

Response to reviewer #1:

We thank the reviewer for their comments on the article. We've responded to the individual comments below.

Summary

The paper summarizes the physical and optical properties of aerosols from biomass smoke from regional to continental scale events. The paper is appropriate, well focused and should eventually be publishable in ACP. I recommend the following minor modifications and additional analysis prior to publication.

Technical Comments

Techniques and analysis seem sound. The criteria for smoke impacts and differentiating LRT and regional smoke events with water vapor seems well-thought out.

Certainly the trend is consistent of lower SSA for the Siberian fires and thus a flaming, higher MCE fire. However, all the SSA values are all relatively high suggesting an MCE on the lower end of the range (mixed to smoldering combustion). This is worth commenting. For reference see the Liu paper below.

Response: This is a good point as all of the SSA's we observed were >0.95 , but low SSA values seem to be only observed in studies of primary emissions. Vakkari et al. (2014) and Yokelson et al. (2009) observed SSA to increase significantly with aging in wildland fire plumes within an hour of emission. In addition, at MBO Briggs et al. (2016) found that BB plumes do not follow the SSA vs MCE parameterization from Liu et al. (2014) and that all SSA values were >0.92 despite very high MCE values. These high SSA values were attributed to SOA formation and increased scattering efficiency driven by particle growth. Given this and that fact that MCE couldn't be accurately derived due to the high CO₂ background, I'm hesitant to say that the equate the relatively high SSA values for the Siberian plumes, which have been atmospherically processed for up to 10 days, to mixed to smoldering combustion.

I have added a paragraph to address this in Section 3.3 (Lines 302-315).

The analysis brought to mind a recent paper by the CSU group examining emitted and aged biomass smoke sizing and radiative properties paper referenced below which may provide a useful intercomparison and context.

Response: The Carrico et al. (2016) paper is a useful paper to compare with our paper. We cited it in comparison with our results for size distributions in Line 388.

Table 1 is useful, however would be more useful with the following additions:

- A summary mean \pm s.d. for the regional versus Siberian events, maybe 2 lines at the bottom

Response: This is a good idea and was added to Table 1.

- Adding in your rough estimate of the age of the plume for each case which was stated as a range elsewhere. **Do the size distributions with Aitken modes correspond to the younger plumes? Are there any other conclusions to be drawn?**

Response: We looked into this but it was too ambiguous to draw any definitive conclusions. Most of the regional BB events were influenced by multiple fires which made it nearly impossible to properly estimate plume age. Additional language to explain this was added in Section 3.2 (Lines 256-260), and Figure 3 was added for a visual explanation.

As far as the events with bimodal distributions, 3 of the 5 are of Siberian origins. Given this we suspect the Aitken mode in these size distributions most likely represents a secondary source from within the boundary layer. We have put this in Line 371.

Figure 4. I'm not sure how the percentiles are done with such small numbers of samples, symbol with whiskers showing the range seems more appropriate. You're really comparing the Siberian to regional fires, why separate into 3 groups? I could only see one small outlier symbol on the chart.

Response: Figure 4 (now Figure 5) was changed to 2 groups, Siberian and Regional fires. Due to the outlier for the Siberian BB AAEs we feel the box plots give a more accurate portrayal of the distribution on the values rather than a mean and whiskers showing the range

Figure 5. Meaning of this? The events symbols are not distinguishable; I would simply delineate Siberian vs. regional with different symbols and colors. With the exception of CO, these parameters are by definition or calculation interdependent. Is the take home message something along the lines of, "Biomass smoke events as indicated by elevated CO concentrations featured shifts to larger sizes driving higher PM mass concentration, light scattering coefficients, and the highest overall mass scattering efficiencies."

Response: Figure 5 was removed and replaced with Figure 7, which is a similar plot but with D_{pm} on the x-axis. This looks at the dependence of different variables on size distribution instead of MSE. For Figure 7 all the events were changed to the same color and not individually identified since D_{pm} was shown not to depend on transport time.

The previous plots showed a correlation between MSE and σ_{scat} , PM1, and CO, all of which can be thought of as surrogates for plume concentration. Since σ_{scat} , PM1, and CO are all correlated with size distribution (Figure 7), the correlations with MSE are likely just a function of particle size and cause no causal effect. In summary it seems the more concentrated the BB plume (higher σ_{scat} , PM1, CO) the larger the size distribution, which in turn increases the MSE.

Given this, we decided to highlight the correlation between σ_{scat} , PM, CO versus D_{pm} instead of MSE.

Language regarding the new figure is in Lines 350-357, and 390-398.

Mechanics and Presentation

The presentation is appropriate in terms of length, style and diction. Figures are appropriate.

Why put the hysplit trajectories plot in supplementary material though? The CALISPO images are appropriately in the supplement. However, the paper is short enough it can accommodate the additional figure rather than the annoyance of looking elsewhere.

Response: This Figure was moved to the main manuscript. It is now Figure 6.

I noted a few inconsistencies (line 158 and 196 for example) in variable, citation, and subscript italics, check throughout.

Response: The manuscript was checked for inconsistencies and proofread.

Line 109 “was located prior to any. . .” Aerosol instrumentation?

Response: This was changed

Line 133, I recommend breaking out as an equation rather than inline.

Response: This was changed

Line 192, I believe you mean Period 2.

Response: This was changed

Line 209, “ascended from the boundary layer (BL) to. . .” MBO?

Response: This was changed

Line 242 superscript missing

Response: This was changed

Line 280 “hygroscopy” replace with hygroscopicity Line 299 “Mei” replace with Mie

Response: This was changed

Line 375 “preformed” replace with performed

Response: This was changed

Liu, S., et al. (2014), Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, *Geophys. Res. Lett.*, 41, 742–748, doi:10.1002/2013GL058392.

Carrico (2016) - Rapidly evolving ultrafine and fine mode biomass smoke physical properties: Comparing laboratory and field results, *JOURNAL OF GEOPHYSICAL RESEARCH: ATMOSPHERES*, Volume 121, Issue 10, 27 May 2016, Pages: 5750–5768