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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraining CO_2 emissions from open biomass burning by satellite observations of co-emitted species: a method and its application to wildfires in Siberia" *by* I. B. Konovalov et al.

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We thank the Referee for the thorough critical evaluation of our manuscript. All of the Referee's concerns have been very carefully addressed in the revised manuscript. Below we describe our point-to-point responses to the Referee's comments. Please note that some of the Referee's critical remarks have already been addressed in an earlier author comment (Konovalov, 2014).

Referee's comment: Firstly, the problem addressed is of very high uncertainty since

it involves two inverse problem solutions to estimate the amount of consumed dry biomass from the satellite observations of AOD and CO column, followed by a scaling to CO2 emission using hugely uncertain factors reported in literature. Each of these steps brings errors. A particular problem is that CO and PM constitute minor fractions of fire smoke, whereas CO2 is its major component. Hence, the approach suggested in the paper tries to constrain the major component of the plumes by observing two minor ones. One can never obtain good accuracy with this.

We agree that the method proposed in our paper is presently associated with considerable uncertainties, but, as argued in Konovalov (2014), the magnitude of uncertainties can be substantially reduced in the future (specifically, as a result of better and more abundant measurements of emission factors and further progress in satellite measurements and modeling of trace species in the lower atmosphere). The method itself can also be developed further and employ, for example, satellite measurements of CO_2 in major plumes for estimation of the emission factor ratios (such an opportunity is mentioned in the Conclusions of the revised manuscript). Note that because CO_2 is a very long lived tracer, CO_2 plumes from fires would be inevitably mixed with plant uptake and fossil CO_2 plumes transported from long distance, resulting in a typically small signal from biomass burning emissions and variable and complex "background" concentrations. On the contrary, CO and aerosols are relatively short lived and not emitted by the vegetation, so their plumes from fires are clearly measurable, and even if they are minor mass fractions of the carbon emitted from fires, they have a much better signal to noise ratio for an inversion. Since the future is, in a general case, hard to predict, we hope that our revised manuscript will be judged by taking into account the present state of the science, including the fact that (to the best of our knowledge) available "bottom-up" CO_2 emission estimates from fires in such a large region as Siberia have never yet been validated with atmospheric measurements.

Referee's comments: Both CO and PM fractions in smoke refer to poor-combustion conditions and therefore are correlated. Odds are high to have their error correlated

too (see detailed comments below). These are bound to dramatically limit the accuracy of the estimates and essentially eliminate the added value of the two inversions, even if the inversions themselves are "perfect".

P.3119, The eqs.11,12 hold only in case of independent estimates, as the authors stated in p.3120. However, both CO and PM emissions refer to burning quality and type of fire (flaming – smoldering). Since the uncertainties in both CO and PM emission factors partly (largely?) originate from uncertainties in the combustion conditions, they become correlated too. The authors ignore it without even trying to check for error covariances. The statement in p.3120 line 19 goes unsupported and doubtful: there is no self-evident reason to believe that.

To address this referee's concern, we performed several modifications of our method. First, we modified our Monte Carlo experiment to take into account co-variations of the differences between simulated and measured data for CO and AOD (specifically, random shuffling of grid cells and days was done in exactly the same way in both CO and AOD datasets). This allowed us to take into account the combined error covariances associated with local variations in burning conditions, spatial patterns of plant population, as well as with possible common errors in transport of CO and aerosol in the model.

Second, we analyzed the relationship between the emission factors for CO and aerosol (specifically, for TPM) reported in literature and representing averages over the measurements made during several dedicated experimental campaigns. The description of this analysis performed separately for fires in extratropical forest and savanna and grassland is provided in the Supplementary material for the revised manuscript. The analysis revealed no evident indications that the regional average values of emission factors for CO and aerosol strongly covariate, and thus it supported our assumption that uncertainties in the CO and aerosol emission factor estimates involved in our CO_2 emission estimation procedure are statistically independent and their covariance can be neglected. Note that the emission factor errors, which are explicitly specified in the

Monte Carlo experiment, are assumed to represent the diversity of the emission factors across the regions in which they were measured; by definition, such uncertainties are decoupled from the uncertainties associated with spatial and temporal variability of the emission factors inside of the study region.

Third, Eqs. 11, 12 were modified for a more general case where the estimates of the FRP-to-BBR conversion factors derived from CO (α^{co}) and AOD (α^{aod}) measurements are not statistically independent. Both the Monte Carlo experiment and estimation procedure were re-done with the modifications. The results support the initial assumption made in the reviewed manuscript that the impact of the error covariances on our estimates of CO_2 emissions is quite negligible, and that they do not eliminate the added value of combination of the AOD and CO inversions.

Note that the results of the updated Monte Carlo experiments performed by taking into account the error covariances were first tested and then analyzed in much more detail than it is described in the revised manuscript (since we had to take care of its length). In particular, we made sure that when the assumed uncertainties in the temporal-spatial fields of AOD and CO data are identical and the uncertainties in the emission factors and in the mass extinction efficiency are not explicitly taken into account, the random samples of the α^{co} and α^{aod} strongly covariate (R^2 $\sim$0.7), as could be expected. (The co-variation was not perfect due to different sensitivities of the modelled CO and AOD fields to the emissions from fires). The samples of α^{co} and α^{aod} obtained with the actual fields of the residual errors (see Section 2.3.3) in the simulations and measurements manifest much smaller covariances (R^2 $\sim$0.1). Interference of those errors with independent uncertainties in the regional estimates of the emissions factors in the framework of the "full" Monte Carlo experiment virtually eliminates the covariance of the errors in α^{co} and α^{aod}.

Although we believe that our uncertainty estimates are sufficiently realistic, we provided the following caveat (see Section 2.4): "...since the exact nature and characteristics of uncertainties in the input data for our analysis are not known (as it is common for vir-

tually any "real world" application of the inverse modelling approach), the uncertainties reported below for our estimates of the conversion factors and CO_2 emissions should be considered with caution." Indeed, estimation of uncertainties in inverse modeling results presents a big common issue, which does not have any easy solution. While most of inverse modeling studies involve subjective (so called, "expert") quantitative characterization of model errors and uncertainties in a priori estimates (which are not used in our study), the important advantage of our method is that we base our uncertainty estimates exclusively on the statistical analysis of the differences between our simulations and available observations.

Note that the preliminary analysis of this issue outlined in Konovalov (2014) involved measurements of emission factors for organic carbon (OC) which are more sparse than the available data for emission factors for TPM. Although the conclusion was essentially the same, nonetheless, the results outlined above supersede the tentative results mentioned in Konovalov (2014).

Referee's comments: Secondly, the paper faced the problem reported by practically all related studies, including earlier works of some of the authors: whereas the CO emission factors deduced in bottom-up and lab studies meet the top-down assessments, the results for PM show about a factor of 3 under-estimation in the bottom-up inventories (with root cause probably being the low emission factors). This mystery is not yet resolved, i.e. simultaneous use of CO and PM literature-based emission factors must include some workaround. It is absent in the paper and, sadly but expectedly, the authors got about 3-fold difference between the mean estimates derived from CO and from AOD inversions (table 2). The authors noticed the problem but waved it out. In particular, they stressed (p.3130) that the uncertainty ranges of these estimates are overlapped. This, however, is not convincing because, firstly, the uncertainty ranges themselves are very poorly known and their tiny overlap can be simply a coincidence. The authors themselves note that their error estimates are rather over- than under-stated, which suggests even higher odds for the difference being formally

"statistically significant". Secondly, the overlap, even if exists, refers more to huge uncertainty ranges (up to a factor of 5 and even more) than to indeed closeness of the estimates. Since the authors are interested in absolute CO2 emissions, which are given in table 3 with 3-digit(!) accuracy, a factor of 3 difference between the outcome of the CO- and AOD-based retrievals is hardly acceptable. Once again, the root cause for this, to my mind, is that the literature-based PM emission factors must be used with the highest care until the problem is resolved. I have not seen much criticism on CO emission factors and assume that they (so far) represent consensus among the researchers. The authors discuss the issue (pp.3132-3133) but somehow ended up with a conclusion that this difference is insignificant. In view of the above, I disagree.

P.3129-3130, table 2. Now the problem comes. It is explained above in "General comments", here I just have to second the statement of p.3130 l.1-5: the combination of CO and PM retrievals using the literature dry-matter-to-CO and –to-PM conversion factors has inherent problems, which questions the value of the whole exercise. A possible way out is to use CO-based emission estimates of CO2 keeping PM-based values as a sensitivity study.

We recognize the potential problem associated with the inversion of AOD measurements, and we are sorry if the referee got the impression that we simply "waved it out". The discussion of this point is considerably extended in the revised manuscript, and a corresponding caveat is provided. Along with the CO_2 emission estimates constrained by both CO and AOD measurements, we provided the emission estimates based only on CO and only on AOD measurements.

Nonetheless, we remain confident that the uncertainty range given for AOD-based estimates of the conversion factors is sufficiently realistic and that thus there are no sufficient *objective* reasons for totally disregarding information provided by the AOD measurements, which automatically gets a smaller weight in our estimation procedure than information derived from CO measurements. We also believe that using PM emissions factors from literature in the framework of our study does not necessarily require any

special "workaround" involving any subjective judgments, since the potential problems associated with the PM emission factor measurements are likely manifested (unless those problems are of trivial nature, which is unlikely) as a diversity of the measurements performed using different instruments in different conditions, regions and seasons. Such diversity is taken into account in our analysis.

Furthermore, we did not have a sufficient ground for expectation that the AOD-based estimates of the FRP-to-BBR conversion factor would be 2-3 times larger than the CO-based estimates. Indeed, although this result is coherent, in particular, with the large correction imposed to aerosol emissions in the GFASv1.0 inventory, it is rather contradictory to the earlier results by Konovalov et al. (2011) who found that their CO and PM_{10} simulations were not consistent with the measurements of near-surface concentrations in the Moscow region in 2010, unless the ratio of CO to PM_{10} emissions was enhanced by about a factor of two with respect to the "standard" settings. A uniform underestimation of AOD in simulations based on the bottom-up inventories is also not supported by Petrenko et al. (2012) who found, in particular, that a global model driven by several bottom-up fire emission inventories tend to overestimates AOD (by up to a factor of 3) over equatorial African region. Finally, the available estimates of CO emission from biomass burning demonstrate a similar degree of uncertainties in Russian regions (see, e.g., Huijnen et al., 2012; Krol et al., 2013). Taking such contradictory results into account, we believe that many more studies involving satellite and ground based measurements of aerosol and co-emitted species along with chemistry transport models using different parameterizations of the key processes are needed to elucidate the potential issues concerning aerosol emissions from biomass burning and their origin. Our study contributed to advancing this active research area by providing (as far as we know, for the first time) the results of parallel inversions of both CO and AOD measurements of biomass burning plumes, as well as the results of tests with different model options. Therefore, we believe that in spite of existence of the probable unresolved problems mentioned above, our results (presented in our manuscript along with appropriate discussion and caveats) will be sufficiently interesting and use-

ful for a broad community specializing in estimation and modeling of biomass burning emissions and their atmospheric effects.

Referee's comments: Finally, the validation section 4.2 re-uses the same observations as were used for emission optimization. Such re-use of the fitted measurements to evaluate the fitting results is absolutely not acceptable. This is especially true because the authors analyze the very parameters heavily affected by the fitting (mean values, biases, RMSE) and ignore those less influenced (correlation coefficient, for instance). Why the authors didn't withhold half of the data from the fitting? The amount of observations is bound to be more than sufficient for that. In the current form the section 4.2 has no value, except for in-situ comparison, which leaves the study practically without any validation. A rigorous workaround to save the paper would be repeating the fitting with half of data withhold but I understand that it may be too painful. One can consider additional periods with strong fires, may be, in other years, although this is not completely painless either.

P.3134, l.1-10. This is the major problem. The wrong statement and an evident crude error in the approach are covered by hand-waving ("would hardly help : : : if emission is wrong"). See the general comments above.

We agree that, fundamentally speaking, optimization and validation data sets should be totally independent. However, we expected that because the number of the parameters optimized in our study was extremely smaller compared to the total number of data points, the use of the same dataset for both optimization and validation purposes could not lead to any wrong conclusions. The results presented in the revised manuscript, where each third day in the period considered was withheld for validation, confirm that expectation. The changes in the optimal estimates of the conversion factors and CO_2 emissions, as well as the changes in the performance statistics of our simulations due to splitting of the initial dataset into two parts, are not considerable.

Referee's comment: P.3102, l.21-23 I did not understand the division between wildfires

and "other types" and the following lengthy but pointless and confusing wording. Why not simply "emissions of CO2 and other species from wildfires are available from: : :"?

Our top down emission estimates, as well as alternative bottom-up estimates mentioned in the concerned paragraph, address emissions not only from wildfires but also from other types of open biomass burning (such as agricultural fires). That is why we could not talk about only emissions from wildfires. Nonetheless, the criticized sentence is shortened in the revised manuscript. Note that wildfires are indeed a dominant source of biomass burning emissions in Siberia (e.g., Lin et al., 2012).

Referee's comment: P.3107, l.23. GFAS emission estimation involves direct scaling to GFED totals as part of the procedure, as mentioned in p.3138. This deserves a clearer explanation here too.

The explanation is added in the revised manuscript.

Referee's comment: P.3113, l.10 I did not understand the reason for such brute-force approach to minimization. Why not to take some standard minimization routine? Just three dimensions of optimization should not be difficult. Problems may arise only if the data scatter is very large resulting in poor convergence. But then the uncertainties of the brute-force minimization will be large too. Explanation is needed here.

As explained in the revised manuscript, direct scanning of the parameter space of the approximation allowed us, on the one hand, to avoid the risk of finding a local minimum of the nonlinear cost function instead of a global one (while most of standard iteration minimization routines might be "trapped" in a local minimum). On the other hand, considerations of computational efficiency were not important in the given case due to relative simplicity of the numerical problem considered.

Referee's comment: P.3115, eq 7. The threshold level notation o is easily mixed with the number 0. The notation should be changed.

The notation is changed in the revised manuscript.

Referee's comment: P.3121 last line. Factual support is needed. How comes that the CO chemistry and secondary aerosol formation from non-fire sources has no impact on the study outcome?I would accept it for grid cells / days, where / when the fire-induced smoke is dominant. But the authors included all cells with fire contribution > 10%, i.e. up to 90% of the pollutants can be from other sources (eq 7, parameter o). For such cells the uncertainties in anthropogenic emission are bound to have strong impact.

The statement questioned by the referee was not formulated quite correctly. We only meant that the emissions of NOx and NMHC from biomass burning do not affect significantly the simulated evolution of pyrogenic CO and PM. The results of a corresponding test for a similar situation are given in Konovalov et al. (2011), Fig. 4. The respective changes are made in the revised manuscript.

Referee's comment: P.3125, l.14.

I did not understand: was MEGAN run online or CHIMERE received precomputed inventory?

Biogenic emissions were calculated "online" by using biogenic emission potentials from the MEGAN global inventory.

Referee's comment: P.3138 l 3-10. A very long and self-contradicting sentence collecting several arguments for and against independence of the GFAS and GFED datasets. Please restate.

The sentence is rewritten in the revised manuscript.

Referee's comment: P.3141, l. 5.

This is a confusing sentence. It should clearly separate the CO/PM model-based inversion to emission fields, which are then simply re-scaled to CO2 using literature data. Note that no evaluation is provided for the last step.

The corresponding part of the Conclusions is clarified as suggested by the referee.

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