**Response to Reviewer #1**

We thank the reviewer for the thoughtful comments. The reviewer raises as a primary concern the measurements of the small absorption enhancement observed at 781 nm and the relationship with the coating-to-core ratio for BC-containing particles, positing that there may be some measurement bias that is leading to a strong deviation from the core-shell behavior.

The primary argument provided by the reviewer is that the SP-AMS might be biased because coatings may not effectively vaporize if they are not engulfing the BC. Certainly this is a possibility. However, we note that if this occurred it would lead to a negative bias and thus the reported coating-to-core ratios would be a lower bound. This would seem to go opposite to the reviewers concern; if the actual coating-to-core ratios were even larger than reported then the disparity only increases. It is true that there is a differential sensitivity of the SP-AMS to BC compared to coating materials owing to how the particle beam overlaps with the laser beam in the instrument (Willis et al., 2014). However, this is more important for absolute quantification than it is for relative quantification (i.e., coating-to-core ratios). Based on Willis et al. (2014), if no accounting of the coating dependence of the detection were accounted for, such detection issues could lead to a bias of ca. 30% in the coating-to-core ratios. While important, such a bias would not materially affect the conclusions here.

We also note that there is an abundance of evidence in the literature for non-core-shell morphologies for fresh biomass-derived BC-containing particles. A just published paper from Adachi et al. (2019) shows images of BC attached to other material, consistent with some of their previous work (Adachi and Buseck, 2008;Adachi et al., 2010;Adachi and Buseck, 2011) and with various other similar measurements (Chakrabarty et al., 2006;China et al., 2013;Torvela et al., 2014). Using an SP2, Sedlacek et al. (2012) observed evidence of non-core-shell morphologies for BC-containing particles in a biomass burning plume, with the fraction of such particles >60% even in a somewhat aged plume. Also with an SP2, (Pan et al., 2017) observed fresh biomass combustion-derived large BC, averaged over many different fuel types and for large (>200 nm diameter) BC cores, exhibits a wide range of estimated shell-to-core diameter ratios and "delay times" that correspond to coating-to-core mass ratios of ca. 0.1 to 3.

One might ask why BC-containing particles from biomass combustion would not readily adopt core-shell morphologies, as the above cited experimental evidence suggests? We suggest BC and coating material existing in the same particles most likely results from near-source coagulation. Sedlacek et al. (2015) observed formation of non-core-shell morphologies from coagulation, albeit not for biomass burning derived particles.

Additionally, it should be considered that the SP-AMS coating-to-core ratios reported here are bulk averages and do not account for the different mixing states of BC-containing particles. The distribution of coating material across the population of particles impacts the absorption enhancement, even when core-shell morphologies are assumed (Fierce et al., 2016). This was also shown in Cappa et al. (2012) for a simple 1:1 bimodal mixture of a mode having smaller coating-to-core ratios (1 or 0.1) and one having values that allowed for matching of the observed coating-to-core ratio. For mixtures of this sort, the predicted absorption enhancement is smaller than obtained if all particles are assumed equivalent. Thus, the issue is not simply one of morphology, but of particle-to-particle mixing state.

The reviewer asks for "convincing theoretical discussion." We lack sufficient information regarding the mixing state (i.e., the distribution of coatings with respect to the BC particle population) and internal

morphology to robustly calculate theoretical absorption enhancements for our experiments. However, example calculations following the approach of Cappa et al. (2012) can give an indication of what conditions might give rise to limited absorption enhancements, even at relatively large bulk-average coating-to-core ratios. Theoretical absorption enhancements, assuming core-shell morphologies, have been calculated for a binary population of particles, with one population "thinly" coated (with coating-to-core = 0.1) and one "thickly" coated (with variable coating-to-core ratios). The fraction of thickly coated particles ($f_{thick}$) was varied from 1 (for which the thickly coated coating-to-core ratio equals the bulk average) to 0.01. We have assumed a BC core diameter of 150 nm with a complex refractive index of 2.0 + 1.0*i*. The complex RI for the coating was assumed as $1.5 + 10^{-8}i$. The absorption enhancement was calculated for each assumed $f_{thick}$ as a function of the bulk-average coating-to-core ratio. The results of these calculations are compared with the observations in Fig. 1a, and the variation in $E_{abs}$ with the $f_{thick}$ for three different bulk-average $R_{coat\text{-}BC}$ values (= 1, 5, and 10) are shown in Fig. 1b. These example calculations indicate that when the population is skewed towards most particles being thinly coated the theoretical $E_{abs}$ can be quite small even when the bulk-average $R_{coat\text{-}BC}$ is large. We note again that these calculations assume a core-shell morphology, so deviations from core-shell would serve to reduce these values further, that is the calculations here are upper-limits. Measurements by Liu et al. (2017) indicate that the core-shell approximation fails for particles having coating-to-core ratios < 3, and even above this value there may still be reductions owing to non-core-shell morphologies. Further, we have considered only a simple binary mixture of thinly and thickly coated particles. Consideration of more complex distributions of material across the BC population would lead to further reductions in the calculated absorption enhancements.

All this is to say that there is a strong experimental and theoretical foundation for observing absorption enhancements lower than the core-shell approximation. Given the simplistic nature of the calculations presented here, the assumptions that go into them, and a lack of experimental constraints regarding the particle mixing state for the particles sampled, we hesitate to add too much of this discussion to the manuscript. Nonetheless, in the revised version we intend to expand somewhat the discussion on Page 10 where we already indicated that the relatively low and constant $E_{abs}$ at 781 nm likely results from a combination of mixing state and morphology effects.

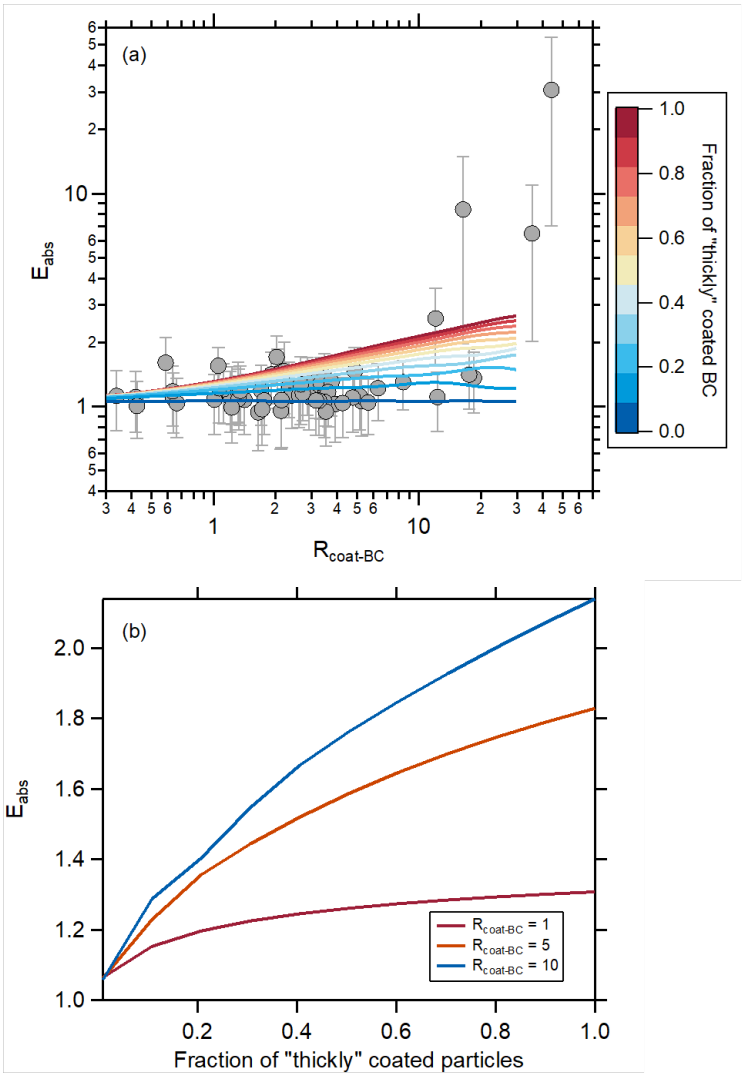


**Figure 1.** (a) The observed (points) absorption enhancement at 781 nm, calculated as the observed mass absorption coefficient divided by the reference value at the limit of no coating, and the calculated (lines) absorption enhancements from core-shell Mie theory as a function of the coating-to-core ratio. The different color lines correspond to different assumptions regarding the fraction of "thickly" coated BC. (b) The calculated absorption enhancement from core-shell Mie theory as a function of the fraction of "thickly" coated particles for three different bulk-average coating-to-core ratios.

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