation coefficients), we used the SciPy package (Jones et al., 2001–)."**
- *p15,l25: Appendix B: Is there any way, based on the present data, to estimate the contribution of charcoal particles to the global absorption aerosol optical depth? Prob- ably not with great precision, but this is a quite open issue. (See e.g. a recent review here: https://link.springer.com/article/10.1007%2Fs40641-018-0091-4)*

- **We added the following text to the Supplementary Material: "In our simulations, we found that the vertically integrated charcoal mass in the atmosphere is approximately one order of magnitude smaller than the mass of dust (using the chosen parameter set). Therefore, charcoal only contributes little to the total aerosol absorption optical thickness in our simulations. However, our simplified approach is very uncertain and does also not consider the non-sphericity of charcoal particles. If the absorption of charcoal were larger than with our simplified estimate, the contribution to the aerosol absorption optical thickness might be somewhat higher, although we do not expect it to be large."**

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