Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-184-RC1, 2016

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Parameterization of Single Scattering Albedo (SSA) and Absorption Angstrom Exponent (AAE) with EC/OC for Aerosol Emissions from Biomass Burning" *by* Rudra P. Pokhrel et al.

Anonymous Referee #3

Received and published: 15 April 2016

This paper presents results from the FLAME IV measurement campaign, in which a range of biomass types, the emissions from which are of great atmospheric relevance, were burned under controlled conditions and monitored by a number of groups. This paper describes measurements of the optical properties of emitted particles (absorption and scattering at multiple wavelengths) along with complementary measurements of organic and elemental carbon (OC/EC), CO and CO2 during these burns. Optical measurements yield estimates for aerosol single scattering albedo (SSA) and absorption angstrom exponent (AAE), which are commonly used in atmospheric models and satellite data retrievals. Previous efforts have shown that the modified combustion ef-

ficiency of a combustion event can be linked to the SSA of the emitted particles. The paper proposes a different relationship that relates SSA and AAE to the EC/OC or EC/(OC+EC) ratios and show that this relationship has better predictive power than that based on MCE over a wider range of conditions. In particular, the MCE-based parameterizations tend to do a poor job at high MCEs (>0.92), which are characteristic of more complete, typically flaming-phase combustion, and also potentially at low MCEs, as fits are distorted due to the prevalence of higher MCE conditions. The OC/EC based parameterizations result in improved fits of the optical properties of particles emitted during combustion of the wide range of fuels. Based on this, the authors suggest that these parameterizations can improve modelers' abilities to represent particle optical properties for both fresh and potentially aged emissions. To support the latter, they include an application of the method to data collected during measurements within a large biomass burning plume, which suggest that the parameterization can be used to track optical properties in an evolving plume.

Improving our understanding of the optical properties of biomass burning emissions is an important goal, and this paper makes a substantial contribution to that, and suggests a way in which available data can be leveraged to improve the representation of particles in models and satellite retrievals. While the work is suited for eventual publication in ACP, there are a number of issues that should be more comprehensively addressed before it is published. In general, the paper is well-written and clear, and should be a valuable contribution to ACP once these issues are addressed.

Major points:

A key concern is with the manner in which OC was quantified, and the potential influence of measurement artifacts on its determination. While the authors offer some discussion of the potential influence of gas-particle partitioning on the observed OC levels, they make no mention of the influence of gas-phase artifacts on quartz-fiber filters, which were used for determination of OC. It has long been known that positive artifacts can contribute the majority of OC mass in some cases (Subramanian et al.

2004). The fact that OC measurements were collected on bare quartz filters, means that there will be a substantial gas-phase artifact, which likely has a larger high-biasing effect on filters with lower OC levels. It is well established at this point that OC from biomass combustion has a large contribution from semivolatile materials, that will partition based on environmental conditions and also readily lead to positive sampling artifacts on quartz filters (Grieshop et al. 2009; Lipsky and Robinson 2006; May et al. 2013). While it is not possible for the extent of this artifact to be measured now (assuming no other groups have done so) an estimate for this effect could be determined using the volatility parametrization from May et al, collected during Flame III. This would also enable a quantitative examination of the impact of dilution on comparison between burns, which was dismissed as unimportant in a not-convincing way (P4, L7-13). Levels of dilution can have substantial effects on the partitioning of organics (See e.g. Fig. 6 in (May et al. 2013)) and since your parameterization is a direct function of OC, it is important to eliminate any biases in the measurements. Overall the authors should be applauded for careful uncertainty analysis, which is too often ignored, but the influence of these processes/artifacts should be included in this analysis. Such artifacts present a complication to the application of this kind of parameterization because both OC and EC measurements are operationally defined and sensitive to sampling conditions. Sampling conditions (average sample concentrations) should be included in the supplement, and efforts made to remove biases resulting from different sampling conditions.

A general comment on the paper is that a number of real-time properties of emissions were characterized at 1 Hz, but only ‘burn-average’ properties discussed. It would be interesting to see how these properties evolved for individual burns as it progressed, as in many cases there will be distinct phases with different properties, and the relative prevalence of these different properties may be very different for the same fuels in different conditions.

It would be helpful if confidence intervals were provided on regression fits.

One important detail left out of the final section, comparing results from the (Yokelson et al. 2009) study, is that ‘PM2.5’ and ‘BC’ in this study were both determined optically (via nephalometer and PSAP, respectively). It is therefore a bit circular to use these to show that a parameterization based on chemical measurements can be used to represent optical properties. The ‘calibration’ of the PM/BC measurements in Yokelson et al does provide an indirect link to ‘mass’ measurements, but they are still optical measurements. This still may be a useful example of the applicability of your result, but needs to be used with proper caveats. To properly do uncertainty analysis on this, the uncertainty in the other assumptions (MAE, MSE) used to estimate BC and PM should also be included.

Minor Points

P3, L10-12 – This sentence is confusing. It makes it sound as if Indonesian Peat is the largest source of organic carbon on the ground (terrestrial). Also, combustion of peat is a varying source and I don’t know if a statement so strong is justified. P4, L6 – Pretty sure you can’t put Perma-pure in a canister this way? Did you use a Nafion dryer? Maybe thinking of some other compound? P4, L13-15 – This is a circular argument unless you have some a priori reason that the quantities you are comparing should have robust correlations. P5, L27 – clarify what is meant by excess P7, L4 – missing a word here, perhaps ‘is’? P8, L8-9 – It is not stated what the chosen functional form is, and why it was chosen. P8, L20 – much of this strong correlation is driven by the fact that there are two clusters of data that are widely spread, through which a line can be drawn. An exponential-type curve could also be driven, and might asymptote at a more reasonable value as EC/TC goes to higher values. P8, L24-26 – This sentence is awkward and difficult to understand. P 9, L15 – it would be helpful if some of this comparison were made graphically, either as a separate plot, or by adding some/all of these points on existing plots P9, L21-22 – Is this really all that can be said about this? If this is the case, it’s really not clear whether it is worth including a table, especially in the main paper. If no systematic point can be made, put the table in the SI and just include a range of differences. P9, L23 – Extra word? P9, L24-25 – Worth a discussion if you are saying your proposed parameterizations won't be applied in models. This is your original motivation – why would it only be used in this limited way if it is so much better than the alternatives? P10, L12-13 – sentence fragment. This should be quantified: 'much worse'. EC-dominated combustion is more common in biofuel use, so may still be an issue. P10, L26 – Any suggestions as to why the AAEs determined for the same burns are so different? P10, L27-28 – There should be a reference to a source with these data. Also, seems to be a missing word in this sentence. P11, L9 – Would be good to mention the physical significance of this intercept. Table 5 –should be no more than 2 significant figures in % difference column

References

Grieshop, A. P., Miracolo, M. A., Donahue, N. M., and Robinson, A. L. (2009). "Constraining the Volatility Distribution and Gas-Particle Partitioning of Combustion Aerosols Using Isothermal Dilution and Thermodenuder Measurements." Environmental Science & Technology, 43(13), 4750–4756.

Lipsky, E. M., and Robinson, A. L. (2006). "Effects of dilution on fine particle mass and partitioning of semivolatile organics in diesel exhaust and wood smoke." Environmental Science & Technology, 40(1), 155–162.

May, A. A., Levin, E. J. T., Hennigan, C. J., Riipinen, I., Lee, T., Collett, J. L., Jimenez, J. L., Kreidenweis, S. M., and Robinson, A. L. (2013). "Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning." Journal of Geophysical Research: Atmospheres, 118(19), 2013JD020286.

Subramanian, R., Khlystov, A. Y., Cabada, J. C., and Robinson, A. L. (2004). "Positive and negative artifacts in particulate organic carbon measurements with denuded and undenuded sampler configurations." Aerosol Science and Technology, 38, 27–48.

Yokelson, R. J., Crounse, J. D., DeCarlo, P. F., Karl, T., Urbanski, S., Atlas, E., Campos,

T., Shinozuka, Y., Kapustin, V., Clarke, A. D., Weinheimer, A., Knapp, D. J., Montzka, D. D., Holloway, J., Weibring, P., Flocke, F., Zheng, W., Toohey, D., Wennberg, P. O., Wiedinmyer, C., Mauldin, L., Fried, A., Richter, D., Walega, J., Jimenez, J. L., Adachi, K., Buseck, P. R., Hall, S. R., and Shetter, R. (2009). "Emissions from biomass burning in the Yucatan." Atmos. Chem. Phys., 9(15), 5785–5812.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-184, 2016.