We would like to thank the reviewer for their valuable suggestions and time. Our responses are given below. We believe the page and line numbers in the reviewer's comments were based on the manuscript that was initially submitted and was modified before publication in ACPD. This resulted in difference between line and page numbers in the comments and the current version of the manuscript in ACPD. Accordingly, we have removed the reviewer's line and page references and inserted the correct page and line numbers to avoid confusion.

Anonymous Referee #1 Received and published: 16 April 2016

Referee Comment: Which parameter is more easily and accurately available for fires on regional and global scales? MCE or the amount of BC and BB OC (or OA)? Since OA/CO ratios for fires are quite variable and hard to predict in models, which SSA parameterization would then lead to the least uncertainty in the radiative effects of wildfire aerosols? See also my specific comment below.

Author Response: While published emission factors for biomass burning are relatively rare, both MCE and BC/OC (or OA) emissions factors are available (Akagi et al., 2011). The emission factors are vegetation (ecosystem) average values making the applicable to regional/global scales. The emission factors for BC/OA have been implemented in GEOS-CHEM (Saleh et al., 2015). While BC/OC (or OA) ratio is difficult to accurately predict in models, it is often easier to keep track of this ratio than MCE for aerosol because many models keep track of aerosol and gas-phase emissions separately. Neither BC/OC (or OA) or MCE can be accurately measured by satellites.

Referee Comment: MCE, EC/OC, and EC/(OC+EC) all have uncertainties/errors associated with them, so the fits are more appropriate if they're ODR with uncertainties as weights of the fit and not least-square linear regression lines as is done throughout the paper. (This is the main reason for my rating of the paper as 'major' revisions since all the figures, tables, and numbers in the text are to be updated accordingly).

Author Response: We agree with the referee that MCE, EC/OC, and EC/(EC+OC) all have uncertainties/error associated with them. Because we are comparing the predictive capabilities of a previously published MCE-based parameterization (that was done with least-squares regression) proposed by Liu et al. with our EC/OC based parameterization, we have decided to do most of the fitting with least-squares regression to compare apples to apples. Additionally, the fits for SSA vs. MCE and SSA vs. EC/OC are non-linear which makes applying least squares regression or ODR of fitting more complex. Given this, we have decided not to change the MCE or EC/OC fits to ODR, but we have made fits for SSA vs. EC/(EC+OC) with ODR and added the results to the SI. We also made a figure comparing the predicted SSA values based on SLR and ODR regression (as explained in section 3.5) and added this in the SI. The predicted SSA values based on SLR are slightly larger than ODR, but the difference is not statistically significant. A two-tailed p value of 0.748 when a two sample t test was performed with the results from both fits. We also add following text in section 3.2 and 3.5

Added Text Location: section 3.2 after the sentence "The y-intercepts of the fits….

Added Text: Fig. S1 shows that regression lines based on a simple liner regression (SLR) model and orthogonal distance regression (ODR) model are fairly similar at wavelengths of 660 and 532 nm for SSA vs. EC/(EC+OC). The regression lines for SSA vs. EC/(EC+OC) at 405 nm show significant deviations when fitted with the SLR vs. ODR methods, especially at higher EC/(EC+OC) ratio. This difference may be due to less data points in that region. Similarly, regression lines for AAE shows larger deviation between SLR and ODR methods at lower EC/(EC+OC) values, possibly due to less data points. ODR-based fits are provided for those who prefer this regression technique.

Added Text Location: section 3.5 after the sentence "This result suggests that the..

Added Text: Similarly, SSA values are predicted based on an ODR model and compared with the SSA predicted by an SLR model. Figure S2 shows the comparison of SSA predicted based on SLR vs. ODR models. The general trend shows that the predicted SSA based on SLR is higher than that based on ODR, but that difference is not statistically significant. We performed a two-tailed t test with the null hypothesis that predicted values are the same from both regression models and found a two-tailed p value of 0.748 at 532 nm.

Referee Comment: P4, L8: The authors discuss possible evaporation of semi-volatile components after sample dilution, but another factor is temperature differences in the relatively long sampling line. What was the temperature of the sampling lines kept at? If it wasn't controlled, how does this temperature difference impact redistribution of semi-volatile components of aerosols?

Author Response: The sampling line was at the room temperature of the lab and there was no temperature control. This is expected to have no impact on room-burn results because emissions were already cooled to room temperature. Stack emissions had also cooled to near-room temperature by the time they reached the top of the stack, though the exact temperature depends on the amount of biomass burned. The fact that fire integrated values of SSA and AAE from stack burns are nearly identical to the room burn values for similar burn conditions, strengthens the argument that neither dilution or the temperature of the sampling line significantly altered the optical properties.

Referee Comment: P4, L28: How is the response of activated carbon monolith to gaseous organic species? Could this sample treatment introduce negative artifacts in organic aerosols?

Author Response: It is possible that the activated carbon utilized to scrub NO_x and ozone could introduce a small negative artifact by removing some volatile organics from the gas-phase and causing aerosol evaporation. However, this is expected to be a negligibly small effect given that the emissions were already diluted into the very large combustion room and had equilibrated to this highly-diluted environment. Again, the fact that nearly identical results were obtained for room-burn (highly diluted) and stack burns (less diluted) strongly suggests that the inlet is not introducing significant artifacts.

Referee Comment: P5, L27: What does 'excess' extinction and absorption mean? I'm not sure how SSA/AAE for stack burns in calculated- I thought it should be just based on average values of abs and ext, but not sure what summing up the 'excess' amounts mean.

Author Response: Excess means above background. We have changed the word "excess" to "background corrected" at P5, L27 and also at P5, L30. SSA/AAE were calculated based on the fire integrated absorption and extinction values during a stack burns. Fire-integrated means you add up all the absorption and extinction that occurred during the course of the burn. A straight time average is misleading because, in a stack burn, SSA and AAE vary by large amount from the flaming dominated part of burn to the smoldering dominated part. A fire may smolder for a long time, but a small fraction of mass emissions occurs during this time so an average value will be biased for those burns which have a shorter flaming period (few second) and a longer smoldering period (couple of minutes). This kind of measurement is common in instantaneous measurement during control burn in laboratory study (McMeeking et al., 2009; Liu et al., 2014; Stockwell et al., 2014).

Referee Comment: P6, Section 3.1: I agree that the data suggest EC/OC ratios are more variable for a given fuel than MCE and therefore relationships of optical properties with EC/OC are more robust. Can the authors elaborate on what specifically changes in the 'burn condition' that leads to such variability in aerosol characteristic? Is the different in the water content of the fuel or the starting temperature of the fire, etc. etc?

Author Response: There are a large number of parameters that determine the EC/OC ratio for a given burn. Some parameters are the surface area to mass of the fuel (stick vs. log vs. pine needle), the way the fuel is stacked or layered, the moisture content of the fuel, and the nature of the fuel (grass vs. wood). Our goal in this paper is not to describe what caused a given EC/OC ratio, for that type of information one must reference an emission factor paper such as Akagi et al., 2011.

Referee Comment: P9, Section 3.4: Can you perform a simple calculation to estimate instantaneous TOA forcing difference of BB aerosol depending on the choice of SSA (new vs. old parameterization) so readers get an idea about the magnitude of the change in forcing? This calculation should be done considering the uncertainties in MCE, EC/OC (or EC/(OA+EC)) and SSA.

Author Response: Unfortunately, there is no easy way to do a simple calculation of this type. Given that different fuels have different EC/OC emissions and that the EC/OC emission depends on moisture etc., one would need an inventory of global fuels, to couple this inventory to the amount of burning that occurs in different regions, then inject the particles in the atmosphere at the appropriate heights and remove them at appropriate rates. This is a complex problem that requires a global model of some type. In general, if SSA drop from 1 to 0.9, TOA forcing can drop by 50 to 100% depending upon the surface albedo (Russell et al., 2002). Saleh et al. (2015) have recently calculated that different parameterizations can significantly change the TOA forcing from biomass burning utilizing GEOS-Chem.

Referee Comment: P9, L24: I'm confused by the statement "While climate models may not directly parameterize optical properties based on EC/OC, the parameterization provides a good sanity check of model schemes to predict optical properties." If models have EC/OC data to do this sanity check, do the authors not recommend the modelers to use this parameterization instead of other estimates? If yes, I think the sentence needs to be rephrased. If not, elaborate why this shouldn't be the recommended approach.

Author Response: The statement will be changed. We will now say, "Because climate models need to mix different emission types, track SSA with extensive aging, and track particle losses, we anticipate that climate models will need parameterizations that include particle-size and refractive index and will not directly implement the parameterizations presented here. However, these parameterizations provide a critical tool to assess if a model implementation, based on assumptions about refractive index and coating thicknesses (Saleh et al., 2015), generates reasonable SSA estimates."

Referee Comment: P10, L21-24: I do understand that the average SSA of peat burning aerosol is lower, but given the uncertainty for the SSA values, the difference at 532 nm vs. 405 nm is not really significant. The relatively high AAE is more convincing for the presence of BrC in peat burning. What is the uncertainty in AAE for this sample? Add that value as well.

Author Response: The uncertainty for the AAE has been added. In terms of uncertainty in SSA, we have modified the error stated to be the error in the mean rather than the error of an individual measurement, which was originally quoted. This makes the difference between 532 and 405 nm more significant.

Referee Comment: P10, Line4: I question the assumption of PM2.5 in a fire being composed of only BC and OM. In most fires, there could also be aerosol nitrate and chloride. How does the ratio of EC/(EC+OC), and therefore, estimate of SSA change if say 5-15% of PM2.5 is assumed to be inorganics? Also, looking at Table 3 in Yokelson et al., ACP 2009, there were direct PM1-OM measurements. Why not use that measure of OM when calculating OC?

Author Response: We agree with the referee that $PM_{2.5}$ in a fire is not composed of only BC and OM. We have modified our calculation to assume 39 % of $PM_{2.5}$ is OC as estimated by Yokelson et al. (2009) during the study. We have corrected the text and updated Fig. 5 with this assumption. The parameterization performs well with this new assumption and no further adjustments to the text were needed.

Added text location: Section 3.5

Added Text: **ΔBC/ΔPM$_{2.5}$** was converted in to EC/(EC+OC) by setting the OC mass fraction to 39 ±9 % of the PM$_{2.5}$ as stated by Yokelson et al. (2009). This calculation assumes BC and EC mass are identical.

Referee Comment: Additionally, BC and EC are not necessarily presenting the same type of species. Can you reference papers that perform both measurements on a series of burned fuels and comment on the 'goodness' of this assumption and how it will impact the predicted SSA?

Author Response: We agree BC and EC are not necessarily presenting the same type of species. But EC is often used as surrogate of BC. We have cited Salako et al. (2012) in our manuscript which state that, for a burn with 82 % biomass burning emissions, 17% diesel emissions, and 1% other, BC = 1.06*EC with R^2 = 0.91. Based on this, our EC/(EC+OC) parameterization and the predicted SSA and AAE will be not significantly different if BC is utilized instead of EC.

Minor Comments:

Referee Comment: P2, L7 primary OA=POA

Author Response: We have changed primary OA to POA

Referee Comment: P3,L16: 'effectiveness of our . . .'

Author Response: We have changed the sentence to, "We also show that predicted SSA based on the EC/(EC+OC) parameterization is similar to measured SSA during the first few hours of aging from the Yucatan peninsula in Mexico (Yokelson et al., 2009)."

Referee Comment: P7, L4-5 "At high MCE, AAE is ~1 because BC absorption proportional to frequency" incomplete sentence. Also, by frequency, do you mean 'wavelength'?

Author Response: We have modified the sentence, it now reads, "At high MCE, AAE is ~1 because BC dominates absorption"

Referee Comment: P7, L5-6: should clarify that low MCE burns give high SSA at long wavelengths, since in the following sentences it's mentioned that the OA in low MCE burns is highly absorbing as BrC.

Author Response: We have changed the sentence to, "In contrast, fuels that burn with low MCE are dominated by OC emissions, which predominantly scatter light at long wavelengths resulting in SSA values nearing unity at 532 and 660 nm and larger values of AAE".

Referee Comment: P9, L18: consider ". . ..how significant of an impact. . ."
Author Response: We have modified the sentence as suggested by referee.

Referee Comment: P9, L23: during . . .? Incomplete sentence
Author Response: We have deleted the extra word "during".

Referee Comment: P10: Indonesian Peat section should be 3.6
Author Response: Indonesian Peat section in now 3.6

Referee Comment: P10, L12: What's the explanation for the parameterization not capturing the measured SSA in plumes with lower SSA? Also, start the sentence with "However" instead of "But", or combine the two sentences

Author Response: This could be a limitation of our parameterization to parameterize burns with extremely high EC (EC/OC much greater than unity). Vakkari et al note that these "dark" plumes are rarely observed in the atmosphere. We change the sentence as "But the predicted values for "dark" plumes are consistently larger than measured values by about 35% on average.

Referee Comment: P10, L27-28: Rephrase the sentence, sounds like summary bullets and not a complete sentence.
Author Response: We change the sentence as "Other peats (North Carolina, Canadian) produced aerosol with similar optical properties to Indonesian peat (values can be found in Table) have less impact on the global radiative budget.

Referee Comment: P11, L2: When giving the range of SSA, separate it for 405 nm vs. 660 nm.
Author Response: We now report SSA ranges for both 405 nm and 660 nm.

Referee Comment: P11, L11-12: Indicate again the errors bars for SSA of peat aerosol at 405 nm and longer wavelengths as well as the error bar for AAE.
Author Response: Errors bars are included for peat aerosol on P11, line 11-12.

References:

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D. and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, doi:10.5194/acp-11-4039-2011, 2011.

Liu, S., Aiken, A. C., Arata, C., Dubey, M. K., Stockwell, C. E., Yokelson, R. J., Stone, E. a, Jayarathne, T., Robinson, A. L., Demott, P. J. and Kreidenweis, S. M.: Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, Geophys. Res. Lett., 41, 742–748, doi:10.1002/2013GL058392, 2014.

McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collett, J. L., Hao, W. M., Holden, A. S., Kirchstetter, T. W., Malm, W. C., Moosmüller, H., Sullivan, A. P. and Cyle E., W.: Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory, J. Geophys. Res., 114, D19210, doi:10.1029/2009JD011836, 2009.

Russell, P. B., Redemann, J., Schmid, B., Bergstrom, R. W., Livingston, J. M., McIntosh, D. M., Ramirez, S. a., Hartley, S., Hobbs, P. V., Quinn, P. K., Carrico, C. M., Rood, M. J., Öström, E., Noone, K. J., von Hoyningen-Huene, W. and Remer, L.: Comparison of Aerosol Single Scattering Albedos Derived by Diverse Techniques in Two North Atlantic Experiments, J. Atmos. Sci., 59(3), 609–619, 2002.

Salako, G. O., Hopke, P. K., Cohen, D. D., Begum, B. A., Biswas, S. K., Pandit, G. G., Chung, Y. S., Rahman, S. A., Hamzah, M. S., Davy, P., Markwitz, A., Shagjjamba, D., Lodoysamba, S., Wimolwattanapun, W. and Bunprapob, S.: Exploring the variation between EC and BC in a variety of locations, Aerosol Air Qual. Res., 12, 1–7, doi:10.4209/aaqr.2011.09.0150, 2012.

Saleh, R., Marks, M., Heo, J., Adams, P. J., Donahue, N. M. and Robinson, A. L.: Contribution of brown carbon and lensing to the direct radiative effect of carbonaceous aerosols from biomass and biofuel burning emissions, J. Geophys. Res. Atmos., 120, doi:10.1002/2015JD023697-T, 2015.

Stockwell, C. E., Yokelson, R. J., Kreidenweis, S. M., Robinson, A. L., DeMott, P. J., Sullivan, R. C., Reardon, J., Ryan, K. C., Griffith, D. W. T. and Stevens, L.: Trace gas emissions from combustion of peat, crop residue, biofuels, grasses, and other fuels: configuration and FTIR component of the fourth Fire Lab at Missoula Experiment (FLAME-4), Atmos. Chem. Phys., 14, 9727–9754, doi:10.5194/acp-14-9227-2014, 2014.

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.: Emissions from biomass burning in the Yucatan, Atmos. Chem. Phys., 9, 5785–5812, doi:10.5194/acp-9-5785-2009, 2009.

We would like to thank the reviewer for their valuable suggestions and time. Our responses are given below. We believe the page and line numbers in the reviewer's comments were based on the manuscript that was initially submitted and was modified before publication in ACPD. This resulted in difference between line and page numbers in the comments and the current version of the manuscript in ACPD. Accordingly, we have removed the reviewer's line and page references and inserted the correct page and line numbers to avoid confusion.

Anonymous Referee #2 Received and published: 11 May 2016

Referee Comment: My main question at this point relates to their choice of r as the figure of merit in their (non-linear) fitting.

Author Response: We agree that r is not a good parameter to assess a non-linear fit. This is why we have given RMSE in Table 3, which we believe is a better parameter for the non-linear fits. We have given r for the non-linear fits because it has been stated in previous publications with which we are comparing. We do believe r is relevant for the EC/(EC+OC) parameterization, which is linear.

Referee Comment: P1, L17: Suggest replacing "significant" with "substantial" or "important" so as not to imply statistical significance.

Author Response: We have changed the word "significant" to "substantial".

Referee Comment: P1, L18: Suggest replacing "inferred" with "predicted".

Author Response: We have changed the word "inferred" to "predicted".

Referee Comment: P1, L20: I find ": : :emission factors for the MCE: : :" to be unclear. EFs of what?

Author Response: We have changed the sentence to read, "It has been suggested that SSA can be effectively parameterized via the modified combustion efficiency (MCE) of a biomass-burning event and that this would be useful because emission factors for CO and CO_2 from which MCE can be calculated are available for a large number of fuels".

Referee Comment: P1, L27 and General Question: Pearson's r is a parameter that describes the linear correlation between two variables. Here, it seems to be applied to one data set that is linearly related (SSA vs. EC/(EC+OC)) and two that are not (SSA vs. MCE and SSA vs. EC/OC). Thus, are the r values really comparable? What does an r value mean for a non-linear relationship? Might a different statistical test be applied? Perhaps Spearman's or Kendall's rank correlation coefficients or a Pearson's Chi Square test?

Author Response: We agree with the referee that Pearson's r gives the measure of linear dependency between two variables and is not very meaningful for nonlinear relationship. However, a key focus of this paper is to compare the predictive capabilities of MCE and EC/OC. A MCE parameterization for SSA (nonlinear relation with SSA) was already published (Liu et al., 2014) and concluded that based on R^2 value that MCE can explain 60% in variability in SSA. To

show EC/OC is better than MCE we also made r value comparison because the article that we are comparing uses r values. Because we recognize the fault in utilizing r or R^2, we also compare the root mean square error (RMSE) of different approaches which is much more useful in comparing the model predictive capabilities. Even though, r value for nonlinear relationship is a rather poor assessment of fit, there are numerous publications that give this result in recent years too (Liu et al., 2014; Lu et al., 2015; Cui et al., 2016).

Referee Comment: P2, L9: The authors cite Stier (2007) as evidence that "most climate models treat organic carbon as purely scattering." However, it is clear from Fig. 1 in Stier (2007) that the OC is somewhat absorbing throughout the visible. In fact, most models treat OC as slightly absorbing (see e.g. the OPAC database).

Author Response: We appreciate the referee's effort in pointing out this improperly cited source and overstatement. We changed the sentence as "Although some climate model treat organic carbon (OC) as purely scattering (Myhre G et al., 2007), OC….".

Referee Comment: P2, L13: The inclusion of the reference to Washenfelder here seems quite selective, as there are many regions where biomass burning has been implicated as a source of BrC. Not that it is not a nice study, but is there a reason why this study is being highlighted?

Author Response: We agree with the referee that biomass burning has been implicated as a source of BrC. But there is significant uncertainty regarding the relative contribution of BrC by different sources (biomass burning, SOA, fossil fuels) in many regions. Washenfelder et al. found that biomass burning is the dominant source of BrC in southeastern US this is the reason why this study is highlighted.

Referee Comment: P2, L17: I suggest that the Saleh reference is removed and only the Feng (global model result) reference is retained.
Author Response: Reference is removed.

Referee Comment: P2, L23: What is meant by SSA and AAE are "commonly implemented in models"? Models don't specify SSA. Similarly, what is meant by "SSA and AAE are also critical for satellite retrievals"? Critical for or are important retrieved information from?

Author Response: The sentence has been modified to read, "Single scattering albedo (SSA) and absorption angstrom exponent (AAE) are commonly used parameters that contain the necessary information on aerosol absorption and scattering to calculate radiative effects".

Referee Comment: P3, L9: No reference to Salako et al. is provided. Also, I would contend that this really remains to be demonstrated as "charring" is known to be a particularly important for

biomass burning. Also, the authors might compare their longest wavelength denuded-particle absorption measurements to the EC measurements to argue that there is a reasonable relationship between BC and EC for this data set.

Author Response: We have inserted the reference to Salako et al. We agree there is some difficulty in equating EC to BC and this is something that needs to be further examined. Because converting our 660 nm absorption to BC would require assuming a MAC, we do not pursue this.

Referee Comment: P7, L4: The sentence starting "At high MCE" is a fragment.

Author Response: We have modified the sentence, it now reads, "At high MCE, AAE is ~1 because BC dominates absorption

Referee Comment: Fig. 1: It seems odd that the least squares fit (red line) doesn't match the data at smaller MCE values at 660 nm (most notably). Is there a reason for this? The functional form used (which should be given in the main text as well) should allow for better agreement at these low MCE values. Also, it is unclear if the fits were performed with/without accounting for the uncertainties in the individual points.

Author Response: A discussion of the reason for the mismatch between fit and data at smaller MCE has been added on Page 7 line 19.

Referee Comment: Fig. 3: The fits the authors retrieve allow for unphysical SSA values > 1. I suggest that they redo their fits, constraining the maximum retrievable SSA to be <= 1. This amounts to constraining the k0 in their fit equation to be <= 1. This links to P8,L14, where the authors note that this fitting does lead to SSA values > 1. But this is a solvable problem. Physical realism can be imposed on the fits.

Author Response: This issue has been corrected by constraining the fits to have a maximum SSA of 1. Text has been added on Page 8 line 14.

Referee Comment: P8, L5: the authors might indicate what they consider the EC/OC value at which composition is "dominated" by EC.

Author Response: The sentence has been modified to read, "When the composition has equal or more EC than OC, there is less dependence of SSA on wavelength and AAE values are less than 2".

Referee Comment: P8, L9: horizontal should be vertical.

Author Response: Horizontal has been changed to vertical.

Referee Comment: P8, L17: Are the data truly more "bunched" or is the difference that Fig. 4 uses a linear scale and Fig. 3 a log scale for the x-axis? I think the latter.

Author Response: We agree with the referee that the effect is due to the use of linear vs log scale. We removed the sentence starting "The simple nature of the linear……"

Referee Comment: P8, General: The authors discuss the robustness of their fits and the ability of EC/(EC+OC) to be used as a predictor. Although I generally agree, a few thoughts: (i) I think that the authors are overstating the case for AAE, as the correlation coefficient is only 0.79. (ii) Regarding the 405 nm measurements, yes, the fit gets a <1 SSA value when EC/(EC+OC) = 0. But it is also clear that the zero intercept here differs substantially from the data points. In other words, the fit is certainly "good" but the model fit and observed SSA values differ by 0.03 or more, which is small yet non negligible. (iii) Can the authors include confidence bands?

Author Response: (i) We agree with the referee that fitting with AAE is not very good. All we mean to say is that we can predict SSA and AAE with the value of EC and OC without the need to reference other properties like size distribution, lensing effect, presence of BrC. We believe including the r for the AAE fit is adequate to show that the fit is significantly weaker than that for SSA. (ii) we absolutely agree with the referee that there is uncertainty here. (iii) a 95% CI for fitting is included for the parameters of the fit. We feel adding the 95% confidence intervals to the plot would make the plot too busy.

Referee Comment: Table 5 and discussion: Do the MCE and EC/OC from the literature for biomass burning emissions generally agree with the observations here in terms of functional form?
Author Response: The values of MCE and BC/OC values from the literature are in the same range that we observed in this study. Also, the nature of BC/OC vs MCE from the literature follow a similar pattern to that shown in Fig. 2 of the main text).

Referee Comment: P9, L24: I find the meaning of the following sentence to be unclear: "While climate models may not directly parameterize optical properties based on EC/OC, the parameterization provides a good sanity check of model schemes to predict optical properties." Can the authors clarify how this table and discussion provides a "sanity check"?
Author Response: The statement will be changed. We will now say, "Because climate models need to mix different emission types, track SSA with extensive aging, and track particle losses, we anticipate that climate models will need parameterizations that include particle-size and refractive index and will not directly implement the parameterizations presented here. However, these parameterizations provide a critical tool to assess if a model implementation, based on assumptions about refractive index and coating thicknesses (Saleh et al., 2015), generates reasonable SSA estimates."

Referee Comment: P10, L10: What is meant by "reasonably good?" As good as the case that is shown? Can this just be shown?
Author Response: We state "the predictions are within roughly ± 5%..." which is what we describe to be reasonably good. We were not able to obtain exact values of SSA at 637 even though we contacted Vakkari et al. The analysis is based on close inspection of their published figures with a program that converts figures to numerical values. We would not feel comfortable publishing exact values without the approval of Vakkari et al.

Referee Comment: P10, L18: If the peat burning was unintentional and a result of e.g. drought, I suggest the authors say "through unintentional peat burning."
Author Response: Indonesian peat burning are mostly anthropogenic (Bompard et al., 1999) but the article that we cited was based on 1997 El Nino event.

Referee Comment: P7, L6: To set things up for later in the paper, the authors might report the mean value for peat here in addition to the maximum. Some discussion of the variability would also be helpful (later in section 3.4).

Author Response: We believe it is most efficient to state this once in Section 3.4 that discusses Indonesian Peat.

Referee Comment: P10, L27: this is a sentence fragment.
Author Response: We change the sentence as "Other peats (North Carolina, Canadian) produced aerosol with similar optical properties to Indonesian peat (values can be found in Table have less impact on the global radiative budget".

Referee Comment: General: I suggest that the authors adopt the terminology "aerosol particles" through-out much of the particle, to indicate that they are looking at the particulate matter and not the associated gaseous material.

Author Response: The title of the article clearly states that this article is about aerosol emissions. We have also modified the first sentence of the results and discussion to read, "Single scattering albedo (SSA) and absorption angstrom exponent (AAE) of aerosol emissions were measured during 41 individual burns of twelve different fuels during FLAME-4

References:

Bompard, J. M. & Guizol, P.: Land Management in South Sumatra Province, Indonesia. Fanning the Flames: The Institutional Cause of Vegetation Fires (European Union Forest Fire Prevention and Control Project and IndonesianMinistry of Forestry and Estate Crops, Jakarta, 1999).

Cui, X., Wang, X., Yang, L., Chen, B., Chen, J., Andersson, A. and Gustafsson, Ö.: Radiative absorption enhancement from coatings on black carbon aerosols, Sci. Total Environ., 551-552, 51–56, doi: 10.1016/j.scitotenv.2016.02.026, 2016.

Liu, S., Aiken, A. C., Arata, C., Dubey, M. K., Stockwell, C. E., Yokelson, R. J., Stone, E. a, Jayarathne, T., Robinson, A. L., Demott, P. J. and Kreidenweis, S. M.: Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, Geophys. Res. Lett., 41, 742–748, doi:10.1002/2013GL058392, 2014.

Lu, Z., Streets, D. G., Winijkul, E., Yan, F., Chen, Y., Bond, T. C., Feng, Y., Dubey, M. K., Liu, S., Pinto, J. P. and Carmichael, G. R.: Light Absorption Properties and Radiative Effects of Primary Organic Aerosol Emissions, Environ. Sci. Technol., 49, 4868–4877, doi: 10.1021/acs.est.5b00211, 2015.

Saleh, R., Marks, M., Heo, J., Adams, P. J., Donahue, N. M. and Robinson, A. L.: Contribution of brown carbon and lensing to the direct radiative effect of carbonaceous aerosols from biomass and biofuel burning emissions, J. Geophys. Res. Atmos., 120, doi:10.1002/2015JD023697-T, 2015.

Vakkari, V., Kerminen, V.-M., Beukes, J. P., Titta, P., Zyl, P. G. van, Josipovic, M., Wnter, A. D., Jaars, K., Worsnop, D. R., Kulmala, M. and Laakso, L.: Rapid change in biomass burning aerosols by atmospheric oxidation, Geophys. Res. Lett., 2644–2651, doi:10.1002/2014GL059396, 2014.

We would like to thank the reviewer for their valuable suggestions and time. Our responses are given below.

Anonymous Referee #3 Received and published: 15 April 2016

Referee Comment: 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.

Author Response: We agree with the reviewer that gas sorption onto quartz fiber filters (QFF) can introduce positive sampling artifacts. To assess such artifacts, back-up quartz fiber filters were collected behind Teflon filters during sample collection in FLAME-4. The quartz filters behind Teflon (QBT) adsorb gases only and thus serve as a measure of gases adsorbed on QFF during sampling (Cheng et al., 2009). For fourteen FLAME-4 burns, representing five different biomass types (grass, rice straw, pine, spruce, peat) gas sorption accounted for an average of 2.4 ± 1.2 % of the OC (ranging from below the instrument limit of detection to 4.7%). In light of the reviewer's comment, we have applied the artifact correction to this data set and describe this in the methods section of the text. All figures and data analysis in the revised manuscript are based upon artifact-correction data.

Added Text Location: Section 2.5, page 6, line 16

Added Text: The effects of positive sampling artifacts due to carbonaceous gas adsorption were assessed using quartz filters behind Teflon (QBT) (Cheng et al., 2009) for 14 of the 96 fires, including grass, rice straw, ponderosa pine, black spruce and peat. For fires with QBT collected, the OC on the backup filter was subtracted directly. For fires without backup filters or those that were below the detection limit, the average OC correction for that fuel type was applied: rice straw (2.0 ± 0.4 %), ponderosa pine (1.2 %), black spruce (2.9 ± 1.6 %) and peat (3.1 ± 0.8 %). For fuels types without backup filters collected, the study average OC artifact (2.4 ± 1.2 %) was subtracted.

Referee Comment: …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). Lev-els 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.

Author Response: We agree that the effect of dilution can be significant and have modified the text on P4, L7 to more explicitly address this issue.

Added Text Location: Section 2.1, page 4, line 4

Added Text: Smoke from the combustion room or stack intended for analysis by the optical

suite of the PAS and CRDS was diluted to achieve extinctions of approximately 500 Mm^{-1} or less to prevent signal saturation in the CRDS. Dilution flow was generated from ambient air by passing it through an active-charcoal and permanganate (Purafil) scrubber to remove gas phase absorbers (O_3 and NO_x) followed by a HEPA filter to remove particulates. Dilution air was introduced to the sample flow ~1 foot from the common inlet. All results presented in this paper explore intensive properties and thus are not sensitive to dilution unless significant evaporation of semi-volatiles occurred, which has been shown to be possible (May et al., 2013). All emissions, weather additionally diluted for optical measurements or not, experienced significant dilution before sampling. A wide range of dilutions are included in the dataset because widely different masses of fuel were burned during each individual burn (see SI Table 2 for details) and some burns were diluted into the combustion room while others were diluted into the much more compact combustion stack. Despite this wide range of dilution conditions, the parameterization of optical properties with EC/(EC+OC) ratio appears robust. It is important to note that this paper is not an attempt to say what the exact EC/OC ratio will be for a given fuel, as this may depend on dilution, but that if the EC/OC ratio is known at a given dilution then the optical properties can be predicted via the parameterizations presented. Accordingly, the authors urge some caution in utilizing EC/OC emission factors from emissions that are not adequately diluted to predict regional optical properties (Akagi et al., 2011).

Referee Comment: 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.

Author Response: We agree with the referee that optical properties are different at different phases of the burn, in fact they can be dramatically different during flaming vs. smoldering. During the measurements, we observed low SSA and AAE when the burn was dominated by flaming but high SSA and AAE when the burn was mostly smoldering. Given that a burn might smolder for a very long time, but that the later-phases of this long smoldering may represent a small fraction of total emissions, it was decided that the most useful approach was to reported burn integrated values (summed absorption, summed extinction). This allows us to compare room burns (which are naturally integrated) to stack measurements and gives the SSA of the sum of particles emitted during the burn. The focused of this paper is to parameterize SSA and AAE with MCE, EC/OC, and EC/(EC+OC). Since MCE and EC/OC were calculated as burn integrated, SSA and AAE were also reported in a similar manner. While observing changing SSA with burn phase is indeed interesting, it is not directly relevant to this paper.

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

Author Response: We have added confidence intervals on regression fits

Referee Comment: 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 nephelometer 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.

Author Response: We agree with the referee's comment. The purpose of this comparison was to test whether the parameterization can capture the effect of aging on SSA that was observed in different studies (Abel et al., 2003; Yokelson et al., 2009; Vakkari et al., 2014). Lack of availability of both EC/OC and SSA data of the aged biomass burning aerosol in these studies makes it difficult to check the performance of our parameterization during aging. This is why we used Yokelson et al. data even though PM/BC was determined optically. We have added following text in the document.

Added Text Location: section 3.5 after "instead of EC/(EC+OC) so several assumptions had to be made to implement our parameterization."

Added Text: One important note is that the $\Delta BC/\Delta PM_{2.5}$ reported in Yokelson et al. (2009) was derived from optical measurements.

Referee Comment: 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.

Author Response: We modified the sentence to read, "Tropical peatlands are one of the largest reservoirs of terrestrial organic carbon".

Referee Comment: 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?

Author Response: We change the sentence to read, "Dilution flow was generated from ambient air by passing it through an active-charcoal and permanganate (Purafil) scrubber to remove gas phase absorbers (O_3 and NO_x) followed by a HEPA filter to remove particulates".

Referee Comment: 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.

Author Response: This entire paragraph has been modified to clarify statements about dilution.

Referee Comment: P5, L27 – clarify what is meant by excess

Author Response: We change the sentence to read, "background corrected" at P5, L27 and also

at P5, L30.

Referee Comment: P7, L4 – missing a word here, perhaps 'is'?

Author Response: We have modified the sentence, it now reads, "At high MCE, AAE is ~1 because BC dominates absorption"

Referee Comment: P8, L8-9 – It is not stated what the chosen functional form is, and why it was chosen.

Author Response: The power law function was chosen because it gave a good fit and was consistent with the power law function utilized to parameterize with MCE**.** We have modified to the text to read, "Figure 3 demonstrates that a power-law parameterization of SSA with EC/OC yields a function."

Referee Comment: 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.

Author Response: We appreciate the reviewer's comment, but we slightly disagree with the statement "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". Since EC/TC and SSA can only vary from 0 to 1, EC/TC changes from 0.005 to 0.2 represent and SSA changes from 1 to 0.8 represent significant variations and the fit tracks the variations in SSA over this range of EC/TC well. Much of the atmospherically relevant biomass burning aerosol fall in this range of SSA. Additionally, if the X axis of the Figure 4 is plotted on a Log scale then the points spread out in a fashion very similar to Figure 3. We agree an exponential (or other functional forms) would also work but because the simple linear regression was equally accurate we did not proceed beyond this. Statistically, the simple model is best than any other complex form.

Referee Comment: P8, L24-26 – This sentence is awkward and difficult to understand.

Author Response: We have changed the sentence to read, "In fact, the robustness of the fits suggests that the EC/(EC+OC) ratio is able to predict the SSA and, to some extent, AAE even though information on particle size distribution, lensing, brown carbon and fuel types are not present, a rather surprising, but useful result."

Referee Comment: 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.

Author Response: We agree with the referee but the previous studies lack MCE and EC/OC data which prevents us from adding these points to existing plots.

Referee Comment: 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.

Author Response: We believe it is important to stress the differences in utilizing different parameterizations and believe it is important enough to leave the table intact. We have left the other wavelengths to the SI.

Referee Comment: P9, L23 – Extra word?

Author Response: We have removed the extra word.

Referee Comment: 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?

Author Response: The statement will be changed. We will now say, "Because climate models need to mix different emission types, track SSA with extensive aging, and track particle losses, we anticipate that climate models will need parameterizations that include particle-size and refractive index and will not directly implement the parameterizations presented here. However, these parameterizations provide a critical tool to assess if a model implementation, based on assumptions about refractive index and coating thicknesses (Saleh et al., 2015), generates reasonable SSA estimates."

Referee Comment: 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.

Author Response: We change the sentence as "But the predicted values for "dark" plumes are consistently larger than measured values by about 35% on average."

Referee Comment: P10, L26 – Any suggestions as to why the AAEs determined for the same burns are so different?

Author Response: We suggest two possible reasons 1) We used different instrument to measure absorption coefficients which would potentially introduced different measurement uncertainties and 2) We estimated AAE from three wavelengths by liner fitting of log(absorption) vs log(wavelengths) while Liu et al calculated between 405/781. These two different method also introduced some variabilities in calculated AAE.

Referee Comment: P10, L27-28 – There should be a reference to a source with these data. Also, seems to be a missing word in this sentence.

Author Response: We change the sentence as "Other peats (North Carolina, Canadian) produced aerosol with similar optical properties to Indonesian peat (values can be found in Table have less impact on the global radiative budget".

Referee Comment: P11, L9 – Would be good to mention the physical significance of this intercept.

Author Response: We agree with the referee and added following text.
Added Text: "which signifies that in absence of EC, SSA due to OC is close to 1 for 532 and 660 nm while it is approximately 0.91 at 405 nm due to effect of brown carbon absorption at 405 nm."

Referee Comment: Table 5 –should be no more than 2 significant figures in % difference column

Author Response: We have modified the % difference column to 2 significant figure.

References:

Abel, S. J., Haywood, J. M., Highwood, E. J., Li, J., and Buseck, P. R.: Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa, Geophys. Res. Lett., 30(15), 1783, doi:10.1029/2003GL017342, 2003.

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D. and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, doi:10.5194/acp-11-4039-2011, 2011.

Cheng, Y., He, K. B., Duan, F. K., Zheng, M., Ma, Y. L., and Tan, J. H.: Positive sampling artifact of carbonaceous aerosols and its influence on the thermal-optical split of OC/EC, Atmos. Chem. Phys., 9, 7243-7256, doi:10.5194/acp-9-7243-2009, 2009.

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.: Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning, J. Geophys. Res. Atmos., 118, 11327–11338, doi:10.1002/jgrd.50828, 2013.

Vakkari, V., Kerminen, V.-M., Beukes, J. P., Titta, P., Zyl, P. G. van, Josipovic, M., Wnter, A. D., Jaars, K., Worsnop, D. R., Kulmala, M. and Laakso, L.: Rapid change in biomass burning aerosols by atmospheric oxidation, Geophys. Res. Lett., 2644–2651, doi:10.1002/2014GL059396, 2014.

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.: Emissions from biomass burning in the Yucatan, Atmos. Chem. Phys., 9, 5785–5812, doi:10.5194/acp-9-5785-2009, 2009.

We would like to thank the reviewer for their valuable suggestions and time. Our responses are given below.

Anonymous Referee #4 Received and published: 19 April 2016

Referee Comment: Pg 3 Ln 25-26. The authors explain about observation of fire transitions from flaming to smoldering. It would be good to add information or clarify how was this done. Also how fractions of flaming to smoldering (Pg 5 Ln 28-30) were maintained during different experiments.

Author Response: We have not attempted to control transitions from flaming to smoldering. Once a fire was ignited, it was left to burn. Fire integrated absorption and extinction were utilized to avoid over-emphasizing properties of the smoldering or flaming portion of the burn.

Referee Comment: Pg 7 Ln 29. This statement about EC/OC depends more on burn conditions than fuel type can mislead. As it is shown in this study (Fig. 2), for certain fuels, such as peat, the different combustion types do not change EC/OC significantly. Therefore, fuel types do influence the EC/OC ratio.

Author Response: We have altered the sentence to read, "Furthermore, this data demonstrates that EC/OC depends significantly on burn conditions in addition to fuel type."

Referee Comment: Pg 8 Ln 24-26. The authors suggest that EC/(EC+OC) ratio is able to predict AAE. I think the r of the least square fit is not that strong for this case (r=-0.79) different from fitting for SSA. The statement may need to be revised.

Author Response: We agree with the referee that r value for AAE fit is not that strong. We have modified to the sentence to read, "In fact, the robustness of the fits suggests that the EC/(EC+OC) ratio is able to predict the SSA and, to some extent, AAE even though information on particle size distribution, lensing, brown carbon, and fuel types are not present, a rather surprising, but useful result."

Referee Comment: Pg 8 Ln 29-30. I think the RMSE values from fit of this study and fit by Liu et al. (Table 3) are not similar. They are about 20-30% different. The statement may need to be revised.

Author Response: The sentence has been modified to read, "For the MCE approach, the RMSE is similar whether coefficients from a least squares fit to our data are used or whether the coefficients proposed by Liu et al. (2014) are used, though the error is slightly lower when the coefficients from the fit to our data are used."

Technical comments

Referee Comment: Pg 4 Ln 6. What does it mean "a canister filled with Perma-Pure"? Perma-Pure is a manufacturer name.

Author Response: We change the sentence to read, "Dilution flow was generated from ambient air by passing it through an active-charcoal and permanganate (Purafil) scrubber to remove gas

phase absorbers (O_3 and NO_x) followed by a HEPA filter to remove particulates".

Referee Comment: Pg 6 Ln 14. What is temperature of filter storage?

Author Response: We have rewritten the sentence to include the temperature of filter storage. It now read, "Filters were stored in clean aluminum foil-lined petri dishes sealed with Teflon tape, and stored frozen (-20^0 C) before and after the analysis".

Referee Comment: Pg 7 Ln 18-19. I am confused with this statement. Liu et al. provided parameterization only for 405 and 532 nm, and on Fig. 1, there is no black fitted line for panel C (660 nm). So what does 660 nm refer to?

Author Response: We have deleted the sentence "At 532 and 660 nm, there are also notable errors at low MCE."

Referee Comment: Pg 9 Ln 22-23. This sentence is not finished?

Author Response: We have removed the extra word.

Referee Comment: Pg 10 Ln 10. Provide correlation plot and value of SSA at 660 nm and at 637 nm in SI.
Author Response: We were not able to obtain exact values of SSA at 637 even though we contacted Vakkari et al. The analysis is based on close inspection of their published figures with a program that converts figures to numerical values. We would not feel comfortable publishing exact values without the approval of Vakkari et al.

Referee Comment: Pg 10 Ln 22-25. In which table we can find the SSA and AAE for Indonesian peat and the other fuels?

Author Response: SSA and AAE values for all burns are available in SI (Table S2).

References:

Liu, S., Aiken, A. C., Arata, C., Dubey, M. K., Stockwell, C. E., Yokelson, R. J., Stone, E. a, Jayarathne, T., Robinson, A. L., Demott, P. J. and Kreidenweis, S. M.: Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, Geophys. Res. Lett., 41, 742–748, doi:10.1002/2013GL058392, 2014.

Vakkari, V., Kerminen, V.-M., Beukes, J. P., Titta, P., Zyl, P. G. van, Josipovic, M., Wnter, A. D., Jaars, K., Worsnop, D. R., Kulmala, M. and Laakso, L.: Rapid change in biomass burning aerosols by atmospheric oxidation, Geophys. Res. Lett., 2644–2651, doi:10.1002/2014GL059396, 2014.

Parameterization of Single Scattering Albedo (SSA) and Absorption Angstrom Exponent (AAE) with EC/OC for Aerosol Emissions from Biomass Burning

Rudra P. Pokhrel[1], Nick L. Wagner[2], Justin M. Langridge[3], Daniel A. Lack[4], Thilina Jayarathne[5], Elizabeth A. Stone[5], Chelsea E. Stockwell[6], Robert J. Yokelson[6] and Shane M. Murphy[1]

[1]Department of Atmospheric Science, University of Wyoming, Laramie, Wyoming, USA
[2]NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, Colorado, USA
[3]Observation Based Research, Met Office, Fitzroy Road, Exeter, EX1 3PB, UK
[4]Transport Emissions, Air Quality and Climate Consulting, Brisbane, Australia
[5]Department of Chemistry, University of Iowa, Iowa City, Iowa, USA
[6]Department of Chemistry, University of Montana, Missoula, Montana, USA

Correspondence to: S. M. Murphy (Shane.Murphy@uwyo.edu)

Abstract. Single scattering albedo (SSA) and absorption angstrom exponent (AAE) are two critical parameters in determining the impact of absorbing aerosol on the Earth's radiative balance. Aerosol emitted by biomass burning represent a significant fraction of absorbing aerosol globally, but it remains difficult to accurately predict SSA and AAE for biomass burning aerosol. Black carbon (BC), brown carbon (BrC), and non-absorbing coatings all make substantial contributions to the absorption coefficient of biomass burning aerosol. SSA and AAE cannot be directly predicted based on fuel type because they depend strongly on burn conditions. It has been suggested that SSA can be effectively parameterized via the modified combustion efficiency (MCE) of a biomass-burning event and that this would be useful because emission factors for CO and CO_2, from which MCE can be calculated, are available for a large number of fuels are available. Here we demonstrate, with data from the FLAME-4 experiment, that for a wide variety of globally relevant biomass fuels, over a range of combustion conditions, parameterizations of SSA and AAE based on the elemental carbon (EC) to organic carbon (OC) mass ratio are quantitatively superior to parameterizations based on MCE. We show that the EC/OC ratio and the ratio of EC/(EC+OC) both have significantly better correlations with SSA than MCE. Furthermore, the relationship of EC/(EC+OC) with SSA is linear. These improved parameterizations are significant because, similar to MCE, emission factors for EC (or black carbon) and OC are available for a wide range of biomass fuels. Fitting SSA with MCE yields correlation coefficients (Pearson's r) of ~0.65 at the visible wavelengths of 405, 532, and 660 nm while fitting SSA with EC/OC or EC/(EC+OC) yields a Pearson's r of 0.94-0.97 at these same wavelengths. The strong correlation coefficient at 405 nm (r = 0.97) suggests that parameterizations based on EC/OC or EC/(EC+OC) have good predictive capabilities even for fuels in which brown carbon absorption is significant. Notably, these parameterizations are effective for emissions from Indonesian peat, which have very little black carbon but significant brown carbon (SSA=0.990±0.001 at 532 and 660 nm, SSA = 0.937 ± 0.005 at 405 nm). Finally, we demonstrate

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that our parameterization based on EC/(EC+OC) accurately predicts SSA during the first few hours of plume aging with data from Yokelson et al. (2009) gathered during a biomass burning event in the Yucatan Peninsula of Mexico.

1 Introduction

Black carbon (BC) aerosol is the dominant atmospheric absorber of visible light and has a significant impact on the Earth's radiative balance (IPCC AR5, 2013). It has been suggested that BC may have the largest positive radiative forcing after carbon dioxide (Jacobson, 2001; Bond et al., 2013). On a global scale, the largest source of BC is open burning of forests and savannas (Bond et al., 2013). Open biomass burning also contributes two-thirds of the primary organic aerosol (POA) emissions globally (Bond et al., 2004; Bond et al., 2013). Although some climate model treat organic carbon (OC) as purely scattering (Myhre G et al., 2007), OC absorption can occur at shorter visible and ultraviolet wavelengths (Barnard et al, 2008; Lack et al., 2012a; Kirchstetter and Thatcher, 2012), which is commonly referred to as brown carbon (BrC). Few models account for BrC absorption or the effect of non-absorbing organic coatings increasing the absorption of BC (commonly called "lensing") in sophisticated ways (Jacobson, 2014; Lin et al., 2014). Recent measurements in the southeastern US show that biomass burning is the dominant source of brown carbon aerosol in this region (Washenfelder et al., 2015). Brown carbon can be a significant contributor to the overall aerosol absorption in biomass burning aerosol (Liu et al., 2014; McMeeking et al., 2014; Lack et al., 2012a). It has recently been suggested that the globally averaged top of atmosphere direct radiative forcing due to carbonaceous aerosols from biomass burning changes from negative to positive values when the effects of BrC are included (Feng et al., 2013).

Including absorption from clear-coating enhancements and brown carbon in biomass burning aerosol into global models necessitates finding a simple and accurate parameterization of the optical properties for biomass burning aerosol. Direct microphysical modelling of aerosol absorption is not feasible because emissions, morphology, mixing state, and aging are not well understood and are not easily accessible from measurements. Even if such understanding and measurements were available, a microphysical model would be too computationally expensive. Single scattering albedo (SSA) and absorption angstrom exponent (AAE) are commonly used parameters that contain the necessary information on aerosol absorption and scattering to calculate radiative effects. SSA and AAE are also critical for satellite retrievals (Ramanathan et al., 2001; McComseky, 2008) and uncertainty in SSA is one of the largest sources of uncertainty in estimating the aerosol direct and semi-direct effects (Jiang and Feingold, 2006; McComseky, 2008). SSA and AAE cannot be directly parameterized based on fuel type because they depend strongly on burn conditions and because there are no large datasets available that relate SSA or AAE to fuel type. It has been suggested that SSA and AAE can be parameterized based on the modified combustion efficiency (MCE), the ratio of CO_2 enhancement to the sum of CO and CO_2 enhancement (see Section 2.4), of a burn. Liu et al. (2014) parameterized SSA with MCE and accounted for much of the variation without including vegetation type into the parameterization, but the parameterization has limited predictive capability at higher MCE's (>0.92) where SSA changes rapidly (McMeeking et al., 2014). MCE has limited ability to predict aerosol BC/OA (Grieshop et al., 2009; Christian et al.,

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2003) on which the absorptivity of organic aerosol has a strong dependence (Saleh et al., 2014). Recent studies show that utilizing the BC/OA mass ratio may be a better way to understand aerosol optical properties than via MCE (Lu et al., 2015; Saleh et al., 2014; McMeeking et al., 2014).

Here we present data collected during the Fourth Fire Laboratory at Missoula Experiment (FLAME-4). A wide range of biomass fuels that represent significant sources of global biomass burning emissions were burned individually, and the resulting smoke was sampled with a range of in-situ instrumentation. SSA and AAE are parameterized with both the elemental to organic carbon mass ratio (EC/OC) and MCE showing that EC/OC is quantitatively superior. EC/OC is utilized in this study as opposed to BC/OA because of the experimental techniques employed (a Sunset Laboratories Instrument) though it is expected to yield similar results to BC/OA when parameterizations are applied to biomass burning emissions (Salako et al, 2012). Fuels studied include Indonesian peat, African grass, crop residue, US brushwood and coniferous trees. Tropical peatlands are one of the largest reservoirs of terrestrial organic carbon (Page et al., 2002). Previous FLAME studies were mainly constrained to the fuels prevalent in the United States (McMeeking et al., 2014; Lewis et al., 2008). Our study includes optical measurements at more wavelengths and covering a wider range of fuels than previous studies (McMeeking et al., 2014). Results are compared and contrasted with those of Liu et al. (2014), who also collected data during FLAME-4 with an independent instrument suite, to demonstrate improvements in SSA and AAE parameterization with EC/OC versus MCE. We also show that predicted SSA based on the EC/(EC+OC) parameterization is similar to measured SSA during the first few hours of aging from the Yucatan peninsula in Mexico (Yokelson et al., 2009).

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2 Materials and Methods

2.1 Fourth Fire Lab at Missoula Experiment (FLAME-4)

Measurements were made during the FLAME-4 lab experiment, a multi-investigator experiment that took place from October 15 – November 16, 2012 at the Fire Sciences Laboratory in Missoula, MT. The combustion room at the Fire Sciences Laboratory measures 12.5 m x 12.5 m x 22 m high and has a 1.6 meter-diameter, 17-meter-tall exhaust stack with a 3.6 meter inverted funnel opening 2 meters above the fuel bed. The room was continuously pressurized with outside air that was conditioned for temperature and humidity. Details of the experimental setup and a summary of fuels burned during FLAME-4 can be found in Stockwell et al. (2014). This paper summarizes results from 12 unique fuels and 41 individual burns. The list of fuels that were analysed and the geographic location where they were sampled from can be found in supplementary Table S1. Out of 41 individual burns, 20 burns were measured by pulling aerosol directly from the top of the exhaust stack (referred to as “stack” burns henceforth). During these stack burns, rapid variation in the aerosol properties are observed as the fire transitions from flaming to smoldering. The remaining 21 burns were measured from an inlet placed in the middle of the combustion room (these are referred to as "room" burns henceforth). Sampling for the room burns began after the smoke was allowed to thoroughly mix in the combustion room (typically 15-20 minutes) and continued for several hours.

The suite of optical instruments used in this work was located in a room directly adjacent to the main combustion room. During

both stack and room burns the smoke was transferred to the optical suite of instruments at 10 liters per minute via a ½" OD copper tube approximately 30 feet long. The transit time through the copper tube was roughly 5 seconds (and varied slightly depending on the amount of dilution air added for a particular burn). Smoke from the combustion room or stack intended for analysis by the optical suite of the PAS and CRDS was diluted to achieve extinctions of approximately 500 Mm^{-1} or less to prevent signal saturation in the CRDS. Dilution flow was generated from ambient air by passing it through an active-charcoal and permanganate (Purafil) scrubber to remove gas phase absorbers (O_3 and NO_x) followed by a HEPA filter to remove particulates. Dilution air was introduced to the sample flow ~1 foot from the common inlet. All results presented in this paper explore intensive properties and thus are not sensitive to dilution unless significant evaporation of semi-volatiles occurred, which has been shown to be possible (May et al., 2013). All emissions, weather additionally diluted for optical measurements or not, experienced significant dilution before sampling. A wide range of dilutions are included in the dataset because widely different masses of fuel were burned during each individual burn (see SI Table 2 for details) and some burns were diluted into the combustion room while others were diluted into the much more compact combustion stack. Despite this wide range of dilution conditions, the parameterization of optical properties with EC/(EC+OC) ratio appears robust. It is important to note that this paper is not an attempt to say what the exact EC/OC ratio will be for a given fuel, as this may depend on dilution, but that if the EC/OC ratio is known at a given dilution then the optical properties can be predicted via the parameterizations presented. Accordingly, the authors urge some caution in utilizing EC/OC emission factors from emissions that are not adequately diluted to predict regional optical properties (Akagi et al., 2011).

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2.2 Instrumentation

During the FLAME-4 experiment, absorption coefficients were measured by a 5-channel photo-acoustic absorption spectrometer (PAS) (Lack et al., 2012b) and extinction coefficients were measured by an 8-channel cavity ringdown spectrometer (CRDS) (Langridge et al., 2011). Absorption coefficients of dry aerosol (RH < 15%) were measured at 405, 532, and 660 nm and absorption of denuded aerosol were measured at 405 and 660 nm. The CRDS measured extinction coefficients of both dry and denuded aerosols at the same wavelengths as the PAS. In this paper, the denuded measurements are not used. The magnitude of absorption by the PAS was determined by sending ozone through both the CRDS and PAS and calibrating the PAS signal using the measured extinction from the CRDS, with Raleigh scattering subtracted (Lack et al., 2012b). The CRDS directly measures extinction without the need to calibrate (Langridge et al., 2011). Both the PAS and CRDS had a common inlet that conditioned the air through a number of steps. First, the aerosol was passed through a cyclone impactor that removed particles with aerodynamic diameters larger than 2.5 microns. Next, the air was dried by two 100-tube Nafion driers (Perma-Pure, Toms River, NJ) in parallel, which reduced the relative humidity in the sample cell to less than 15%. Following the Nafion driers, an activated carbon monolith (MAST Carbon, Basingstoke, UK) was used to scrub NO_x and O_3 from the sample air while transmitting the particles. The removal of NO_x was continuously tracked by a CRDS gas-phase channel at 405 nm. A filter was periodically inserted into the sample stream to remove particles and confirm baseline stability.

Uncertainty in the PAS absorption measurements is the sum of two terms, one accounting for calibration accuracy and the other for instrumental drift. The accuracy of the absolute calibration of the PAS is 5%, as detailed by Lack et al., (2006). The PAS was calibrated at the beginning and end of each measurement day during FLAME-4 and some instrument drift was observed between calibrations. The maximum change in slope between the daily calibrations was 5%. Addition of these two 5% errors in quadrature yields the overall uncertainty for the PAS measurements (7%) for this study. For the CRDS,

Langridge et al. (2011) demonstrated that the accuracy of measurements depends on relative humidity of the aerosol. For dry aerosol, as measured in this work, errors were shown to be < 2 % and that is the error utilized in this paper.

2.3 Calculation of SSA and AAE

Single scattering albedo is defined as:

$$SSA = \frac{b_{scat}}{b_{scat}+b_{abs}} = \frac{b_{scat}}{b_{ext}} = 1 - \frac{b_{abs}}{b_{ext}} \quad (1)$$

where b_{scat} is the scattering coefficient and b_{abs} is the absorption coefficient. The sum of the scattering and absorption coefficients is known as the extinction coefficient (b_{ext}). SSA in this study was calculated by inserting measurements of absorption by the PAS and extinction by the CRDS into the farthest right version of Equation 1. Errors for SSA in this study were calculated by propagating the uncertainties described above for the extinction and absorption measurements and adding the standard deviation of the actual fire measurements in quadrature. The result of this error propagation is that, based on instrumental error alone, the errors in SSA are much larger for low SSA measurements than higher SSA measurements. The instrumental error generates an error in SSA of 0.004 at an SSA of 0.95 while the error at an SSA of 0.5 is an order of magnitude higher at 0.036. Absorption angstrom exponent is defined as:

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$$b_{abs} = a\,\lambda^{-AAE} \quad (2)$$

where b_{abs} is the absorption coefficient and the constant, a, is independent of wavelength. AAE is determined in this study from the slope of a least squares fit to the logarithm of absorption coefficients versus the logarithm of wavelengths. Data from three wavelengths (405, 532, and 660 nm) are used to determine AAE. Errors for AAE were calculated as one standard deviation of the slope of the least squares fit.

During the room burns, SSA and AAE were found to be nearly constant after emissions fully mixed in the dark combustion room and the results reported in this paper are the average of 1 Hertz measurements from an approximately 1-hour period after the smoke was fully mixed. The situation was different for stack burns, where rapid fluctuations in both SSA and AAE were observed as the amount of flaming and smoldering combustion within the fire varied. In order to obtain representative measurements for the stack burns, fire-integrated SSA and AAE were calculated. To generate fire-integrated SSA, the background corrected extinction and absorption coefficients were summed for the duration of the burn and then the SSA was calculated with these integrated parameters. This procedure gives more weight to periods of the burn that produced the most extinction or absorption and should generate numbers similar to what is observed during the room burns for burns with similar fractions of flaming and smoldering. Fire-integrated AAE is generated by summing the background corrected absorption coefficients measured during a stack burn then determining the AAE as described for the room burns.

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2.4 Modified Combustion Efficiency (MCE)

The modified combustion efficiency is defined as

$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \quad (3)$$

where ΔCO and ΔCO_2 are the background-subtracted CO and CO_2 mixing ratio (Ward and Radke, 1993; Yokelson et al., 1997). Background mixing ratios were measured before the ignition of each burn. The CO and CO_2 mixing ratio were measured by an open path Fourier transform infrared spectrometer (Stockwell et al., 2014). The MCE reported in this study is the fire-integrated value.

2.5 Determination of the Elemental Carbon to Organic Carbon Ratio (EC/OC)

Fine particulate matter ($PM_{2.5}$) was selected by a cyclone operating at a flow rate of 42 liters per minute and was collected on to 37 mm quartz fiber filters (QFF) (PALL, Port Washington, NY) at ambient temperature. Field blanks were collected at a rate of one in seven samples. Prior to use, QFF were pre-cleaned by baking at 550℃ for 18 hours. Filters were stored in cleaned aluminum foil-lined petri dishes sealed with Teflon tape, and stored frozen (-20^0 C) before and after analysis. OC and EC were measured by thermal optical analysis (Sunset Laboratories, Forest Grove, OR, USA) following the IMPROVE-A protocol where the EC/OC split was determined by thermal optical transmittance. The effects of positive sampling artifacts due to carbonaceous gas adsorption were assessed using quartz filters behind Teflon (QBT) (Cheng et al., 2009) for 14 of the 96 fires, including grass, rice straw, ponderosa pine, black spruce and peat. For fires with QBT collected, the OC on the backup filter was subtracted directly. For fires without backup filters or those that were below the detection limit, the average OC correction for that fuel type was applied: rice straw (2.0 ± 0.4 %), ponderosa pine (1.2 %), black spruce (2.9 ± 1.6 %) and peat (3.1 ± 0.8 %). For fuels types without backup filters collected, the study average OC artifact (2.4 ± 1.2 %) was subtracted. Analytical uncertainties for OC were propagated from the standard deviation of field blanks (0.7 μg cm^{-2}) and 5% of the OC concentration. For EC, uncertainties were propagated from an estimate of the instrument precision (0.1 μg m^{-2}), 5% of EC concentration and 5% of pyrolyzed carbon (which forms from OC charring on the filter during analysis). The value of 5 % is a conservative estimate of the precision error in replicate sample analysis, which is typically 1-3 % (NIOSH., 1999). Analytical uncertainties for the EC/OC ratio were propagated from the individual EC and OC uncertainties.

3 Results and Discussion

Single scattering albedo (SSA) and absorption angstrom exponent (AAE) were measured during 41 individual burns of twelve different fuels during FLAME-4. Both quantities showed significant variability both from one fuel to another and also between different burns of the same fuel.

3.1 SSA Parameterization with MCE

Figure 1 shows SSA and AAE plotted versus MCE. A consistent trend of decreasing values of SSA and AAE with increasing MCE is observed for all fuels. This trend has been observed before and can be explained by the fact that more BC and less OC is produced during the flaming part of a burn when MCE is highest, while more OC and less BC is produced during the smoldering part of a burning when MCE is lowest (Ward et al., 1992; Christian et al., 2003; McMeeking et al., 2009, Liu et al., 2014). Accordingly, fuels that burn efficiently produce larger amounts of BC relative to OC and, because BC has significantly lower SSA than OC, the ensemble of particles from these efficient burns have lower values of SSA. Observed SSA values reached a minimum value of 0.257 ±0.059 at 660 nm. At high MCE, AAE is ~1 because BC dominates absorption. In contrast, fuels that burn with low MCE are dominated by OC emissions, which predominantly scatter light at long wavelengths resulting in SSA values nearing unity at 532 and 660 nm and larger values of AAE. The maximum AAE of 10.43 ±1.11 was observed for a peat burn. From the 9 different burns having MCE less than 0.90, the average SSA at 405 nm was 0.923 ±0.005 whereas the average SSA at 532 and 660 nm were 0.987 ±0.001 and 0.990 ±0.001 respectively. AAE values for these burns range from 3.68 ±0.47 to 10.43 ±1.11 with an average value of 6.51. These results strongly suggest that burns with low MCE have significant emissions of brown carbon, which has notable absorption at 405 nm, but is largely scattering at 532 and 660 nm.

While the general trend of decreasing SSA with increasing MCE is robust, a focus on the region with MCE greater or equal to 0.92 reveals that any parameterization based on MCE will have difficulty predicting the variability of SSA values in this region because SSA varies by up to 0.7 while MCE is nearly invariant. This difficulty has also been noted by McMeeking et al. (2014) based on data from the FLAME-3 set of experiments. Panels A and B of Fig. 1 show a least-squares fit to our FLAME-4 data along with the parameterizations proposed by Liu et al. (2014). The least-squares fits were made using the same functional form proposed by Liu et al. and allowing the coefficients to vary. Fitting coefficients and Pearson's correlation coefficients are given in Table 1. While the Liu et al. parameterizations have predictive capability, there are significant errors at high MCE suggesting that MCE may not be the optimal variable for parameterization. The fitted curves are significantly different than the data points at low MCE because the shape of the least-squares fitted curve is largely determined by the majority of points that occur at MCE > 0.90. A curve that fit the low MCE data better would have even larger errors than the curves shown at high MCE. The trends for AAE are similar to those for SSA. Panel D of Fig. 1 shows that parameterization of AAE with MCE can lead to significant errors at both low and high MCE.

Figure 2 demonstrates one reason why it is difficult to accurately predict SSA based on MCE. For similar values of MCE (at high MCE), EC/OC can vary by a factor of up to 6. This trend, which has also been noted by Christian et al. (2003) for a different suite of fuels, suggests that MCE is a poor proxy at the high MCE for the mass content of EC and OC on which absorption and scattering properties ultimately depend. Additionally, EC/OC for the same fuel was much more variable between lab burns than MCE. Eight different Sawgrass burns yielded MCE ranging in the narrow interval of 0.949 to 0.963,

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while EC/OC for the same burns varied from 0.010 to 4.358. Furthermore, this data demonstrates that EC/OC depends significantly on burn conditions in addition to fuel type.

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3.2 SSA Parameterization with EC/OC

Figure 3 shows the variation of SSA and AAE as a function of the EC/OC ratio for burns where EC/OC data from the Sunset Labs EC/OC instrument was available. At small EC/OC, aerosol composition is dominated by organic carbon resulting in SSA values approaching unity (at wavelengths > 532 nm) and large AAE values (>2). In contrast, at large EC/OC, composition is dominated by elemental carbon, which produces minimum SSA values as low as 0.257 ± 0.059 at 660 nm and AAE values as low as 0.854 ±0.21. The wavelength dependence of SSA is pronounced when aerosol composition is dominated by organic carbon. In general, when there are significant amounts of organic carbon SSA values at 532 and 660 nm are fairly close to one another while SSA values at 405 nm are significantly smaller. When the composition has equal or more EC than OC, there is less dependence of SSA on wavelength and AAE values are less than 2.

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These results are consistent with brown carbon causing absorption and lowering SSA at short wavelengths (<532 nm) (Barnard et al., 2008; Lack et al, 2012a; Kirchstetter and Thatcher, 2012) but only scattering at longer wavelengths. Figure 3 demonstrates that a power-law parameterization of SSA with EC/OC yields a function with a gentler change in slope that is less vertical at high SSA and will be shown to result in a lower error in predicted SSA than parameterizations based on MCE. The least square fits of SSA to EC/OC have significantly larger correlation coefficients (average of 0.96 as shown in Table 2) than those for SSA fits based on MCE (average of 0.64 as shown in Table 1). The least squares fit of AAE to EC/OC gives a slightly larger r value (r = 0.80) than the fit of AAE to MCE (r = 0.74) but the improvement is less than for the SSA fits. The equations and Pearson's correlation coefficients for each fitting line are given in Table 2. All fits are constrained with an upper limit for SSA of unity. Figure 4 demonstrates that when SSA and AAE are plotted vs. the EC/(EC+OC) ratio, the resulting fits are linear with similar correlation coefficients to the fits based on EC/OC. The y-intercepts of the fits vs. EC/(EC+OC), with SSA less than or near 1 at all wavelengths, are more physically realistic than the fits vs. EC/OC which significantly overshoot an SSA of 1 at low EC/OC ratios at 660 and 532 nm. Figure S1 shows that regression lines based on a simple liner regression (SLR) model and orthogonal distance regression (ODR) model are fairly similar at wavelengths of 660 and 532 nm for SSA vs. EC/(EC+OC). The regression lines for SSA vs. EC/(EC+OC) at 405 nm show significant deviations when fitted with the SLR vs. ODR methods, especially at higher EC/(EC+OC) ratio. This difference may be due to less data points in that region. Similarly, regression lines for AAE shows larger deviation between SLR and ODR methods at lower EC/(EC+OC) values, possibly due to less data points. ODR based fits are provided for those who prefer this regression technique.

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Given the effectiveness and linearity of the fit of SSA with EC/(EC+OC), one might conclude that the only truly important parameters for determining SSA are absorption by elemental carbon and scattering by organic carbon. However, Figure 4 demonstrates that the y-intercept of the 405 nm fit is significantly less than the y-intercepts for the 532 and 660 nm fits. This suggests that the EC/(EC+OC) ratio is able to capture the effect of brown carbon, an idea that has also been suggested by Saleh et al. (2014; 2015). In fact, the robustness of the fits suggests that the EC/(EC+OC) ratio is able to predict the SSA

and, to some extent, AAE even though information on particle size distribution, lensing, brown carbon, and fuel types are not present, a rather surprising but useful result.

To compare the performance of the different regression models discussed in this study, we calculated the root mean square error (RMSE) for the EC/OC and EC/(EC+OC) based models and compared with the RMSE of the MCE based model first proposed by Liu et al. (2014). The RMSE values for the different regression models are reported in Table 3. For the MCE approach, the RMSE is similar whether coefficients from a least squares fit to our data are used or whether the coefficients proposed by Liu et al. (2014) are used, though the error is slightly lower when the coefficients from the fit to our data are used. The main result is that the RMSE of the models based on EC/OC or EC/(EC+OC) are significantly less than RMSE for the model based on MCE.

3.3 Comparison of SSA and AAE from This Study with Previous Results

Given the useful robustness of the fits of SSA with EC/OC and EC/(EC+OC), it is important to confirm that the SSA values measured during this study are representative. As mentioned earlier, the FLAME-4 study analyzed a wide range of globally relevant fuels. Some fuels were burned in both FLAME-4 and FLAME-3 at similar MCE. In these cases, the SSA values from this study are, within experimental error, similar to those derived by McMeeking et al. (2014). The range of SSA values for ponderosa pine (varies from 0.832±0.012 to 0.987±0.001) and black spruce (varies from 0.899±0.007 to 0.929±0.005) at 532 nm are similar to previously reported SSA values measured on temperate and boreal forest at wavelengths of 540 and 550 nm (ranged from 0.83±0.11 to 0.97±0.02) (Hobbs et al., 1996; Radke et al., 1998; Radke et al., 1991). AAEs reported by Lewis et al., (2008) are comparable to AAEs estimated for burns with MCE greater than 0.90 during this study. However, for burns with MCE less than 0.90, AAEs from this study are larger than many previously reported for biomass burning aerosols (Lewis et al., 2008; Liu et al., 2014), but McMeeking et al. (2014) is closer to our values. For burns with MCE less than 0.90, the AAE values from this study are comparable to previous studies that estimated the AAE of organic material, without BC, from biomass burning, as shown in Table 4. This is consistent with our study including burns (mainly peat combustion and two low efficiency ponderosa pine burns) that produced very little black carbon and behaved optically like pure organic emissions. Overall, the data collected during this study are reasonably similar to previous studies, encompass a wide range of burn conditions, and produce new insights.

3.4 Impact of Parameterization with EC/OC

We next determined how significant of an impact parameterizing SSA with EC/OC as opposed to MCE could have in climate models. Climate models typically begin with vegetation-based emission factors, so we compared the SSA predicted by the MCE and EC/OC parameterizations using the literature average emission factors of a wide variety of global fires types compiled by Akagi et al. (2011). As shown in Table 5, the estimated SSA at 532 nm varies significantly for some fuels while it is similar for others. The differences for other wavelength is similar in magnitude and is given in Table S3. The maximum difference in the predicted SSA at 532 nm between EC/OC and MCE approach is ~ 48 % for garbage burning. Because climate

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models need to mix different emission types, track SSA with extensive aging, and track particle losses, we anticipate that climate models will need parameterizations that include particle-size and refractive index and will not directly implement the parameterizations presented here. However, these parameterizations provide a critical tool to assess if a model implementation, based on assumptions about refractive index and coating thicknesses (Saleh et al., 2015), generates reasonable SSA estimates.

3.5 Robustness of the EC/OC parameterization of SSA in aging aerosol

Significant changes in the SSA of biomass burning aerosol can occur during the first few hours of aging (Abel et al., 2003; Yokelson et al., 2009; Vakkari et al., 2014). In a typical example, Yokelson et al. (2009) documented a BB plume where the SSA at 530 nm changed from ~0.75 to ~0.93 during 1.4 hours of aging. This presents a key question of whether the parameterizations of SSA based on EC and OC presented in this work can capture the observed aging effects. To probe this question, we calculated the SSA that would be predicted based on our EC/(EC+OC) regression model and compared it to the actual SSA observed by Yokelson et al. (2009) because that study provided reasonable auxiliary data. However, Yokelson et al. (2009) only measured $\Delta BC/\Delta PM_{2.5}$ instead of EC/(EC+OC) so several assumptions had to be made to implement our parameterization. One important note is that the $\Delta BC/\Delta PM_{2.5}$ reported in Yokelson et al. (2009) was derived from optical measurements. $\Delta BC/\Delta PM_{2.5}$ was converted in to EC/(EC+OC) by setting the OC mass fraction to 39 ±9 % of the $PM_{2.5}$ as stated by Yokelson et al. (2009). This calculation assumes BC and EC mass are identical. Figure 5 shows the measured and predicted SSA during the first 1.4 hrs of aging, which was the extent of aging observed in this dataset. The predicted SSA based on the EC/(EC+OC) parameterization closely tracks the measured SSA. The shaded region in Fig. 5 shows the 95% confidence interval of the predicted SSA values, and this interval encompasses most of the measured SSA values. This result suggests that the parameterization remains valid during initial aging of biomass plumes. Similarly, SSA values are predicted based on an ODR model and compared with the SSA predicted by an SLR model. Figure S2 shows the comparison of SSA predicted based on SLR vs. ODR models. The general trend shows that the predicted SSA based on SLR is higher than that based on ODR, but that difference is not statistically significant. We performed a two-tailed t test with the null hypothesis that predicted values are the same from both regression models and found a two-tailed p value of 0.748 at 532 nm. A similar approach was used to check the performance of our SSA parameterization with observations from Vakkari et al. (2014) of biomass burning plumes with variable ages in southern Africa. The predicted SSA at 660 nm shows reasonably good agreement with the measured SSA at 637 nm. The predictions are within roughly ± 5% for biomass burning plumes with SSA > 0.75, which encompasses most field observations of BB plumes. But the predicted values for "dark" plumes are consistently larger than measured values by about 35% on average. However, Vakkari et al. (2014) note that the "dark" plumes they observed are rarely observed in the atmosphere or laboratory studies.

3.6 Indonesian Peat

One fuel that deserves special mention is Indonesian Peat because of the large emissions from this source and the minimal available data for it. During the 1997 El Niño event, 0.19-0.23 Gt of carbon was estimated to be emitted to the

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atmosphere through peat burning, which was equivalent to ~40% of the mean annual global carbon emissions from fossil fuel (Page et al., 2002). Chakrabarty et al. (2015) recently presented results for emissions from Boreal peat, but not for Indonesian peat, which is thought to currently be a more significant global source of emissions. Chand et al. (2005) reported an SSA of 0.99 for Indonesian peat burned in the laboratory at a wavelength of 540 nm. This SSA is similar to the SSA at a wavelength of 532 nm for Indonesian peat burned during this study (0.990 ± 0.001) and by Liu et al. (2014). However, this study and Liu et al. (2014) present a significant new FLAME-4 finding that the SSA of the aerosol from burning Indonesian peat at 405 nm is 0.937 ± 0.005, demonstrating that this fuel emits significant amounts of brown carbon and is not only scattering. Our derived average AAE of 7.7 ± 0.42 further demonstrates the importance of brown carbon from this fuel and enables modelling of its absorption properties across the visible spectrum. Our AAE is higher than reported for the same peat fuels by Liu et al. (2014). Other peats (North Carolina, Canadian) produced aerosol with similar optical properties to Indonesian peat (values can be found in Table), have less impact on the global radiative budget.

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4.0 Conclusions

We examined the SSA and AAE of aerosol emissions from the combustion of a variety of globally significant biomass fuels under controlled laboratory conditions. It was found that SSA and AAE varied significantly, but that the variation could be explained by dependence on burn conditions without a need to explicitly account for fuel type. Measured SSA ranged from 0.257 ± 0.059 to 0.997 ± 0.001 at 660 nm, 0.361 ± 0.046 to 0.968 ± 0.002 at 405 nm, and AAE ranged from 0.85 ±0.21 to 10.43 ±1.11. We demonstrate that SSA parameterization with EC/OC and EC/(EC+OC) is quantitatively superior to parameterization with MCE and that the best fit of SSA with EC/(EC+OC) is linear. By applying various parameterizations of SSA to global emission factors measured by Akagi et al. (2011), we demonstrate that parameterizations based on EC/OC and EC/(EC+OC) yield similar results to one another, but yield significantly different results than parameterizations based on MCE. Biomass burning aerosol emissions with compositions dominated by OC give higher AAE than those with more BC content, suggesting that the organic fraction of these emissions contains significant brown carbon. The effect of brown carbon on SSA parameterization with EC/(EC+OC) is to cause the y-intercept to be near unity (0.98-0.99) at 532 and 660 nm and less than unity (0.91) at 405 nm. Which signifies that in absence of EC, SSA due to OC is close to 1 for 532 and 660 nm while it is approximately 0.91 at 405 nm due to effect of brown carbon absorption at 405 nm. Emissions from the burning of peat, one of the largest source of terrestrial organic carbon (Page et al., 2002), yielded an SSA at 532 and 660 nm close to unity (0.990 ± 0.001 on average), but an average SSA at 405 nm of 0.937 ± 0.005. This wavelength dependent SSA, and an average AAE of 7.7 ± 0.42, show that brown carbon absorption from peat combustion emissions are significant. Finally, we demonstrate that our parameterization of SSA based on EC/(EC+OC) accurately predicts SSA during the first few hours of plume aging with ambient data from Yokelson et al. (2009) gathered during a biomass burning event in the Yucatan Peninsula of Mexico.

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References

Abel, S. J., Haywood, J. M., Highwood, E. J., Li, J., and Buseck, P. R.: Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa, Geophys. Res. Lett., 30(15), 1783, doi:10.1029/2003GL017342, 2003.

Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D. and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys., 11, 4039–4072, doi:10.5194/acp-11-4039-2011, 2011.

Barnard, J. C., Volkamer, R. and Kassianov, E. I.: Estimation of the mass absorption cross section of the organic carbon component of aerosols in the Mexico City Metropolitan Area (MCMA), Atmos. Chem. Phys., 8, 6665–6679, doi:10.5194/acp-8-6665-2008, 2008.

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J. H. and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi:10.1029/2003JD003697, 2004.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., Deangelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380–5552, doi:10.1002/jgrd.50171, 2013.

Chakrabarty, R. K., Gyawali, M., Yatavelli, R. L. N., Pandey, A., Watts, A. C., Knue, J., Chen, L.-W. A., Pattison, R. R., Tsibart, A., Samburova, V. and Moosmuller, H.: Dominance of brown carbon in aerosol emissions from burning of boreal peatlands, Atmos. Chem. Phys. Discuss., 15, 28793–28813, doi:10.5194/acpd-15-28793-2015, 2015.

Chand, D., Schmid, O., Gwaze, P., Parmar, R. S., Helas, G., Zeromskiene, K., Wiedensohler, A., Massling, A. and Andreae, M. O.: Laboratory measurements of smoke optical properties from the burning of Indonesian peat and other types of biomass, Geophys. Res. Lett., 32, L12819, doi:10.1029/2005GL022678, 2005.

Cheng, Y., He, K. B., Duan, F. K., Zheng, M., Ma, Y. L., and Tan, J. H.: Positive sampling artifact of carbonaceous aerosols and its influence on the thermal-optical split of OC/EC, Atmos. Chem. Phys., 9, 7243-7256, doi:10.5194/acp-9-7243-2009, 2009.

Christian, T. J., Kleiss, B., Yokelson, R. J., Holzinger, R., Crutzen, P. J., Hao, W. M., Saharjo, B. H. and Ward, D. E.: Comprehensive laboratory measurements of biomass-burning emissions: 1. Emissions from Indonesian, African, and other fuels, J. Geophys. Res., 108(D23), 4719, doi:10.1029/2003JD003704, 2003.

Feng, Y., Ramanathan, V. and Kotamarthi, V. R.: Brown carbon: A significant atmospheric absorber of solar radiation, Atmos. Chem. Phys., 13, 8607–8621, doi:10.5194/acp-13-8607-2013, 2013.

Grieshop, A. P., Logue, J. M., Donahue, N. M. and Robinson, A. L.: Laboratory investigation of photochemical oxidation of organic aerosol from wood fires 1: Measurement and simulation of organic aerosol evolution, Atmos. Chem. Phys., 9, 1263–1277, doi:10.5194/acp-9-1263-2009, 2009.

Hobbs, P. V., Reid, J. S., Herring, J. A., Nance, J. D., Weiss, R. E., Ross, J. L., Hegg, D. A., Ottmar, R. D., and Liousse, C.: Particle and trace-gas measurements in smoke from prescribed burns of forest products in the Pacific Northwest, in: Biomass Burning and Global Change, Vol. 1, edited by: Levine, J. S., MIT Press, New York, 697–715, 1996.

Intergovernmental Panel on Climate Change: Anthropogenic and Natural Radiative Forcing, Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, 659–740, doi:10.1017/ CBO9781107415324.018, 2013.

Jacobson, M. Z.: Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, Nature, 409, 695–697, 2001.

Jacobson, M. Z.: Effects of biomass burning on climate, accounting for heat and moisture fluxes, black and brown carbon, and cloud absorption effects, J. Geophys. Res. Atmos., 119, 8980–9002, doi:10.1002/2014JD021861, 2014.

Jiang, H. and Feingold, G.: Effect of aerosol on warm convective clouds: Aerosol-cloud surface flux feedbacks in a new coupled large eddy model, J. Geophys. Res. Atmos., 111, D01202, doi:10.1029/2005JD006138, 2006.

Kirchstetter, T. W. and Thatcher, T. L.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys., 12, 6067–6072, doi:10.5194/acp-12-6067-2012, 2012.

Lack, D. a., Lovejoy, E. R., Baynard, T., Pettersson, A. and Ravishankara, a. R.: Aerosol Absorption Measurement using Photoacoustic Spectroscopy: Sensitivity, Calibration, and Uncertainty Developments, Aerosol Sci. Technol., 40(9), 697–708, doi:10.1080/02786820600803917, 2006.

Lack, D. A., Langridge, J. M., Bahreini, R., Cappa, C. D., Middlebrook, a. M. and Schwarz, J. P.: Brown carbon and internal mixing in biomass burning particles, Proc. Natl. Acad. Sci., 109, 14802–14807, doi:10.1073/pnas.1206575109, 2012a.

Lack, D. A., Richardson, M. S., Law, D., Langridge, J. M., Cappa, C. D., McLaughlin, R. J. and Murphy, D. M.: Aircraft Instrument for Comprehensive Characterization of Aerosol Optical Properties, Part 2: Black and Brown Carbon Absorption and Absorption Enhancement Measured with Photo Acoustic Spectroscopy, Aerosol Sci. Technol., 46, 555–568, doi:10.1080/02786826.2011.645955, 2012b.

Langridge, J. M., Richardson, M. S., Lack, D., Law, D. and Murphy, D. M.: Aircraft Instrument for Comprehensive Characterization of Aerosol Optical Properties, Part I: Wavelength-Dependent Optical Extinction and Its Relative Humidity Dependence Measured Using Cavity Ringdown Spectroscopy, Aerosol Sci. Technol., 45, 1305–1318, doi:10.1080/02786826.2011.592745, 2011.

Lewis, K., Arnott, W. P., Moosmüller, H. and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, J. Geophys. Res., 113, D16203, doi:10.1029/2007JD009699, 2008.

Lin, G., Penner, J. E., Flanner, M. G., Sillman, S., Xu, L., and Zhou, C.: Radiative forcing of organic aerosol in the atmosphere and on snow: effects of SOA and brown carbon, J. Geophys. Res. Atmos., 119, 7453–7476, doi:10.1002/2013JD021186, 2014.

Liu, J., Scheuer, E., Dibb, J., Ziemba, L. D., Thornhill, K. L., Anderson, B. E., Wisthaler, A., Mikovinỳ, T., Devi, J. J., Bergin, M. and Weber, R. J.: Brown carbon in the continental troposphere, Geophys. Res. Lett., 41, 2191–2195, doi:10.1002/2013GL058976, 2014.

Liu, S., Aiken, A. C., Arata, C., Dubey, M. K., Stockwell, C. E., Yokelson, R. J., Stone, E. a, Jayarathne, T., Robinson, A. L., Demott, P. J. and Kreidenweis, S. M.: Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, Geophys. Res. Lett., 41, 742–748, doi:10.1002/2013GL058392, 2014.

Lu, Z., Streets, D. G., Winijkul, E., Yan, F., Chen, Y., Bond, T. C., Feng, Y., Dubey, M. K., Liu, S., Pinto, J. P. and Carmichael, G. R.: Light Absorption Properties and Radiative Effects of Primary Organic Aerosol Emissions, Environ. Sci. Technol., 49, 4868–4877, doi:10.1021/acs.est.5b00211, 2015.

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.: Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning, J. Geophys. Res. Atmos., 118, 11327–11338, doi:10.1002/jgrd.50828, 2013.

McComiskey, A., Schwartz, S. E., Schmid, B., Guan, H., Lewis, E. R., Ricchiazzi, P. and Ogren, J. a.: Direct aerosol forcing: Calculation from observables and sensitivities to inputs, J. Geophys. Res. Atmos., 113, D09202, doi:10.1029/2007JD009170, 2008.

McMeeking, G. R., Kreidenweis, S. M., Baker, S., Carrico, C. M., Chow, J. C., Collett, J. L., Hao, W. M., Holden, A. S., Kirchstetter, T. W., Malm, W. C., Moosmüller, H., Sullivan, A. P. and Cyle E., W.: Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory, J. Geophys. Res., 114, D19210, doi:10.1029/2009JD011836, 2009.

McMeeking, G. R., Fortner, E., Onasch, T. B., Taylor, J. W., Flynn, M., Coe, H. and Kreidenweis, S. M.: Impacts of nonrefractory material on light absorption by aerosols emitted from biomass burning, J. Geophys. Res. Atmos., 119, 12272–12286, doi:10.1002/2014JD021750, 2014.

Myhre, G., Bellouin, N., Berglen, T. F., Berntsen, T. K., Boucher, O., Grini, A., Isaksen, I. S. a., Johnsrud, M., Mishchenko, M. I., Stordal, F. and Tanré, D.: Comparison of the radiative properties and direct radiative effect of aerosols from a global aerosol model and remote sensing data over ocean, Tellus B, 59, 115–129, doi:10.1111/j.1600-0889.2006.00226.x, 2007.

NIOSH: Manual of Analytical Methods. Elemental carbon (diesel particulate): Method 5040, Edited by: Eller, P. M.; Cassinelli, M. E. Publisher: National Institute for Occupational Safety and Health, Cincinnati, OH, 1999.

Radke, L. F., Hegg, D. A., Hobbs, P. V., Nance, J. D., Lyons, J. H., Larsen, K. K., Weiss, R. E., Regan, P. J., and Ward, D. E.: Particulate and trace emissions from large biomass fires in North America, in: Global Biomass Burning: Atmospheric, Climatic, and Biospheric Implications, edited by: Levine, J. S., MIT Press, Cambridge, MA, 209–224, 1991.

Radke, L. F, Hegg, D. A., Lyons, J. H., Brock, C. A., Hobbs, P. V., Weiss, R., and Rassmussen, R.: Airborne measurements on smokes from biomass burning, in: Aerosols and Climate, edited by: Hobbs, P. V. and McCormick, M. P., Deepak Publishing, Hampton VA, 411–422, 1988.

Page, S. E., Siegert, F., Rieley, J. O., Boehm, H.-D. V., Jaya, A. and Limin, S.: The amount of carbon released from peat and forest fires in Indonesia during 1997, Nature, 420, 61–65, doi:10.1038/nature01141, 2002.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T. and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle., Science, 294, 2119–2124, doi:10.1126/science.1064034, 2001.

Salako, G. O., Hopke, P. K., Cohen, D. D., Begum, B. A., Biswas, S. K., Pandit, G. G., Chung, Y. S., Rahman, S. A., Hamzah, M. S., Davy, P., Markwitz, A., Shagjjamba, D., Lodoysamba, S., Wimolwattanapun, W. and Bunprapob, S.: Exploring the variation between EC and BC in a variety of locations, Aerosol Air Qual. Res., 12, 1–7, doi:10.4209/aaqr.2011.09.0150, 2012.

Saleh, R., Robinson, E. S., Tkacik, D. S., Ahern, A. T., Liu, S., Aiken, A. C., Sullivan, R. C., Presto, A. a, Dubey, M. K., Yokelson, R. J., Donahue, N. M. and Robinson, A. L.: Brownness of organics in aerosols from biomass burning linked to their black carbon content, Nat. Geosci., 7, 647–650, doi:10.1038/ngeo2220, 2014.

Saleh, R., Marks, M., Heo, J., Adams, P. J., Donahue, N. M. and Robinson, A. L.: Contribution of brown carbon and lensing to the direct radiative effect of carbonaceous aerosols from biomass and biofuel burning emissions, J. Geophys. Res. Atmos., 120, doi:10.1002/2015JD023697-T, 2015.

Srinivas, B. and Sarin, M. M.: Light absorbing organic aerosols (brown carbon) over the tropical Indian Ocean: impact of biomass burning emissions, Environ. Res. Lett., 8, 044042, doi:10.1088/1748-9326/8/4/044042, 2013.

Srinivas, B. and Sarin, M. M.: Brown carbon in atmospheric outflow from the Indo-Gangetic Plain: Mass absorption efficiency and temporal variability, Atmos. Environ., 89, 835–843, doi: 10.1016/j.atmosenv.2014.03.030, 2014.

Stockwell, C. E., Yokelson, R. J., Kreidenweis, S. M., Robinson, A. L., DeMott, P. J., Sullivan, R. C., Reardon, J., Ryan, K. C., Griffith, D. W. T. and Stevens, L.: Trace gas emissions from combustion of peat, crop residue, biofuels, grasses, and other fuels: configuration and FTIR component of the fourth Fire Lab at Missoula Experiment (FLAME-4), Atmos. Chem. Phys., 14, 9727–9754, doi:10.5194/acp-14-9227-2014, 2014.

Sun, H., Biedermann, L. and Bond, T. C.: Color of brown carbon: A model for ultraviolet and visible light absorption by organic carbon aerosol, Geophys. Res. Lett., 34, L17813, doi:10.1029/2007GL029797, 2007.

Vakkari, V., Kerminen, V.-M., Beukes, J. P., Titta, P., Zyl, P. G. van, Josipovic, M., Wnter, A. D., Jaars, K., Worsnop, D. R., Kulmala, M. and Laakso, L.: Rapid change in biomass burning aerosols by atmospheric oxidation, Geophys. Res. Lett., 2644–2651, doi:10.1002/2014GL059396, 2014.

Washenfelder, R. A., Attwood, A. R., Brock, C. A., Guo, H., Xu, L., Weber, R. J., Ng, N. L., Allen, H. M., Ayres, B. R., Baumann, K., Cohen, R. C., Draper, D. C., Duffey, K. C., Edgerton, E., Fry, J. L., Hu, W. W., Jimenez, J. L., Palm, B. B., Romer, P., Stone, E. A., Wooldridge, P. J. and Brown, S. S.: Biomass burning dominates brown carbon absorption in the rural southeastern United States, J. Geophys. Res. Lett., 42, 653–664, doi:10.1002/2014GL062444, 2015.

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Stier, P., Seinfeld, J. H., Kinne, S. and Boucher, O.: Aerosol absorption and radiative forcing, Atmos. Chem. Phys., 7, 5237–5261, doi:10.5194/acp-7-5237-2007, 2007.

Ward, D. E. and Radke, L. F.: Emissions measurements from vegetation fires: A Comparative evaluation of methods and results, in: Fire in the Environment: The Ecological, Atmospheric and Climatic Importance of Vegetation Fires, edited by: Crutzen, P. J. and Goldammer, J. G., John Wiley, New York, 53–76, 1993.

Ward, D. E., Susott, R. a., Kauffman, J. B., Babbitt, R. E., Cummings, D. L., Dias, B., Holben, B. N., Kaufman, Y. J., Rasmussen, R. a. and Setzer, a. W.: Smoke and fire characteristics for cerrado and deforestation burns in Brazil - BASE-B experiment, J. Geophys. Res., 97, 14601–14619, doi:10.1029/92JD01218, 1992.

Yokelson, R. J., Ward, D. E., Susott, R. A., Reardon, J. and Griffith, D. W. T.: Emissions from smoldering combustion of biomass measured by open-path Fourier transform infrared spectroscopy, J. Geophys. Res., 102, 18865–18877, 1997.

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.: Emissions from biomass burning in the Yucatan, Atmos. Chem. Phys., 9, 5785–5812, doi:10.5194/acp-9-5785-2009, 2009.

Zhong, M. and Jang, M.: Dynamic light absorption of biomass-burning organic carbon photochemically aged under natural sunlight, Atmos. Chem. Phys., 14, 1517–1525, doi:10.5194/acp-14-1517-2014, 2014.

Table 1. Fitting coefficients for SSA as a function of MCE ($SSA = k_0 + k_1(MCE)^{k2}$) and AAE as a function of MCE ($AAE = a\ (MCE)^b$) for fuels analyzed in this study with Pearson's r values for each fit. Numbers in parentheses are one standard deviation of the fitting coefficients.

	Wavelengths (nm)	k_0	k_1	k_2	r
SSA	405	0.920(± 0.043)	-0.632(± 0.221)	26.877(±11.2)	0.649
	532	0.933(± 0.038)	-1.637(± 1.05)	58.492(± 23.3)	0.64
	660	0.941(± 0.041)	-1.687(± 1.05)	56.45(± 22.3)	0.644
AAE	405/532/660	a	b		
		2.454 (±0.231)	-3.292(±0.418)		0.738

Table 2. Fitting coefficients for SSA as a function of EC to OC ratio ($y = k_0 + k_1(EC/OC)^{k2}$) and AAE as a function of EC to OC ratio ($y = a\ (EC/OC)^{b}$) for fuels analyzed in this study with Pearson's r values for each fit. Numbers in parentheses are one standard deviation of the fitting coefficients.

	Wavelengths (nm)	k_0	k_1	k_2	r
SSA	405	1.000(± 0.048)	-0.497(± 0.056)	0.429(±0.079)	0.973
	532	1.000(± 0.043)	-0.469(± 0.06)	0.576(± 0.119)	0.960
	660	1.000(± 0.055)	-0.483(± 0.078)	0.602(± 0.157)	0.937
AAE	405/532/660	a	b		
		1.802 (±0.275)	-0.232(±0.041)		0.796

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Table 3. Root mean square error (RMSE) for SSA and AAE parameterization with EC/OC, EC/(EC+OC), MCE with fit coefficients from a least squares fit of this study's data, and MCE with coefficients from Liu et al., (2014).

	λ (nm)	EC/OC	EC/(EC+OC)	MCE (Liu et al., 2014)	MCE (Fit to This Study)
SSA	405	0.041	0.04	0.163	0.115
	532	0.057	0.043	0.17	0.133
	660	0.077	0.06	0.224	0.144
AAE	405/532/660	0.911	0.92	NA	1.324

Table 4. AAE calculated for burns with MCE less than 0.90 compared with previous studies that derived the AAE of organic mass (without BC) for biomass burning emissions.

Reference	Wavelength (nm)	Aerosol Component Analyzed	AAE
This Study	405/532/660	Entire Aerosol	3.7 to 10.4 Average = 6.5
Srinivas, B. and Sarin, M. M. (2013)	365-700	Water Soluble Organics	3 to 19 Average = 9
Srinivas, B. and Sarin, M. M. (2014)	300-700	Water Soluble Organics	8.3 ± 2.6
Feng et al., (2013)	400-700	Organic	6.6
Sun et al., 2007	NA	Water Soluble Organics	6
Zhong, M. and Jang, M (2014)	400-700	Organics	4.74
Kirchstetter and Thatcher (2012)	400-700	Organics	3.0 to 7.4 Average = 5

Table 5. Comparison of SSA at 532 nm predicted based on MCE (with Liu et al. (2014) coefficients), EC/OC, and EC/(EC+OC) parameterizations for emission factors of various biomass fuels from Akagi et al. (2011). The values in % difference in predicted SSA column are percentage difference for EC/OC / EC/(EC+OC) approaches respectively. BC/OC is assumed to be equivalent to EC/OC.

Biomass Types	MCE[1]	BC/OC[1]	SSA_532 MCE Approach	SSA_532 EC/OC Approach	SSA_532 EC/(EC+OC) Approach	% Difference in Predicted SSA
Tropical Forest	0.95	0.11	0.79	0.87	0.88	-10/-11
Savanna	0.96	0.14	0.67	0.85	0.86	-27/-28
Crop Residue	0.94	0.33	0.82	0.75	0.73	9/11
Pasture Maintenance	0.92	0.09	0.89	0.88	0.89	1/0
Boreal Forest	0.92	-	0.88	-	-	-
Temperate Forest	0.95	-	0.77	-	-	-
Extra tropical Forest	0.93	0.07	0.87	0.9	0.92	-3/ -6
Peat land	0.9	0.03	0.94	0.93	0.95	1/1
Chaparral	0.96	0.35	0.69	0.74	0.72	-7/-4
Open Cooking	0.95	0.29	0.75	0.77	0.76	-3/-1
Patsari Stoves	0.97	0.39	0.58	0.73	0.7	-26/-21
Charcoal Making	0.86	0.03	0.97	0.94	0.96	3/1
Charcoal Burning	0.93	0.77	0.87	0.59	0.54	32/38
Dung Burning	0.89	0.29	0.95	0.77	0.75	19/21
Garbage Burning	0.97	0.12	0.58	0.86	0.87	-48/50

[1]Data from Akagi et al., 2011

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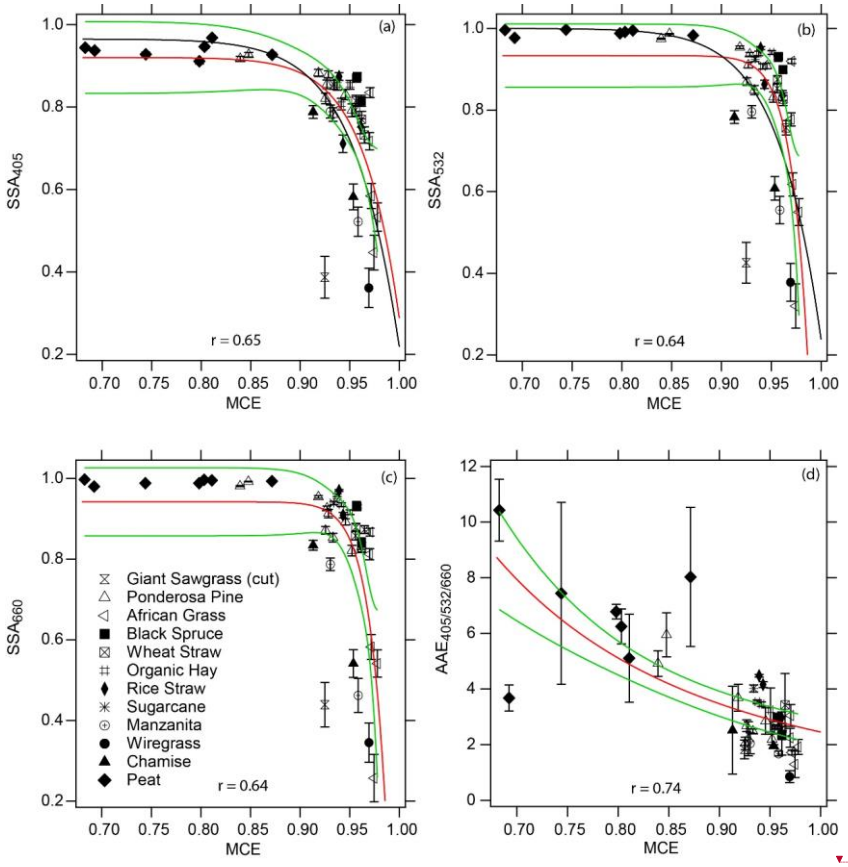

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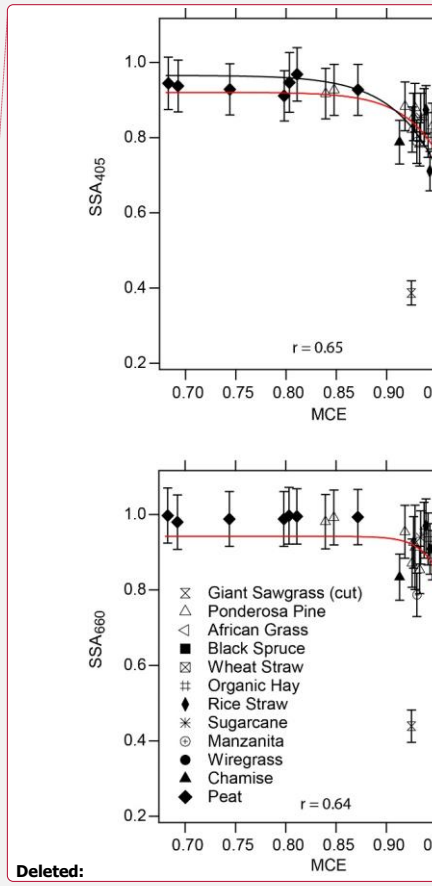


Figure 1. Single scattering albedo at (a) 405 nm, (b) 532 nm, and (c) 660 nm plotted as a function of MCE measured during the FLAME-4 experiment. The solid red lines are least-squares fits of the data with details given in Table 1 and the black lines are parameterizations proposed by Liu et al. (2014) which have the same functional form as the red-line fits. Error bars for SSA are calculated by taking 7% uncertainty in absorption measurement, 2% uncertainty in extinction measurement and one standard deviation of the average value for room burns and adding these uncertainties in quadrature. For stack burns, the error is the quadrature sum of the uncertainty in the PAS and CRDS measurements. (d) Absorption angstrom exponent as a function of MCE. The error bars for AAE are one standard deviation of the least squares fit to the averaged data for a given fuel. Green lines are the 95% confidence interval of the fits.

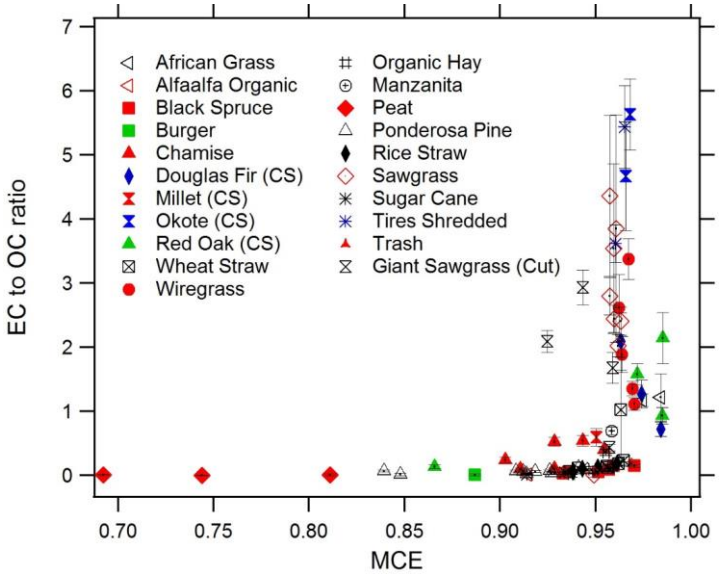
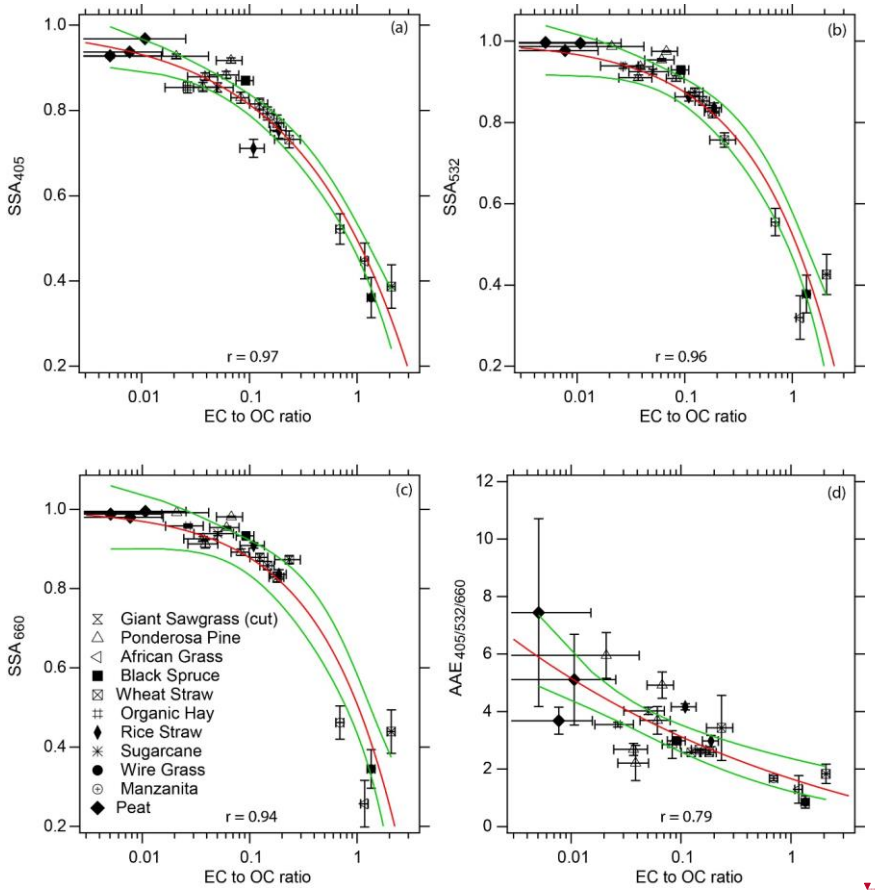


Figure 2. EC/OC ratio plotted as a function of MCE. Different symbols and colours represent different fuels as listed in the legend. The error bars are the propagation error calculated from the uncertainty associated with the EC and OC measurement. This plot contains data from additional fuels from FLAME-4 for which SSA and AAE were not calculated.



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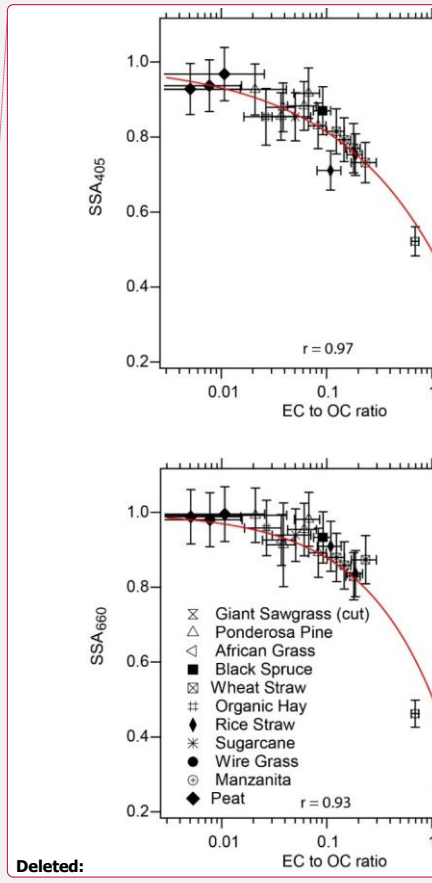
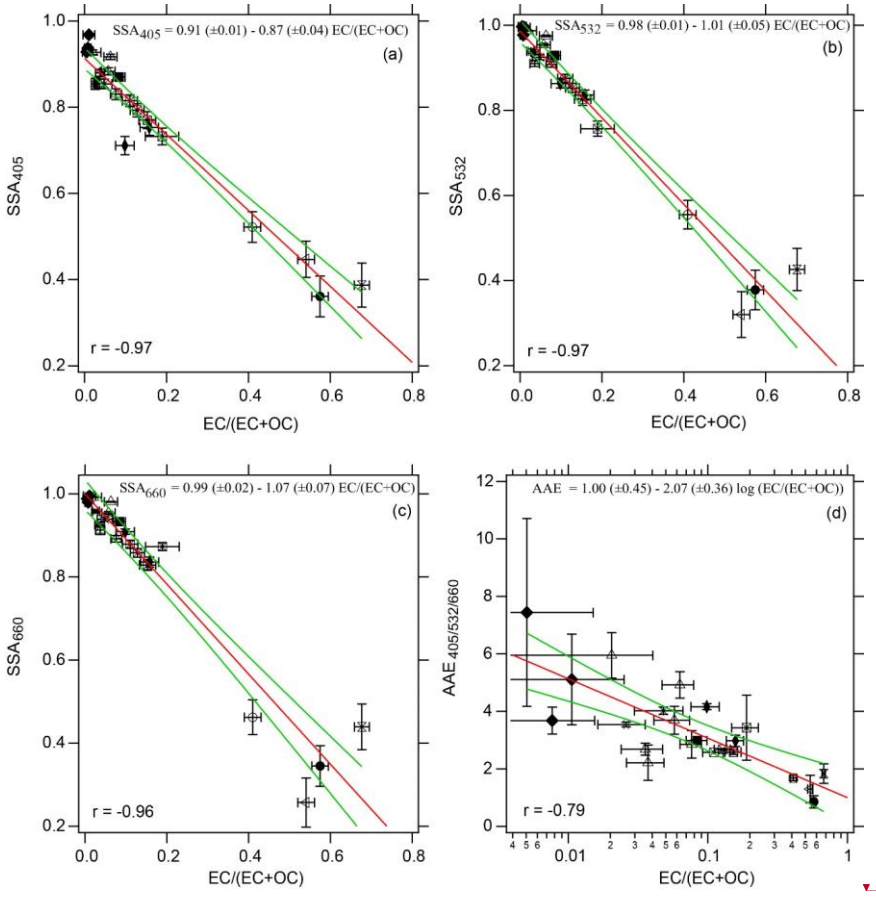


Figure 3. SSA at (a) 405 nm, (b) 532 nm, and (c) 660 nm plotted as a function of EC/OC. Solid red lines are least-squares best fits of the data that are constrained to force SSA to be less than one. The error bars on SSA are the propagation error calculated by taking 7 % uncertainty in absorption measurement, 2% uncertainty in extinction measurement and one standard deviation of the average value for room burns. Whereas for stack burn it is the propagation error calculated form uncertainty in PAS and CRDS measurements. Error bars on the EC to OC ratio are the propagation error calculated from the uncertainty associated with the EC and OC measurement (d) AAE as a function of EC/OC with error bars being one standard deviation of the measurement. Green lines are the 95% confidence interval of the fits.



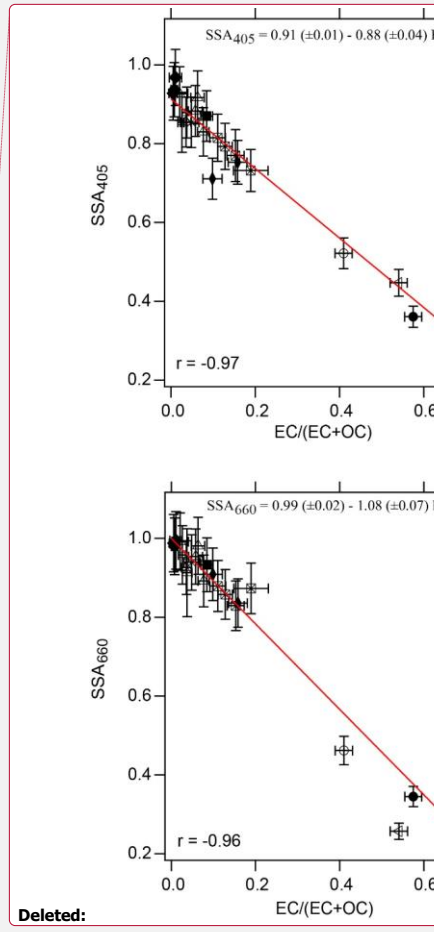


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Figure 4. Same as Fig. 3 but for SSA plotted vs. $\frac{EC}{(EC+OC)}$. Equations for the fits are given in each panel. Green lines are the 95% confidence interval of the fits.

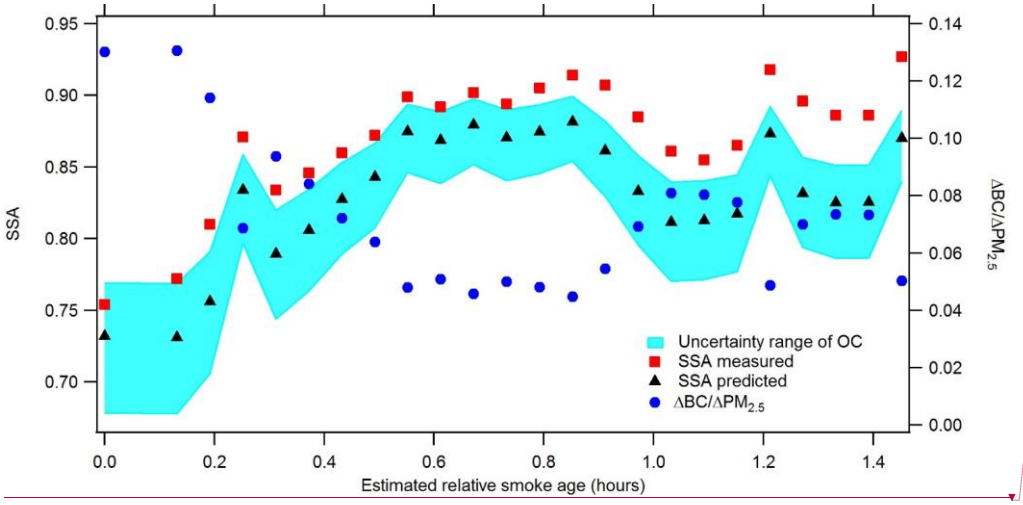

Figure 5. Measured (Yokelson et al., 2009) and predicted SSA at 532 nm for biomass burning emissions during the initial 1.4 hours of aging. Shaded region indicate the predicted SSA values on accounting uncertainty ranges of OC mass.

Deleted: 95 % confidence interval of the predicted SSA.

Supplementary Information For

Parameterization of Single Scattering Albedo (SSA) and Absorption Angstrom Exponent (AAE) with EC/OC for Aerosol Emissions from Biomass Burning

Rudra P. Pokhrel[1], Nick L. Wagner[2], Justin M. Langridge[3], Daniel A. Lack[4], Thilina Jayarathne[5], Elizabeth A. Stone[5], Chelsea E. Stockwell[6], Robert J. Yokelson[6] and Shane M. Murphy[1]

[1]Department of Atmospheric Science, University of Wyoming, Laramie, Wyoming, USA
[2]NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, Colorado, USA
[3]Observation Based Research, Met Office, Fitzroy Road, Exeter, EX1 3PB, UK
[4]Transport Emissions, Air Quality and Climate Consulting, Brisbane, Australia
[5]Department of Chemistry, University of Iowa, Iowa City, Iowa, USA
[6]Department of Chemistry, University of Montana, Missoula, Montana, USA

Table S1. Summary of fuels analyzed in this study and sampling locations.

Fuel	Stack Burn	Room Burn	Fuel type	Sampling location
African grass (tall)	0	1	Savana/Sourveld/Tall grass	Kruger National Park, R.S.A
African grass (short)	4	0	Savana/Sourveld/Short grass	Kruger National Park, R.S.A
Giant Cutgrass	0	1	Marsh	Jasper CO., SC
Wiregrass	0	1	Pine forest understory	Chesterfield Co., SC
Peat (CAN)	2	0	Boreal Peat	Ontario & Alberta, Canada
Peat (NC)	2	1	Temperate Peat	Green Swamp & Alligator River NWR, NC
Peat (IN)	2	0	Indonesian Peat	Central Kalimantan
Organic Hay	1	2	Crop residue	Fort Collins, CO
Organic Wheat Straw	1	2	Crop residue	Fort Collins, CO
Conv. Wheat Straw	0	1	Crop residue	Walla Walla CO., WA
Sugar Cane	0	1	Crop residue	Thibodaux, LA
Rice Straw	1	2	Crop residue	CA, China, Malaysia, Taiwan
Ponderosa Pine	5	4	Temperate Forest	Outskirts of Missoula, MT
Black Spruce	0	3	Boreal Forest	South of Fairbanks, AK
Chamise	1	1	Chaparral	San Jacinto Mountains, CA
Manzanita	1	1	Chaparral	San Jacinto Mountains, CA
Total	20	21		

Table S2. SSA, AAE, MCE, EC to OC ratio, and mass of fuel burned for all fuels measured during stack and room burns of this work. S and K represent the short and tall African grass and the number after S or K indicate the collection plots. The ± values after AAE are the one standard deviation of the slope and for EC/OC is the propagated error from EC and OC uncertainty. Errors for SSA are not shown because of space considerations, but are ± 7% of the quantity (1-SSA). For instance, the error for an SSA of 0.95 is ± 0.004 while the error at an SSA of 0.5 is ± 0.036.

		SSA						
ID	**Fuel Type**	**405**	**532**	**660**	**AAE**	**MCE**	**EC/OC**	**Mass (g)**
9	Manzanita	0.79	0.8	0.79	2.05 ± 0.37	0.93	NA	270
18	Ponderosa Pine	0.88	0.95	0.95	3.69 ± 0.48	0.918	0.062 ± 0.018	202
20	Chamise	0.79	0.78	0.83	2.52 ± 1.58	0.913	NA	250
32	Rice Straw	0.71	0.86	0.91	4.16 ± 0.11	0.943	0.111 ± 0.028	430
61	NC Peat	0.94	1	1	10.43 ± 1.11	0.683	ND	NA
62	Ponderosa Pine	0.83	0.91	0.89	2.85 ± 0.48	0.946	0.084 ± 0.016	4029
70	Ponderosa Pine	0.93	0.99	0.99	5.95 ± 0.79	0.848	0.021 ± 0.018	274
81	African Grass S2	0.72	0.78	0.81	2.25 ± 0.33	0.97	NA	436
82	African Grass S1	0.53	0.55	0.54	1.92 ± 0.27	0.978	NA	416
84	Ponderosa Pine	0.82	0.87	0.87	2.05 ± 0.23	0.925	NA	3860
87	Organic Hay	0.81	0.91	0.94	3.46 ± 0.08	0.941	NA	412
91	African Grass S1	0.84	0.92	0.87	3.05 ± 0.39	0.97	NA	580
92	African Grass S2	0.58	0.62	0.58	1.72 ± 0.08	0.972	NA	455
95	Ponderosa Pine	0.78	0.85	0.85	2.48 ± 0.08	0.933	NA	150
96	Organic Wheat	0.73	0.76	0.87	3.43 ± 1.13	0.965	0.239 ± 0.054	154
112	Canadian Peat	0.97	1	1	5.11 ± 1.58	0.811	0.011 ± 0.012	NA
113	NC Peat	0.94	0.98	0.98	3.68 ± 0.47	0.692	0.008 ± 0.007	NA
114	Indonesian Peat	0.93	1	0.99	7.44 ± 3.27	0.744	0.005 ± 0.010	NA
124	Canadian Peat	0.91	0.99	0.99	6.78 ± 0.26	0.798	NA	NA
125	Indonesian Peat	0.93	0.98	0.99	8.03 ± 2.50	0.872	NA	NA
129	Ponderosa Pine brown/green	0.92	0.98	0.98	4.92 ± 0.46	0.839	0.068 ± 0.018	200
130	California Rice Straw	0.75	0.84	0.84	2.98 ± 0.20	0.961	0.192 ± 0.031	147
131	Black Spruce	0.87	0.93	0.93	2.98 ± 0.05	0.957	0.095 ± 0.018	639
132	Organic Wheat	0.82	0.88	0.88	2.58 ± 0.01	0.956	0.127 ± 0.024	638
133	Conventional Wheat	0.79	0.85	0.86	2.67 ± 0.03	0.956	0.150 ± 0.023	494
134	Black Spruce	0.87	0.93	0.93	2.97 ± 0.12	0.957	NA	1077
135	Chamise	0.58	0.61	0.54	1.95 ± 0.06	0.954	NA	667
136	Manzanita	0.52	0.56	0.46	1.68 ± 0.09	0.959	0.711 ± 0.060	1064
137	Black Spruce	0.82	0.9	0.84	2.32 ± 0.70	0.962	NA	1602
138	Organic Hay	0.85	0.94	0.92	3.25 ± 0.79	0.95	NA	592.2

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140	Ponderosa Pine	0.88	0.94	0.91	2.21 ± 0.61	0.928	0.039 ± 0.012	1672
141	Wire Grass	0.36	0.38	0.35	0.85 ± 0.21	0.969	1.386 ± 0.113	540
142	Ponderosa Pine brown/green	0.79	0.83	0.82	2.19 ± 0.19	0.952	NA	1529
143	California Rice Straw	0.87	0.95	0.97	4.48 ± 0.05	0.939	NA	902
144	Ponderosa Pine	0.86	0.91	0.93	2.68 ± 0.21	0.927	0.037 ± 0.013	1731
145	African Grass K3	0.45	0.32	0.26	1.29 ± 0.48	0.975	1.206 ± 0.097	1078
146	Organic Hay	0.85	0.94	0.96	3.55 ± 0.08	0.937	0.027 ± 0.010	1335
147	Sugarcane	0.85	0.93	0.94	4.02 ± 0.13	0.934	0.052 ± 0.021	867
148	Giant Saw Grass	0.39	0.43	0.44	1.83 ± 0.34	0.925	2.148 ± 0.177	2000
149	Organic Wheat	0.77	0.83	0.83	2.58 ± 0.08	0.962	0.185 ± 0.029	393
150	North Carolina Peat	0.95	0.99	1	6.25 ± 0.63	0.803	NA	NA

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Table S3. Same as Table 5 of main text, but for 405 nm.

Biomass Types	MCE[1]	BC/OC[1]	SSA_405 MCE Approach	SSA_405 EC/OC Approach	SSA_405 EC/(EC+OC) Approach	% Difference In Predicted SSA
Tropical Forest	0.95	0.11	0.74	0.81	0.83	-9 /-12
Savanna	0.96	0.14	0.63	0.78	0.80	-24 /-27
Crop Residue	0.94	0.33	0.77	0.69	0.70	10 /9
Pasture Maintenance	0.92	0.09	0.84	0.82	0.84	2 /0
Boreal Forest	0.92	-	0.84		-	-
Temperate Forest	0.95	-	0.73		-	-
Extra tropical Forest	0.93	0.07	0.83	0.85	0.86	-2 / -4
Peat land	0.9	0.03	0.90	0.89	0.89	1 /1
Chaparral	0.96	0.35	0.64	0.68	0.68	-6 /-6
Open Cooking	0.95	0.29	0.70	0.71	0.72	-1 /-3
Patsari Stoves	0.97	0.39	0.54	0.67	0.67	-24 /-24
Charcoal Making	0.86	0.03	0.93	0.89	0.89	4 /4
Charcoal Burning	0.93	0.77	0.82	0.55	0.53	33 /35
Dung Burning	0.89	0.29	0.90	0.7	0.71	22 /21
Garbage Burning	0.97	0.12	0.54	0.8	0.82	-48 /52

[1]Data from Akagi et al., 2011

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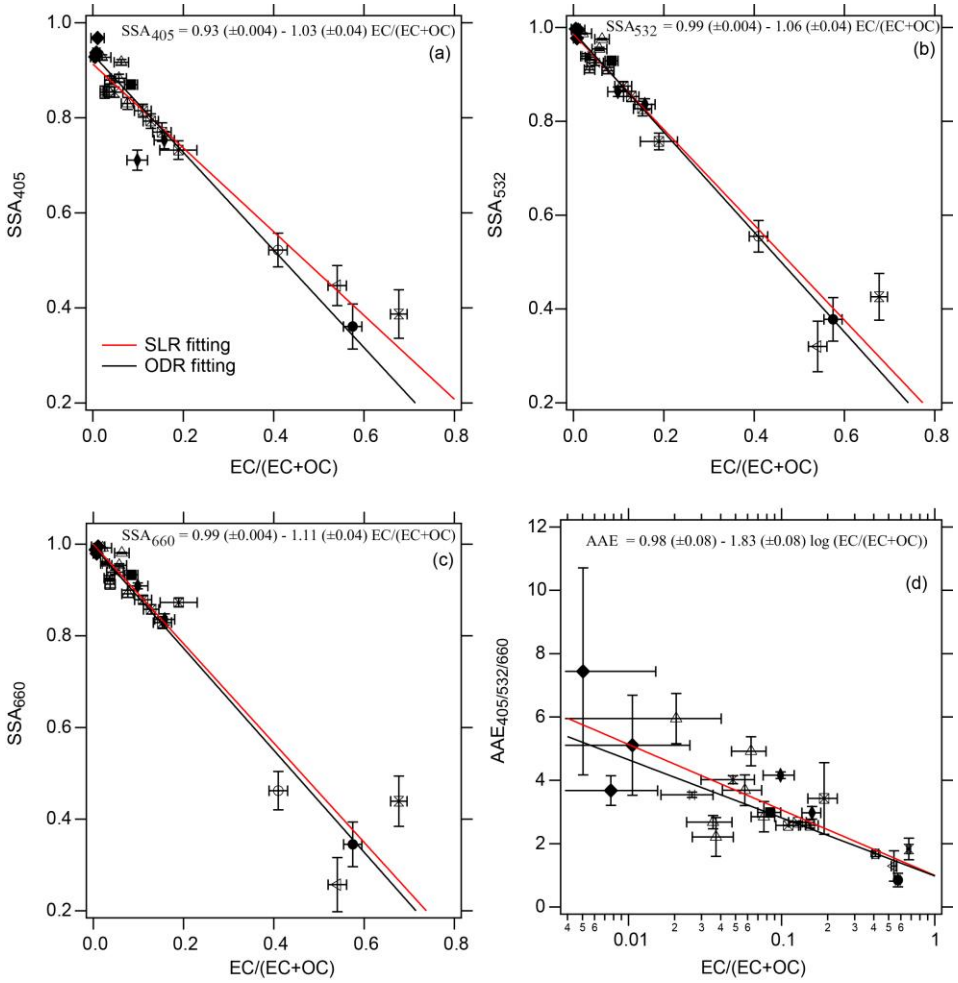

Figure S1. SSA at (a) 405 nm, (b) 532 nm, (c) 660 nm, and (d) AAE plotted as a function of EC/(EC+OC). Solid red lines are simple linear regression fitting lines and solid black lines are orthogonal distance regression fitting lines. Equations presented in each figures are the ODR best fit equations for respective SSA and AAE.

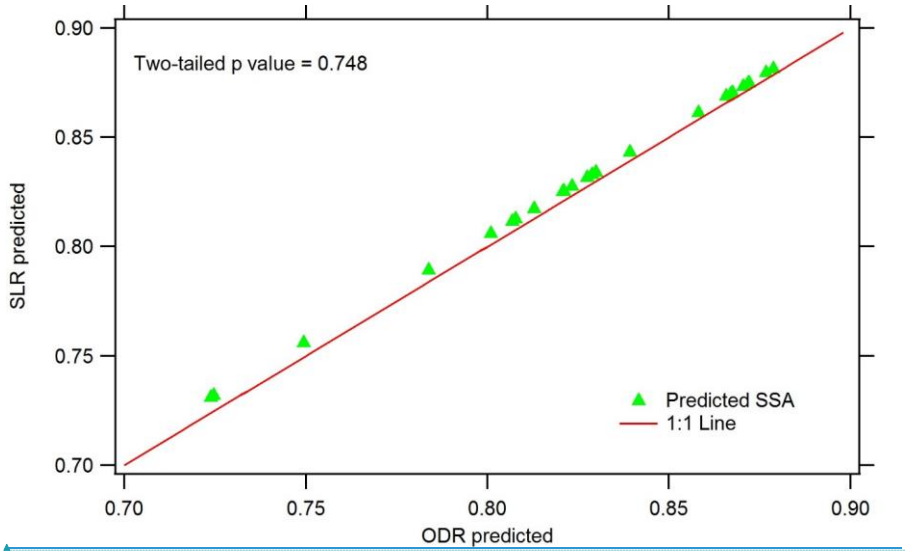

Figure S2. Comparison of predicted SSA values based on SLR and ODR models. Solid red line is one to one line.

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