

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 CO₂ 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 k_0 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 $\pm 5\%$...” 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:

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