

Interactive comment on “Measurement of size dependent single scattering albedo of fresh biomass burning aerosols using the extinction-minus-scattering technique with a combination of cavity ring-down spectroscopy and nephelometry” by Sujeeta Singh et al.

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Responses to questions and comments by Rudra Pokhrel

COMMENT: Authors wrote “The SSA and AAE values in this work do not fit well with current schemes that relate these factors to the modified combustion efficiency (MCE) of a burn” but they have not done proper validation or check because they don’t have MCE calculation but present as a major finding in their abstract. They concluded burn

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condition does not control the SSA and AAE and mention as a one of the major findings in abstract but have not tested SSA and AAE correlation with either MCE or BC/OA ratio. Recently Pokhrel et al. (2016) shows BB SSA shows very strong correlation with $EC/(EC+OC)$.

AUTHORS RESPONSE-The authors did not have the capability to measure CO and CO₂ to derive MCE at the time of this work, nor was MCE recognized as being important until much later. We also lack the capability to measure EC and OC content. We are currently working to address these important aspects of BB aerosols, including chemical analysis and optical properties as a function of aging using an indoor smog chamber in forthcoming work. We thank Pokhrel for bringing his work to our attention. However, the authors did not conclude that combustion conditions and MCE have no effect on SSA or AAE. We recognize that these factors (including fuel type, fuel state, and burning process) are important aspects of BB. The focus of this work was on fuel type. The discussion of the conclusions in question can be found on lines 439 through 446 on page 13. In short, such large AAE values were only seen in the scheme of Liu et al. at SSA values >0.85 at 532 nm, though much smaller SSA values were observed in our work. A lack of SSA wavelength dependence was only observed below an MCE of 0.92, though the absolute values of SSA were very low and correspond to MCE values greater than 0.92; it cannot be both. The lack of spectral dependence in SSA was based on the fitting parameters found in Liu et al. It appears that Pokhrel et al. arrived at different coefficients, though it is not clear why “allowing the coefficients to vary” would produce this result. The authors will determine if this argument is still consistent with the coefficients found in Pokhrel et al.

For white pine, we observed AAE values from 2.17-4.20. This would correspond to a $EC/(EC+OC)$ less than 0.2, as shown in Figure 4 of Pokhrel et al. However, Pokhrel et al. show SSA values greater than ~ 0.75 at 532 nm for $EC/(EC+OC)$ less than 0.2. Our observed SSA values range from 0.46-0.74, which is only seen at $EC/(EC+OC)$ significantly greater than 0.2. The flaming stage of cedar does not seem to be as

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problematic in EC/(EC+OC)-based scheme. These conclusions will be included in the final paper.

COMMENT: How authors divide flaming and smoldering stages of fires?

AUTHORS RESPONSE-This differentiation was done visually. The visual differentiation was based Tillman's description of combustion of wood as three distinct, but overlapping stages (Tillman 1981). The first ignition stage involves use of heat to drive off moisture and bring the wood to the pyrolysis temperature. In the second, stage (flaming), the wood undergoes pyrolysis (thermal decomposition under oxygen-poor conditions) when it reaches around 500-600°F. This process leads to production of organic gases with increasing high molecular weight as combustion progresses. The remaining portion is charcoal, which burns at about 1100°F. Once volatiles are driven off, direct combustion of black carbon occurs (smoldering). Any unburned gas-phase residue will be in the form of smoke or condensed pyrolysis gases. Complete combustion requires plenty of oxygen and the three elements of temperature, turbulence, and time.

Tillman, D.A., Rossi, A.J., Kitto, W.D., (1981). Wood Combustion: Principles, Processes, and Economics. Academic Press: New York, NY.

COMMENT: Page 7 line 249: Author's mentions "even though the size distribution did not change over the course of weeks, but observed decrease in optical values suggests that could be due to changes in chemical properties of the soot, but in line 241 they wrote that result in this work will only be compared to literature observation of fresh shoot". How could chemically change soot be compared with fresh soot?

AUTHORS RESPONSE-The authors recognize that some chemical changes had occurred over the course of several weeks, and took measures to account for these changes. For the most part, these changes seem quite slow. The samples were kept in distilled water and not exposed to the environment. We have several reasons for comparing our values to fresh BB soot. While some changes in the particle size distri-

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bution were observed upon impingement, this sampling scheme (or, really, any offline sampling scheme) is unlike any natural atmospheric processing. No photochemical changes were allowed to take place. The system would be too dilute for SOA formation via acid chemistry. Semi-volatile species would be almost immediately put into the condensed phase, though solubility would be a more important factor in determining their interactions with the particulate phase. The consistency of the measurements (with very small changes) done following sample collecting and several days and weeks later also gives us confidence that the chemical change is not significant. In short, our samples had more in common with the physical and chemical properties of fresh BB soot than processed soot, so our results were put in the context of fresh BB soot observations.

COMMENT: Page 8 line 255: what is the logic behind to adjust 580-660 nm range, not 500-580 nm range?

AUTHORS RESPONSE-The authors acknowledge that some more experimental detail is needed regarding this topic. Our main reasoning for this was that data in the 580-660 nm had poorer S/N than data in the 500-580 nm range. This is due to the smaller reflectivity of the mirrors in that range. The values for extinction, scattering, and absorption cross sections were high in the 580-660 nm range for 300 nm particles and low for 400 nm particles but maintained the same slope. For the same-day run for both wavelength ranges, we found nearly the same values for the 400 nm particles. In all cases the SSA did not change significantly due to adjusting the extinction and scattering values. Measurements were done several times at different days and the results are consistent.

COMMENT: Page 10 line 329: Author's mentions Cedar have higher SSA at flaming state than the smoldering stage for 300 nm particles. What is the [reason] behind this? Do authors want to say organics absorb more than black carbon in 500-660 nm? Since more BC is produced during flaming and more OC is produced during smoldering (Ward et al., 1992).

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AUTHORS RESPONSE-Only 300 nm diameter particles exhibited a higher SSA in flaming stage than smoldering stage. Several others had SSA values that were indistinguishable between stages. The reviewer is correct, in that more discussion is warranted for these findings, and revisions to the paper will reflect that. Given that we do not have insight into the relative contributions of OC and EC, saying that OC is more absorbing than EC would be pure conjecture. While the authors agree with the conclusions of Ward et al. and many others, it is important to recognize that their observations were done over the entire size distribution, or at least the entirety of PM_{2.5}. It has been shown that the smoldering phase emits larger, higher SSA particles (Reid et al., 2005). It is likely that at small particle diameters, such as 300 nm, EC has a greater contribution to than OC, giving rise to lower SSA values for the smoldering stage of this fuel at this size. To make up for this, larger particles could have a greater contribution of OC, resulting in its greater abundance in PM_{2.5}. This is consistent with the larger observed SSA values for larger diameter particles in the smoldering stage of cedar combustion.

COMMENT: Page 10 line 351-354: Author's mentions there was no observable trend of SSA as a function of particle size and again wrote SSA diverged significantly at larger diameters. What do these sentences signify? And in Line 353 they mention particle size clearly plays the major role in determining the scattering or absorption properties. How that does not affect the SSA?

AUTHORS RESPONSE-It is not to say that there are not differences as a function of particle size, but there lacks a trend for the flaming stage. For the smoldering stage, as noted earlier in that paragraph, SSA increased with particle size for each fuel investigated. For the flaming stage, there is no systematic change of SSA with particle size.

COMMENT: Page 11 Line 394: Author's concluded that due to lack of variability in their SSA values for different wavelength the MCE of the burns for their work is < 0.92 based on Liu et al. (2014) study. If so, then why they called flaming for such burns? Based on Yokelson et al. (1996) definition, MCE of 0.9 represents roughly equal amount of

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flaming and smoldering and MCE ~ 0.8 are pure smoldering.

AUTHORS RESPONSE-Liu's results show that SSA varies strongly with fire-integrated modified combustion efficiency (MCE)—higher MCE results in lower SSA values and greater spectral dependence of SSA. SSA values between 0.6 and 0.8 correspond to MCF values between 0.9-0.95 for all samples at $\lambda = 405\text{nm}$, 532 nm and 781nm . As the reviewer indicated for pine for example for flaming MCE = 0.990, for smoldering MCE = 0.835, and MCE = 0.96 for total fire integrated (Yokelson et al. 1996). Other earlier measurements are also consistent with these results, For example Reid et al. (2005) defined as MCE > 0.9 for flaming combustion, MCE < 0.9 for smoldering combustion and McMeeking, et al. (2009) had MCE = 0.80 smoldering phase of the fire and MCE = 0.99 for the flaming phase. Our conclusions were based on the most recent results of Liu et al. There is no statement on line 394 suggested by the reviewer calling it a flaming burn.

Reid, J. S., R. Koppmann, T. F. Eck, and D. P. Eleuterio (2005), A review of biomass burning emissions part II: Intensive physical properties of biomass burning particles, *Atmos. Chem. Phys.*, 5, 799 – 825. Gavin R. McMeeking et al. (2009), Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory; *Journal of Geophysical Research* 114, D19210 doi:10.1029/2009JD011836,

COMMENT: Page 12 Line 398-400: Author's mention that higher SSA values in field measurements than their observation suggest that the MCE values of wildfires are higher than controlled laboratory burns. They did not explain why burn with higher MCE could have higher SSA values. It is clear from laboratory studies that burns with higher MCE have lower SSA (Pokhrel et al., 2016; Liu et al., 2014; McMeeking et al., 2014) supporting the fact that more BC will produce during flaming stages of burns (Ward et al., 1992).

AUTHORS RESPONSE-The reviewer is correct. We will amend the text, stating "This could suggest that the MCE values of wildfires are lower than controlled laboratory

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burns.” This is likely due to the lower abundance of oxygen in wildfires, and the revised text will mention this as well. Yucatan Peninsula results will also be mentioned earlier in the paragraph. The remainder of the text stands.

COMMENT: Page 12 Line 408-410: Author’s mentions, despite of having similar burn conditions with Hopkins et al. (2007), they found different SSA values for white pine than that of ponderosa pine needles/twigs. But they do not mention how they compare the burning conditions because they don’t have either BC/OA or MCE calculations in their study.

AUTHORS RESPONSE-The authors are using the phrase “similar burning conditions” fairly loosely. In Hopkins et al. the fuel was simply burned on a platform. While this work was employed a burning drum, air flow was not especially restricted. This is opposed to other significantly different burning conditions, such as the reduced oxygen conditions of a forest fire or the high efficiency combustion done in a cook stove.

COMMENT: Throughout the document, authors wrote soot. Do they want to say only soot produced during biomass burning?

AUTHORS RESPONSE-The authors recognize that discussion is limited to soot produced from biomass burning (BB), and will make a statement at the end of our introduction regarding this. There are some discussions that apply to soot more generally from other sources (diesel exhaust, acetylene flames, etc.). We will review our use of the word “soot” throughout the document and specify BB soot if it aids in understanding and reduces confusion. However as pointed out by Buseck et al. (2012), there still exists ambiguity on the definitions of soot, black carbon, and carbonaceous aerosols.

Buseck, P. R.; Adachi, K.; Gelencsér, A.; Tompa, É.; Pósfai, M., Are black carbon and soot the same? Atmos. Chem. Phys. Discuss. 2012, 12 (9), 24821-24846.

Other things: We will remove the words “or mineral dust” from line 387 and add to the phrasing of the next sentence.

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