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: $\Delta BC/\Delta PM_{2.5}$ was converted in to $EC/(EC+OC)$ by setting the OC mass fraction to $39 \pm 9 \%$ 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.

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