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:

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