Evaluation of the BC / OC ratios for aerosol emissions from biomass burning in Siberia using AERONET retrievals

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INTRODUCTION

Biomass burning (BB) aerosol is known to have a considerable impact on the radiation budget of the atmosphere as a result of both scattering and absorption of the solar shortwave radiation and increasing the downward longwave radiation. A balance between the cooling and warming effects of aerosol critically depends on the ratio of the major absorbing and scattering components in particles, such as black carbon (BC) (or elemental carbon, EC) and organic carbon (OC), respectively. Available direct measurements of this ratio in remote regions (like Siberia) are limited and rather uncertain.

We examine a simple method to estimate the BC (EC) / OC ratio in BB aerosol by using retrievals of aerosol optical properties from Aerosol Robotic Network (AERONET) measurements along with an empirical relationship between the EC / OC ratio and the single-scattering albedo (SSA).

METHOD

Model and measurements data

Measurements data

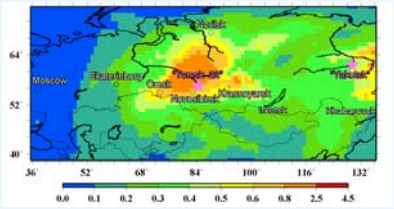
AERONET Version 2, Level-2 aerosol inversion product: SSA at the 440-, 675- and 869-nm wavelengths; AOD for the 440- and 500-nm wavelengths.

FRP data (the MODIS Collection 6 Level 2 data product)

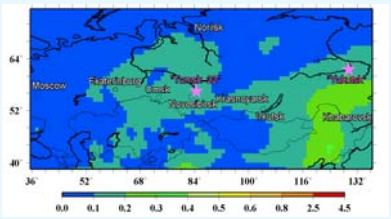
Model data

CHIMERE chemistry-transport model (v2016): simulations of AOD, BB aerosol photochemical age [Konovalov et al., 2017a] , RH in BB aerosol columns, EC/OC ratio in BB aerosol (based on GFED4.1 emission factors)

WRF v.3.6 (meteorology)



Averages of AOD^{500} from CHIMERE simulations with (above) and without (below) fire emissions, calculated only over the data and hours, that were supplied with measurement data from the Tomsk-22 AERONET site (Jun.–Aug. 2012)



An empirical relationship between EC/OC ratio and SSA [Pokhrel et al., 2016]:

$$\omega_0^{\lambda} \cong a^{\lambda}\frac{[EC]}{[EC]+[OC]}+b^{\lambda} \Rightarrow \frac{[EC]}{[EC]+[OC]} \cong (\omega_0^{\lambda}-b^{\lambda})/a^{\lambda} \quad (1)$$

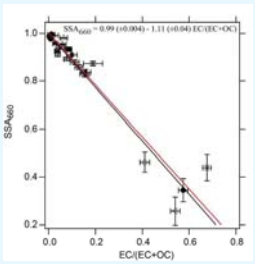
ω_0^{λ} is the single scattering albedo (SSA) at a given wavelength λ

[EC],[OC] are the mass concentrations of EC and OC

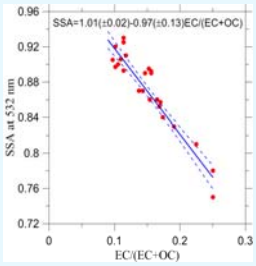
a^{λ},b^{λ} are the coefficients of the linear regressions

The coefficients of the linear regressions fitted to the Fourth Fire Laboratory at Missoula Experiment (FLAME-4) [Pokhrel et al., 2016]

Wavelength λ	Slope (a^{λ})	Intercept (b^{λ})
405 nm	−1.07 (±0.08)	0.94 (±0.007)
532 nm	−1.06 (±0.04)	0.99 (±0.004)
660 nm	−1.11 (±0.04)	0.99 (±0.004)



A linear fit (red line) and an ODR fit (black line) for the relationship between the EC/(EC + OC) ratio and SSA^{660} [Pokhrel et al., 2016].



A linear fit and the 90% confidence intervals for the relationship between the EC/(EC + OC) ratio and SSA^{532} (based on aircraft observations by Yokelson et al. [2009]).

Analysis

Selection criteria applied to the AERONET observations:

1	AOD at 500nm	>0.5
2	Relative humidity in the BB aerosol column	<60%
3	BB aerosol photochemical age	<30 h

EC/OC estimate given by Eq. (1) is very sensitive to uncertainties in b^{λ} and estimates of b^{λ} from the FLAME-4 may be not representative for aged BB aerosol in boreal region. A way to exclude it from analysis:

$$\frac{[EC]}{[EC]+[OC]} \cong (\omega_0^{869}-1)/a^{869} \text{ (assuming } b^{869}=1)$$

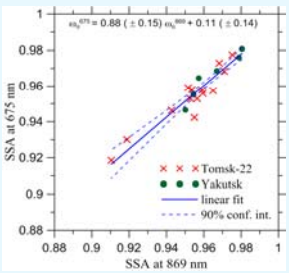
$$\omega_0^{\lambda} \cong A^{\lambda}\omega_0^{869}+B^{\lambda},\ A^{\lambda}=\frac{a^{\lambda}}{a^{869}},\ B^{\lambda}=b^{\lambda}-\frac{a^{\lambda}}{a^{869}}$$

$$\frac{[EC]}{[OC]} \cong \left[\frac{1}{(\omega_0^{869}-1)A^{\lambda}/a^{\lambda}}-1\right]^{-1} \quad (2)$$

Distinctive features of the different cases of estimation of the EC/OC ratio using the AERONET observations

Case 1 (base case)	**The estimates are obtained using Eq. (2) and SSA observations at 675 and 869 nm**
Case 2	**The same as Case 1, but using SSA observations at 440 nm**
Case 3	**The estimates are obtained using the available parameterization (Eq. 1) for SSA at 660 nm**
Case 4	**The same as Case 3, but using the parameterization for SSA at 405 nm**

RESULTS

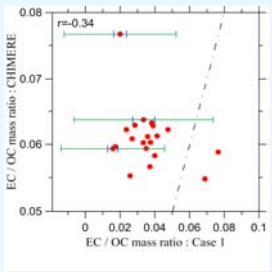


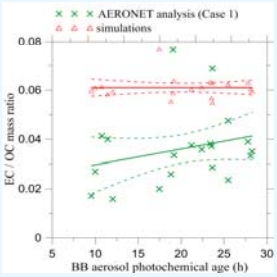
The AERONET SSA observations (ω_0) at 675 nm plotted as a function of the corresponding observations at 869 nm.

Average values of the EC/OC ratio estimates derived from the AERONET observations according the different estimation cases along with the corresponding value predicted by simulations with the CHIMERE model.

Case 1	**0.036 (±0.009)**
Case 2	**0.038 (±0.035)**
Case 3	**0.031 (±0.009)**
Case 4	**0.002 (±0.011)**
CHIMERE	**0.061**

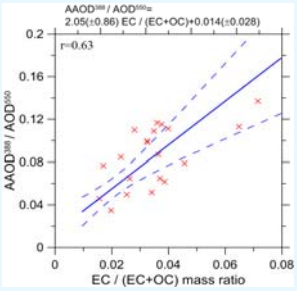
The relationship between the EC/OC mass ratios derived from the AERONET data and predicted with the CHIMERE. The 90% conf. intervals (green error bars) considering the uncertainty of the approximation and the probable error (of 0.03) of individual SSA observations, while the 90% confidence intervals (blue brackets) include only the uncertainty of the approximation. The dot-dashed line depicts the 1:1 ratio.

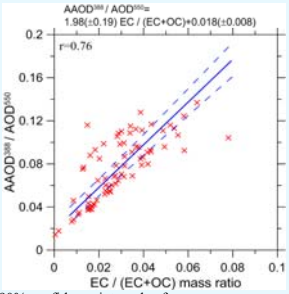




The dependence of the EC/OC mass ratio on the photochemical age of BB aerosol according to the analysis based on the AERONET data (green solid line) and the simulations with the CHIMERE CTM (red solid line). The 90% confidence intervals of the linear fits (simple linear regressions) are shown by the green and red dashed lines.

The ratio of $AAOD^{388}$ and AOD^{550} evaluated using AERONET observations with (left) and without (right) applying selection criteria as a function of the EC/(EC + OC) mass ratio





A linear regression (fitted with the ODR method) and the 90% confidence intervals of the fit are shown by solid and dashed lines, respectively.

$$\omega_0^{440} = f([EC]/[EC]+[OC]) = 1-\frac{AAOD^{440}}{AOD^{440}}$$

$AAOD^{440}$ and AOD^{440} are extrapolated to the 388-nm and 550-nm wavelengths, respectively, using the corresponding *Ångström* exponents, α_a and α_e:

$$\alpha_a = -\frac{\log(AAOD^{440}/AAOD^{675})}{\log(440/675)}$$

$$\alpha_e = -\frac{\log(AOD^{440}/AOD^{500})}{\log(440/500)}$$

Please see Konovalov et al. [2017b] for further details

CONCLUSIONS

- We used the AERONET observations to estimate the EC/OC mass ratio in BB aerosol in Siberia. The EC/OC ratio values in June-August 2012 are found to range from 0.015–0.077, with the mean value of 0.036 (±0.009). The estimated mean value of the EC/OC ratio is found to be significantly smaller than the corresponding value (0.06) predicted by the BB aerosol simulations based on the emission factors specified in the GFED4.1 emission inventory, but in a very good agreement with the value of the EC/OC enhancement ratio (0.038) evaluated in long-term in situ measurements of carbonaceous aerosol at Zotino [Mikhailov et al., 2017].
- The EC/OC ratio estimates derived from the AERONET SSA observations using the combination of observations at 869-nm and 675-nm wavelengths are found to be consistent with the corresponding estimates derived from the combination of observations at 869-nm and 440-nm wavelengths. There are significant discrepancies between the estimates derived from the AERONET data with the original FLAME-4 parameterizations for SSA of fresh BB aerosol at the 660- and 405-nm wavelengths. The parameterization for 405 nm is found to strongly underestimate the EC/OC ratio; this may be due to a decrease of BrC absorption in aging BB aerosol plumes.

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REFERENCES

1. Konovalov, I.B., Beekmann, M., Berezin, E.V., Formenti, P., Andreae, M.O. Probing into the aging dynamics of biomass burning aerosol by using satellite measurements of aerosol optical depth and carbon monoxide. *Atmos. Chem. Phys.*, **2017a**, 17, 4513–4537.
2. Konovalov, I.B., Lvova, D.A., Beekmann, M. Estimation of the Elemental to Organic Carbon Ratio in Biomass Burning Aerosol Using AERONET Retrievals. *Atmosphere*, **2017b**, 8(7), 122.
3. Pokhrel R.P., Wagner, N.L., Langridge J.M., et al. Parameterization of single-scattering albedo (SSA) and absorption Ångström exponent (AAE) with EC / OC for aerosol emissions from biomass burning. *Atmos. Chem. Phys.*, **2016,** 16, 9549–9561.
4. Yokelson, R.J., Crounse, J.D., DeCarlo, P.F., et al. Emissions from biomass burning in the Yucatan. *Atmos. Chem. Phys.*, **2009**, 9, 5785–5812.
5. Mikhailov, E., Mironova, S., Mironov, G., et al. Long-term measurements (2010–2014) of carbonaceous aerosol and carbon monoxide at the Zotino Tall Tower Observatory (ZOTTO) in central Siberia. *Atmos. Chem. Phys. Discuss.*, **2017**, doi:10.5194/acp-2017-409.