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Estimation of black carbon emissions from Siberian fires using satellite observations of absorption and extinction optical depths

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S1. Sensitivity analysis of the empirical AAOD parameterization

In this supplementary section, we report several supplementary tests that were designed to evaluate the sensitivity of the empirical AAOD parameterization (see Eq. 4) discussed in Sect. 2.2.3 with respect to possible biases in the simulated AAOD and AOD data for the "bgr" scenario (see Eq. 5). The different test cases involved different $AAOD_b$ and AOD_b values that were modified by applying to them the constant scaling factors, s_1 and s_2, respectively.

First, we considered the case where the both s_1 and s_2 were equal zero. That is, an impact of the "background" aerosol on the columnar SSA values was entirely disregarded. This case is analogous to one addressed in Konovalov et al. (2017a), except that the data selection criteria were different (see Sect. 2.2.3). The estimate of κ_1 obtained in this case (see Fig. S1) is 15 % lower than that for the base case (see Fig. 4). The difference can be explained by the uncertainty at the 90th percentile confidence level. Using the parameterization presented in Fig. S1 for estimation of BB BC emission estimation procedure would accordingly result in about 15 % larger top-down estimates of BC emissions than those reported in Sect. 3.5.

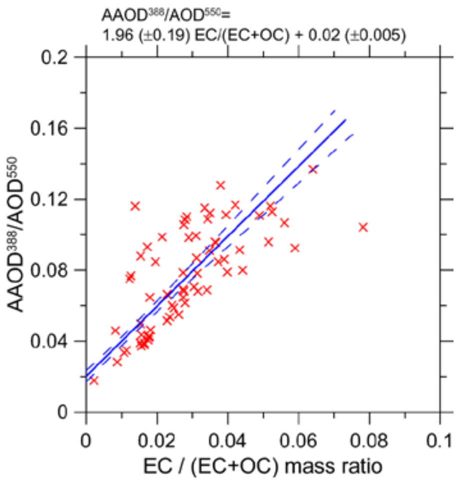

Figure S1: The AAOD/AOD ratio as a function of the EC/(EC+OC) ratio. Both ratios shown by red crosses have been derived from the observations at the AERONET sites as explained in Sect. 2.2.3, except that the $AAOD_b$ and AOD_b values involved in Eq. (5) were zeroed. A linear regression fitted with the ODR method and its 68.3 % (1-sigma) confidence intervals are shown by solid and dashed blue lines, respectively.

In the second test, s_1 was equal to 0.5 or 1.5 ($AAOD_b$ was increased or decreased by 50 %), while s_2 was equal to 1.0. The results indicate (see Fig. S2) that the parameterization is quite insensitive to the large changes in the background AAOD values. Although we cannot estimate possible biases in $AAOD_b$, the fact that the decrease of the $AAOD_b$ values decreases the uncertainty of both κ_1 and κ_2 may be regarded as evidence that $AAOD_b$ is biased positively.

Finally, in the third test, the $AAOD_b$ values were kept constant (s_1 was equal 1.0) but the AOD_b values were increased or decreased by 50 % (s_2 was set to be 0.5 or 1.5). The decrease of AOD_b results in a rather small decrease of κ_1 (by 8 %), but the increase of AOD_b leads to a substantial increase (by 20 %) of the same coefficient. Therefore, if AOD_b were strongly underestimated, the AAOD/AOD ratio would also be considerably underestimated in our simulations, while the BB BC emissions obtained in our analysis would be accordingly overestimated. However, although we do not have any information about probable biases in AOD_b at the AERONET sites considered in this study, there is evidence (see Sect. 3.6) that AOD_b is actually overestimated by about 40 % on average for the whole study region. Therefore, the underestimation of AAOD/AOD ratio by the parameterization presented used in our analysis (see Fig. 4) is possible but not likely.

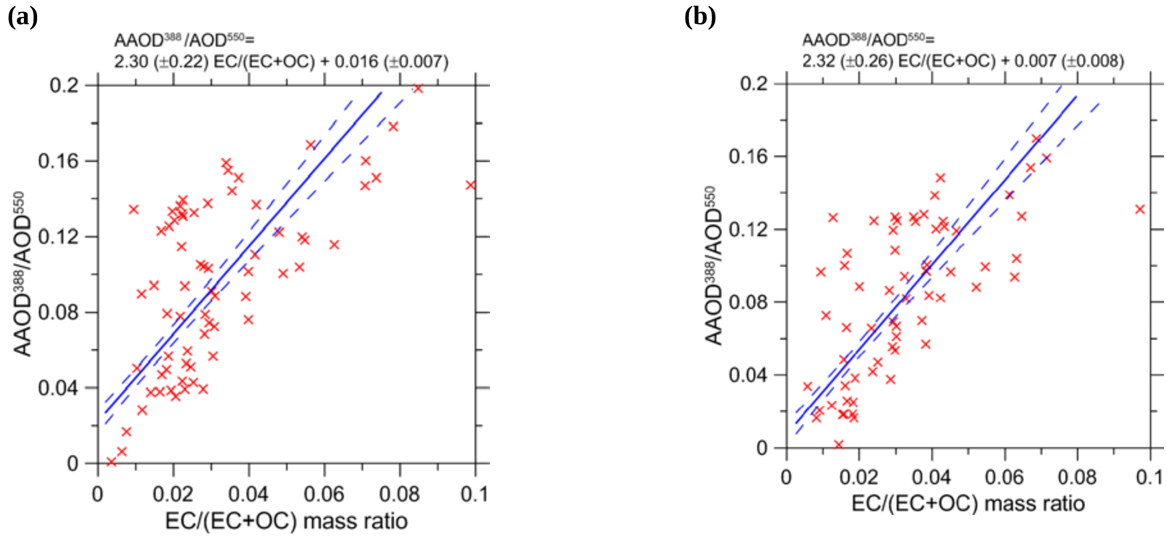

Figure S2: The same as in Fig. S1, but with the original AOD_b values and with the $AAOD_b$ values (see Eq. (5)) that were (a) decreased by a factor 0.5 and (b) increased by a factor of 1.5.

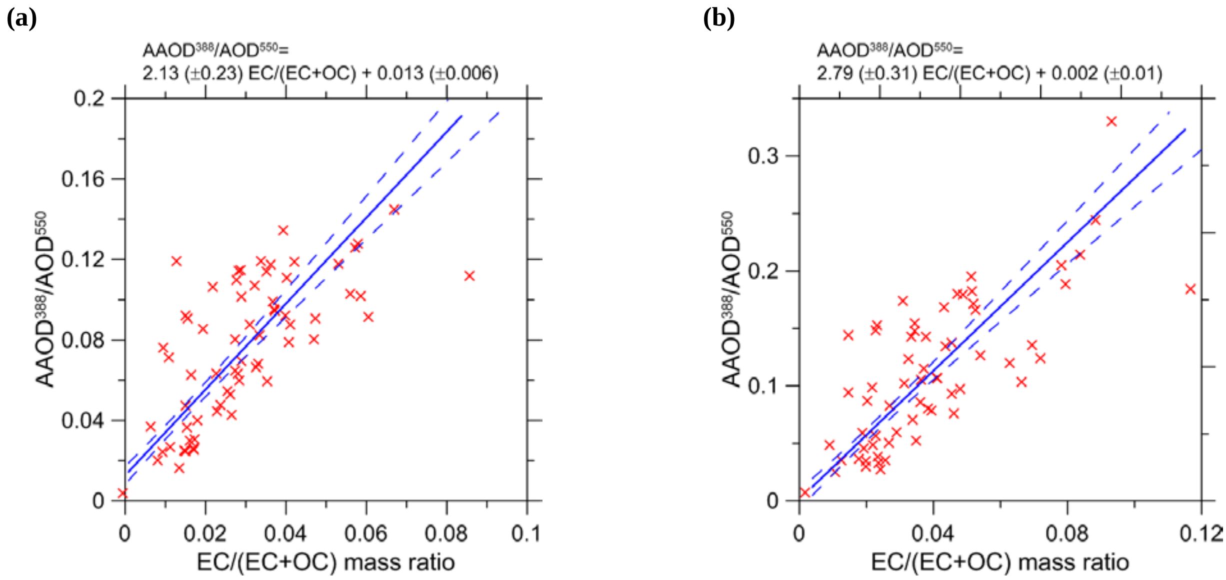

Figure S3: The same as in Figs. S1 and S2, but with the original $AAOD_b$ values and with the AOD_b values that were (a) decreased by a factor 0.5 and (b) increased by a factor of 1.5.

S2. The effect of the observation and model errors on the top-down BC emission estimates

Here we briefly analyze an impact of the observation and model errors on our top-down BC emission. Let us V_o be the observed AAOD value in an arbitrary grid cell and day of a given month. We assume that V_o depend linearly on the BB BC emissions, $\boldsymbol{E}$, as follows:

$$V_o = \sum_j S_j E_j + V_b + \varepsilon_o \tag{S1}$$

where $\boldsymbol{S}$ is a vector describing the sensitivities of AAOD to the BB BC emissions, V_b is the corresponding background AAOD value, and ε_o is an observational error. Note that the components of the vector $\boldsymbol{E}$ are the BC emissions in different grid cells of the study region and different days of a given month. We further can assume similar relationships between the simulated AAOD and the corresponding emission fields:

$$V_m = \sum_j S_j e_j F^{BC} + V_b + \varepsilon_s \tag{S2}$$

where $\boldsymbol{e}$ is the BB BC emission field (in a general case, different from $\boldsymbol{E}$) specified in the model, F^{BC} is the emission correction factor, and ε_s is a model error. For simplicity, we assume that $\boldsymbol{S}$ and V_b represent exact values of the sensitivities and of the background part of AAOD (otherwise, the corresponding errors could be represented by ε_s). We assume also that the modeled AOD values involved into our simulations of AAOD have already been optimized.

By requiring (in accordance to Eq. 7) that the mean values of V_o and V_s are approximately equal and using Eqs. (S1) and (S2), we obtain the following estimates for the F^{BC}:

$$F^{BC} \cong \frac{\left\langle \hat{S}E \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_o \right\rangle}{\left\langle \hat{S}e \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_m \right\rangle}, \tag{S3}$$

where angular brackets denote averaging over values available (in different grid cells and days) for a given month, N_v is the total number of the available AAOD or AOD data points, N_e is the product of the total numbers of days and grid cells in the month and region considered, and $\hat{S}$ specifies the cumulative contribution of the BB BC emissions in a given grid cell / day to AAOD values throughout the study region and a month considered*:*

$$\hat{\boldsymbol{S}} = \sum_i \boldsymbol{S}_i \,, \tag{S4}$$

where i is the index of a grid cell / day. Similarly, we can obtain an estimate for the total BC emissions, $\hat{E}_{tot,}$ and to evaluate the relative error, δ_e, of $\hat{E}_{tot}$:

$$\hat{E}_{tot} \cong \frac{\left\langle \hat{S}E \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_o \right\rangle}{\left\langle \hat{S}e \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_s \right\rangle} e_{tot}\,, \tag{S5}$$

$$\delta_e \cong \frac{\left\langle \hat{S}E \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_o \right\rangle}{\left\langle \hat{S}e \right\rangle + N_e^{-1} N_v \left\langle \varepsilon_s \right\rangle} e_{tot} E_{tot}^{-1} - 1, \tag{S6}$$

where E_{tot} is the true (unknown) value of the total BB BC emissions.

We can further transform Eq. (S6) as follows:

$$\delta_e \cong \frac{\left\langle \hat{S} \right\rangle E_{tot} + N_e \left\langle \hat{S}'E' \right\rangle + N_v \left\langle \varepsilon_o \right\rangle}{\left\langle \hat{S} \right\rangle e_{tot} + N_e \left\langle \hat{S}'e' \right\rangle + N_v \left\langle \varepsilon_s \right\rangle} e_{tot} E_{tot}^{-1} - 1 \;, \tag{S7}$$

where $\hat{S}'$, E', and e' denote the deviations from the corresponding mean values. It is reasonable to expect that because the sensitivity of the AAOD values to BC emissions is mostly determined by transport processes independent of the emissions, the co-variances $<\hat{S}'E'>$ and $<\hat{S}'e'>$ should approach zero if the number of the data is sufficiently large and they are sufficiently representative of an entire region and a period considered. Then, if both the observations and simulations of AAOD are unbiased (that is, $<\varepsilon_o>$ and $<\varepsilon_s>$ are negligible), the estimation error approaches zero. In other words, the above analysis indicates that given an unlimited amount of the unbiased observational and simulated data, our optimization procedure based on Eq. (7) is expected to yield an unbiased estimate of the total BC emissions.

In the real situation considered in this study, the amount of the data is limited, and thus our estimate is likely to be affected by uncertainties. We also cannot exclude that the observations and simulations are affected by some biases. However, we expect that the resulting uncertainty associated with the random observation and model errors is included in the confidence intervals evaluated by means of a bootstrapping procedure (see Sect. 2.2.4). Possible biases in the simulated and observational data and their effect on our BC emission estimates are discussed in Sect. 3.6.

Note, finally, that if some emission sources are systematically underrepresented in the available observation data, the co-variances $< \hat{S}'E' >$ and $<\hat{S}'e'>$ will likely be different from zero, and the emission estimate will be affected by the aggregation error. However, unlike the case of a scarce ground-based monitoring network discussed, e.g., by Kaminski et al. (2001), the satellite data cover our study region quasi-uniformly. Moreover, at least a part of this potential error is accounted for in our bootstrapping procedure described in Sect. 2.2.4. Therefore, we believe that the aggregation error does not exceed the confidence intervals for our estimates. Nonetheless, we cannot provide a reliable quantitative estimate for the aggregation error in our case. The likely presence of the aggregation error emphasizes the importance of validation of our estimates by using independent observations (see Sect. 3.3 and 3.4).

S3. Estimation of the correction factors for BB BC and OC emissions in a selected sub-region

In this section, we present the optimal estimates of the correction factors, F^{BC} and F^{OC}, for a sub-region covering the south-west part (50-57° N, 60-115° E) of our study region (see purple dashed rectangles in Fig. 2). Figure S4 shows our calculations of the integral fire radiative energy released from forest and other (mainly agricultural and grass) fires in this sub-region. Values shown in Fig. S4 can be compared with similar values calculated for the entire study region and presented in Fig. 3. Evidently, compared to the whole study region, the sub-region features much larger contributions (ranging from 44 % in June to 88 % in July) of agricultural and grass fires to FRP in any month considered. Furthermore, agricultural and grass fires provide the predominant contribution to FRP (exceeding 70 %) in each month except June.

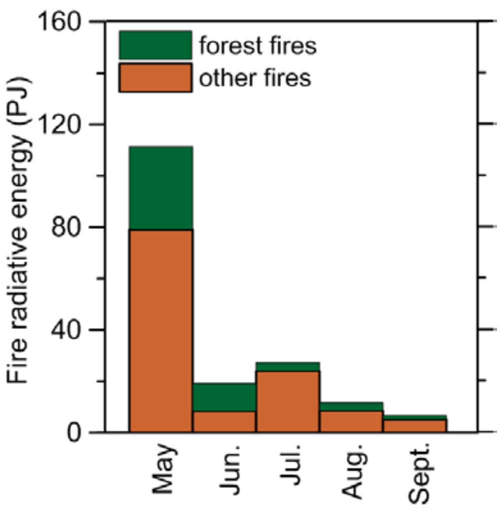

Figure S4: The same as in Fig. 3 but for the sub-region covering the south-west part (50-57°N, 60-115° E) of the study region.

We employed the same estimation procedure as that described in Sect. 2.2.4, except that the satellite AAOD and AOD data outside of the selected sub-region were disregarded. Note that the F^{BC} and F^{OC} estimates obtained in this way are, to some extent, affected by BB aerosol that was emitted outside of the given sub-region but transported into it. Nonetheless, for the purposes of the analysis discussed here, it is sufficient that the selected observations are more sensitive to the emissions in the considered sub-region than the disregarded observations made outside it.

The correction factor estimates derived from the observations corresponding to the considered sub-region are reported in Table S1 and are discussed in Sect. 3.1. Note that the estimates for September could not be obtained, as there were no satellite data satisfying the common criteria specified in Sect. 2.2.4 and corresponding to the given sub-region.

Table S1. The same as in Table 2 but for the selected sub-region (50-57°N, 60-115°E) of the study region.

Correction factor	May	June	July	August	September
F^{BC}	3.32 (± 1.31)	2.15 (±0.72)	1.94 (±0.68)	5.21 (±2.27)	NA
F^{OC}	2.87 (± 0.95)	2.16 (±0.55)	2.66 (±0.65)	3.77 (±1.34)	NA
F^{BC}/F^{OC}	1.16 (± 0.37)	0.99 (± 0.32)	0. 73 (± 0.23)	1.38 (± 0.50)	NA

References

Kaminski, T., Rayner, P. J., Heimann, M., and Enting, I. G.: On aggregation errors in atmospheric transport inversions, J. Geophys. Res., 106, 4703, doi:10.1029/2000jd900581, 2001.