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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of black carbon emissions from Siberian fires using satellite observations of absorption and extinction optical depths" *by* Igor B. Konovalov et al.

Anonymous Referee #2

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The main goal of the manuscript by Kanovolov et al. is to investigate the feasibility of constraining biomass burning (BB) BC emissions by using OMI measurements of aerosol absorption optical depth (AAOD). A long-standing difficulty with using OMI AAOD for this goal is that brown carbon can impact the measurements in ways that are difficult to account for within the inverse modeling process. In this work, a combination of relationships between EC, OC, single scattering albedo and AOD measurements are used to cleverly circumvent this traditional shortcoming. The work is thus novel and interesting in this regard, methodologically. This technique is then used to estimate BB emissions in the arctic, providing some valuable top-down estimates of emissions in this region that are significantly different than widely used inventories (e.g., GFED4).

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The manuscript is overall fairly well written, although there are some aspects of the inverse modeling procedure that could be clarified. It would also further strengthen the paper if the authors more clearly evaluated the impact of their top-down estimates as compared to simulations without any optimization of emissions scaling factors. These comments and other smaller issues and questions are described below. Addressing these likely amounts to minor revisions, after which this paper will be suitable for publication in ACP.

Comments:

Major:

5.7: I'm not sure that not making us of a priori information is touted as being a feature, nor that this statement is strictly correct. Bayesian inference is superior for a reason, in that the use of prior knowledge is essential to the solution of inverse problems. Further, there are plenty of sources of a priori information used in the methods described here. CHIMERE uses a prior anthropogenic inventory (HTAP) which helps define background values that contribute to the inversion. In another sense, the FRP data serves as a source of prior information to build a BB emission inventory following Konovalov 2014. Further, one could think of the land cover information used therein as being counted as prior information, etc.

11.10: So BC aging not included in CHIMERE? I can understand neglecting SOA, but BC aging affects its lifetime and transport, particularly to the arctic, as shown in previous studies. Accounting for aging should go beyond keeping track of particle age, as a threshold, as aging changes the particle properties and susceptibility to scavenging and removal. Comment on how this assumption biases the results shown here.

Eq 4: I wonder why the authors took this approach rather than relating AAOD/AOD to SSA (which seems a bit more direct, given that SSA is measured, whereas EC/(OC+EC) is estimated from Eq 3 and they have the problem of random errors in the

independent variable so have to use ODR instead of least squares). Then they could use Eq 3 to substitute SSA for the ratio of EC/(EC+OC), and derive an equation similar to Eq 6 that allows for estimation of AAOD without using a model-calculated SSA for BB organic aerosol.

16.4: This is starting to sound a bit circular. Use of CHIMERE here to separate the background AAOD and AOD would imply that CHIMERE has the correct BB emissions. How much do these estimates change if the authors were to run CHIMERE using their posterior BB emissions estimates? Does this entire procedure need to be iterated? Or, by "background", do the authors mean non-BB AOD and AAOD? This would make a little more sense, being as they stated earlier that they don't use CHIMERE for computing AOD and AAOD of BB aerosol given uncertainties in refractive index of BB OC. Still.. It took me several times reading this section and section 2.1 to piece together the process being used. It could benefit significantly from a more clear explanation, that more clearly maps to the schematic in Fig 1. At present, Fig 1 is misleading, as only part (the non BB part) of the AOD and AAOD estimated from CHIMERE are used for calculation of the AAOD value that ultimately is used within the estimation procedure in red. So this part of the figure needs more detail. The source of the non-BB aerosols (HTAP) is another input to the modeling that should be indicated.

Eq 7: Minimization of the least absolute value of the model error corresponds (in a Bayesian sense) to assuming that the observation errors follow a Laplacian distribution. This the of L1 minimization is best applied when the data is suspected of having outliers. The authors state on p 18 that they suspect multiplicative errors. This would most likely result in a log-normal distribution, if the errors being multiplied are Gaussian, rather than a Laplacian distribution. Thus it would make more sense to perhaps optimize the log of the model error. This is, in some sense, what the authors have done by dividing the model error by the observations so that they consider only the relative model error. But the form is not quite the same. Instead, the authors should normalize by the observation error covariance. Not all of the observed AAOD and AOD measure-

ments have the same errors, and thus shouldn't be considered equally in the objective function being minimized here.

18.12: I agree the satellite observations help indicate the location of the fires. But there is still considerable uncertainty in the variability of emissions factors from one fire location to another, especially given the crude representation of land cover types (considering only 5 classes). This sort of uncertainty could lead to large errors in the spatial distribution of the magnitude of BC emissions, even if the fire locations and intensity are perfectly known. Thus, the authors are going to need to admit here that the aggregation error is an unavoidable consequence of the chosen computational method.

12.21: Later in the paper (Figs 5, 6), I was struggling to clarify what constituted the "base" simulation. Is this a simulation using initial estimates of BB emissions, or an estimate based on the application of the scaling factors optimized from Eq 7? It would be useful to specify here. In presentation of figures of simulations that use the optimized emissions, a clearer name (e.g., "optimized") would be better.

Figs 5, 6: As mentioned above, not clear what is "base" here. Simulation with scaling factors of 1.0? Or the "base case option" (p18, line 25) of the optimized results? Please clarify.

Fig 6: Either way, it would be instructive to see Fig 6 with the emissions scaling factors of 1.0 as well as with the optimized emissions scaling factors, to see the improvement owing to the inversion.

Minor:

Intro: Missing some papers on BC transport to the arctic, of which a few more have been recently published.

3.9-11: References on use of atmospheric observations to constrain BB emissions is a bit thin. There have been numerous studies in this area. Even some of the BB

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emission products use ambient measurements as part of their constraints, such as QFED. Update: some such studies are referenced later in the introduction; still it seems like something is missing at this point.

4.16-19: And also dust? Perhaps that's not a problem when focused on regions suspected of being influenced by BB, but the statement as currently written is more general than that. Also, aerosol layer height is an important source of uncertainty in these types of studies.

11.31: I'm confused by this statement. It seems the previous content was all about describing how AAOD is calculated in CHIMERE. But here the authors state that AAOD values from CHIMERE that include BB emissions are not used? This seems to contradict page 10, line 21 "we took into account BB emissions . . ." UPDATE: after reading the entire paper, this becomes clear. But it could perhaps be written here in a way that doesn't lead to an initial source of confusion.

18.7: Or it would require note more computational resources, but more computational tools, such as an adjoint or other (i.e. Lagrangian) source-oriented model. CHIMERE has an adjoint (Menut et al., 2000) although perhaps it is antiquated.

Fig 4: Does the non-zero intercept of this line tell you something about the refractive index of the OC?

Fig 7(b): I'm not sure this shows much other than the fact that the modeled EC/(EC+OC) ratio is rather consistent, which is likely owing the fact that the model is based on a fixed EC/OC ratios in the emissions. Still, the authors should comment on the one feature that runs counter to this trend in the figure, the cluster of green points above the 1:1 line, which potentially indicates sampling of regions influenced by non-BB sources.

23.7: The authors need to clarify the writing here with regards to precision vs accuracy. If the errors were truly random, they would not lead to bias.

Fig 8: The way the background concentrations are included has the downside of skewing the vertical scale such that differences between the model and observations appear minimized, visually. Instead, a linear scale should be used, and background concentrations could be plotted using a different vertical axis on the right hand side, or scaled by x10-100 so they could be plotted on the same scale. Also, more interesting than showing the background concentrations would be to show the model estimates when using the BB emissions with the optimized vs unoptimized emissions factors. Since "base" wasn't clearly defined in the presentation of the results, it's not clear to me which of these cases is being presented anyways.

Fig 10: The two cases don't appear to be significantly different; I would suggest just showing one. Also, I don't understand what the purple dots in panel (b) represent.

24.27: I thought it was stated in the CHIMERE model description that there was no aging of BC included in the model.

26.7: While uncertainty information isn't provided with the GFED4 inventory, there have been numerous studies evaluating this inventory compared to other bottom-up inventories as well as evaluations based on top-down constraints using ambient measurements. These studies provide some range of estimates for the uncertainty in GFED4 emissions that could be referenced. In other words, from previous work we know that it's not something small like 1%, and certainly error as large as x2 (or even x10, regionally) have been noted.

Editorial:

4.5 and other locations: Careful with random switches between present and past tense.

4.10: GEOS-Chem

8.12: "measurement data" is a little redundant.

10.32: distributions –> distribution

11.4: are anyway not –> are not

17.5: The authors cite Enting 2001, but the bibliography only includes Enting 2002.

Fig 2 and 9: Text in the figure is illegible. Suggest using solid black, font that is easier to read.

Fig 5: Small suggestion on labeling: Don't write "CHIMERE - bgr" or "CHIMERE - base", as that at first looks like subtraction. Just leave out the hyphen, or use a colon.

21.17: Figure 5?

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