

The authors' effort to address the comments are acknowledged and appreciated. However, I still believe that my two major concerns from the previous review still hold. I think it is best to make my points by doing the following simplified calculation.

Assuming no effect of internal mixing (just to simplify the calculation), one can calculate MAC of the aerosol (BC + OA) as a weighted average of the MACs of the components:

$$MAC_{BC+OA} = MAC_{BC} C_{BC} + MAC_{OA} C_{OA} \quad (1)$$

Where C_{BC} and C_{OA} are the concentrations. This can be written in terms of BC-to-OA ratio as:

$$MAC_{BC+OA} = MAC_{BC} (1 + BC\text{-to-OA}) + MAC_{OA} \left(1 + \frac{1}{BC\text{-to-OA}} \right) \quad (2)$$

And AAE is:

$$AAE_{BC+OA} = \frac{d\ln(MAC_{BC+OA})}{d\ln(\lambda)} \quad (3)$$

Solving equations (1-3), one can mathematically reproduce the data in Figure 4 and SI Figure 1 (Figure 6 in the authors' response).

The blue curve in Figure R1 was calculated using $MAC_{BC} (532nm) = 10 \text{ m}^2/\text{g}$, $MAC_{OA} (532nm) = 0.2 \text{ m}^2/\text{g}$ [just an assumption based on general values in the literature. You can use different values, but won't change the general picture], $AAE_{BC} = 1.1$ [the value assumed by the authors], and $AAE_{OA} = 6$ [chosen to reproduce measurements at low BC-to-OA].

The red curve was calculated using the same values, with the only difference being $AAE_{BC} = 1.8$. Let's put the difference between the blue and red curves aside for now. Looking at any of the two curves, one can see that AAE_{BC+OA} increases with decreasing BC-to-OA ratio in a fashion very similar to what the authors report, simply due to the decreased contribution of BC to AAE. In other words, a constant AAE_{OA} can explain the data (whether that OA is primary or secondary does not matter). Therefore, the authors' conclusion that "brown" aerosol is exclusively secondary OA (e.g. line 296 and line 455) does not necessarily follow from the data. The correlation with f44 and the droplet mode is not enough. All they can say is that aged OA contributes to the brown aerosol, but they cannot say that the brown aerosol is exclusively secondary (unless they show that all OA is secondary, which I don't think is the case).

Now to the second major point concerning AAE_{BC} . In the previous review, I made the point that AAE_{BC+OA} should converge to AAE_{BC} at large BC-to-OA ratios. This is supported by the calculations shown in Figure R1. The authors assume AAE_{BC} of 1.1 in their analysis, while their data (Figure 4 and SI Figure 1) clearly show that AAE plateaus at ~1.8 at large BC-to-OA ratios.

They explained this discrepancy in the revised manuscript as “*due to any spectrally light absorbing material that the AMS could not detect (refractory material, or material in particles smaller than 100 nm and larger than 1 μm).*”

This is not convincing. First, what is the light-absorbing refractory material with such a high AAE (it needs to be $\gg 1$ in order to have such a big influence)? It could be dust, but the authors say that they exclude data that had contribution from dust.

Second, let’s assume that the contribution is from OA particles that the AMS could not see (too small or too large particles). That would mean the AMS missed A LOT of OA mass. This can be explained by looking at the difference between the blue and red curves in Figure R1. The red curve is very similar to the authors’ data (e.g. Figure 4 in the manuscript). AAE plateaus at 1.8. If AAE_{BC} is 1.1, that would mean what the authors report as BC-to-OA = 20, should actually be 0.5 (where the dashed black line intersects the blue curve in Figure R1). Of course, this calculation is simplified, but the point is that the BC-to-OA has to be grossly underestimated (at least an order of magnitude) for the authors’ explanation to hold.

I don’t think this is the case. The more logical explanation is that the AAE measurements, for some reason, are overestimated by ~80%. And as I pointed out in the previous review, this would explain the unusually large AAE_{BC+OA} reported in this study.

I think the authors should try to address this bias, or at the very least clearly state it in the manuscript and discuss the implications.

Finally, it is not clear why the authors define “brown” aerosol as something different than brown carbon. Do they mean that there are non-organic (non-dust) components that are also brown? If yes, they need to justify. If not, it seems to me that brown aerosol and brown carbon are synonymous.

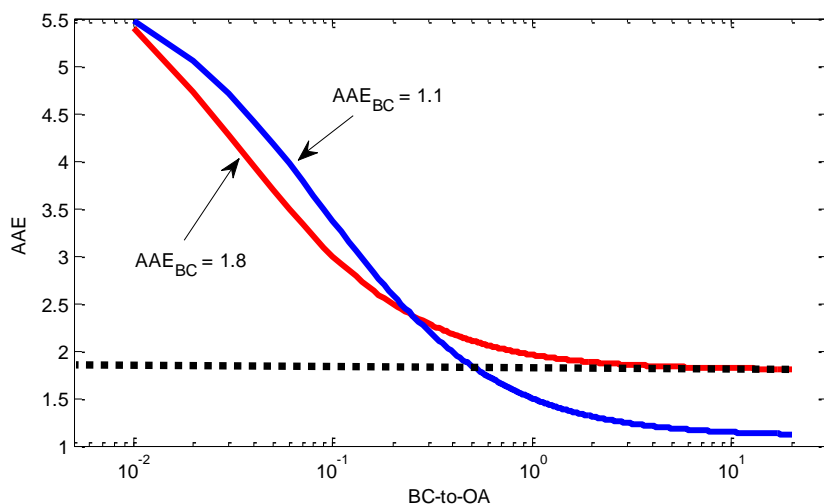


Figure R1