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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Brown carbon: a significant atmospheric absorber of solar radiation?" *by* Y. Feng et al.

Anonymous Referee #1

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Summary: The authors include "brown carbon" in a global circulation model and attempt to assess the impacts of this BrC on global radiative forcing. They first compare their model results, particularly the absorption Angstrom exponent (AAE) to values derived from AERONET, and seem to conclude that inclusion of BrC leads to better agreement, despite substantial inconsistencies in certain regions (S. America and S. Africa). There is also a misunderstanding of how model AAE values, which are based on spherical particle Mie theory calculations, can be compared with ambient measurements. Their base model formulation gives an AAE = 0.53, which is unrealistically low compared to observations and is simply a function of using spherical particle theory to describe particles that are non-spherical. This means that the model AAE starts from too low a value (0.53 instead of closer to 1), and thus increases in the model AAE due to the inclusion of BrC are starting from the wrong reference point. Further, I find that

there is much that is not explained sufficiently.

Certainly, understanding and quantifying the influence of BrC on climate is an important problem, and estimates must be made even in the face of uncertainty. This manuscript provides one such estimate. However, I have concerns that the radiative forcing estimates provided here lack appropriate uncertainty analysis (for example, specific values are given in the abstract, but it is not even clear whether these come from the strongly or weakly absorbing simulations) and I have some concerns about the fundamental model formulation.

My specific comments are below, but overall I suggest that this paper is in need of substantial revisions before it can possibly be reconsidered for publication, if it should be reconsidered at all.

Specific comments:

P2800, L8: The authors need to provide a reference and explicit justification for the use of the 1:1.4 ratio for conversion of OC to POM, accounting for secondary formation, when dealing with biomass emissions. Is this just made up?

P2800, L26: The authors state that "The emissions of BC and organic matter are from Bond et al. (2007), including fossil fuel, bio-fuel, and open biomass burning sources." However, as best I can tell Bond et al. provide emissions estimates for only "energy related" combustion sources, which does not include open biomass burning sources. So where exactly do the estimates for open biomass sources come from? Not from Bond et al. (2007).

Fig. 1: I suggest that the authors also present a map of the ratio between BC and BrC, as this is crucial to determining the contribution of BrC relative to BC.

P2802, L9: I do not see how the RI reported here for black carbon comes from Bond and Bergstrom (2006). As best I can tell, this RI is not reported anywhere by Bond and Bergstrom. Further, the authors make it look as if 1.8 + i0.74 is the value suggested by

Bond and Bergstrom. This is not the case. They suggest 1.95 + i0.79. In all likelihood, this difference is inconsequential to the actual calculations, but it must be noted and the associated statements corrected.

P2802, L19: The authors must state within this manuscript what BC size (or size distribution) is assumed. This is critical to the determination of the absorption cross-section and, in particular, the increase in the cross-section upon coating with non-absorbing material. Many of my comments below stem from this fundamental issue.

P2802, L29: The authors indicate that Bond and Bergstrom suggest a lower bound of 5 m2 g 1 for "observed submicron BC particles" and 7.5 m2 g 1 for freshly emitted aggregates. I can see where they recommend 7.5 m2 g 1, including for ambient uncoated particles, but I do not see where they ever recommend 5 m2 g 1. Bond and Bergstrom also make it abundantly clear that spherical particle Mie theory commonly underestimates absorption cross-sections for black carbon. Therefore, I believe that the focus on the absolute values in this section is misplaced.

P2802/2803: Continuing from my previous comment, I have substantial concerns that the authors methods lead to an underestimate in black carbon absorption simply because they start from the wrong place, namely a calculated MAC for their uncoated particles that is lower than what is likely in the atmosphere.

P2802, L22: The authors need to be more precise in their discussion of absorption enhancements, especially as they relate to brown carbon. For non-absorbing coatings, the enhancement comes from only one source: the lensing effect of the coating. However, for brown carbon coatings the enhancement comes from two places: lensing + direct absorption by brown carbon. Given this, of course the brown carbon coatings exhibit the largest enhancements. . .this is because it includes direct absorption by the BrC! Why is this important? Because if the BrC were externally mixed from BC it would still lead to an "enhancement" (if enhancement is defined as any increase above the absorption attributable to BC) whereas non-absorbing species would not.

Figure 3: I recommend the authors show a difference plot in addition.

P2803, L13: Looking at Fig. 3, I would conclude that over Asia the "hotspot" is only approximately captured: there appears to be a distinct offset in the exact location within East Asia.

P2804/Figure 4: The authors need to make clear that the SSA measurements sample a different subset of data points than the AOD measurements. My understanding is that AERONET can only extract SSA when the AOD > 0.2. Thus, the data considered are biased in some way towards regions/periods with large optical depths.

P2804, L7: By "with a coefficient of 0.51" do the authors mean specifically with an r value of 0.51? Thereby making r2 = 0.26, meaning that the correlation is pretty poor (which is evident just by looking at Fig. 4). Also, it is evident that no fit line is actually on the graph, only the 1-1 line. Since they state that the results "correlate" with a "coefficient of 0.51" they should provide their best fit line and the slope associated with this line. It will most certainly not be 1. In fact, I suggest that the report the slope and the r2 value for each of the different model runs (with strongly absorbing OC, moderately absorbing OC, no absorbing OC) in a table, as well as show the model results in different panels. The overlapping points are exceptionally difficult to distinguish in Fig. 4. Better yet, they would report the values for each model run for each region considered in the discussion, as well as for the global average.

P2804, L9: The authors note that the mean model error and bias in SSA is 2% and 3% respectively. However, given the poor fit in Fig. 4 I do not feel that these are really the most descriptive metrics. I suggest that the authors provide a more robust description of their model/measurement agreement that includes the actually poor relationship and doesn't seem to be trying to pass off what I would take as poor agreement as good agreement just because the mean agrees well. In other words, I find this somewhat misleading.

Fig. 5: The citation to Kirchstetter in the caption is unnecessary.

Fig. 5: The authors should also present the fraction of AAOD attributable to BrC, and not just the increase in AAOD. Fractional metrics here provide greater context for understanding what is going on, while absolute changes are useful in characterizing the radiative impacts.

P2805, L8: The authors note that the mean calculated AAE without BrC is somewhat less than the AAE inferred from Aeronet (0.9 vs. 1.2). However, it is not made clear whether this is significant or not. The AAE one calculates for BC depends on the size used to represent the BC, as well as the extent of coating (see Gyawali et al., Atmos. Chem. Phys., 9, 8007–8015, 2009 or Lack and Cappa, Atmos. Chem. Phys., 10, 4207–4220, 2010). Since no information is given in this MS regarding the uncoated BC size distribution or the mean coated BC size distribution, it is difficult to compare. But the results from these two studies make it clear that AAE values that are only slightly greater than 1 cannot be taken as evidence of BrC since this cannot be separated from other effects on the AAE. Therefore, the conclusions arising from comparisons between the model results and Aeronet are not valid. This does not mean that the authors cannot state with some confidence that their model results indicate that the presence of BrC will, in general, lead to an increase in the observed AAE. It only means that they cannot make such direct comparisons that are valid within both the model uncertainty (dictated by the fundamental formulation of the problem) and measurement uncertainty.

P2805 and Fig. 6: It is unclear what gives rise to the broadness of the AAE for the simulations without BrC. Presumably, this is variations in coating thickness by region. This must be stated.

P2805, L20: The authors state “Although no BrC is explicitly assumed to be associated with fossil fuel combustion, the AAE distribution [when BrC is included] is in good agreement over Europe and North America, which are dominated by fossil fuel emissions.” However, they provide no explanation as to why this would be the case. They must explain this result.

P2805, General: I find that the authors do a generally poor job of distinguishing between the different calculations within the text. It is often not clear when they are referring to non, weakly and strongly absorbing BrC.

P2806 and Fig. 6: The substantial overestimate of the AAE when “strongly” absorbing BrC is assumed in the region that is most dominated by BrC from biomass burning (and which is the origin of the strongly absorbing BrC properties) is certainly a weakness of the study. Importantly, the particularly poor agreement in this region indicates that consideration of global mean values (which is done throughout the manuscript) are likely not particularly meaningful. The authors should focus on regional comparisons. For example, I don’t see much worth in comparing the global mean contribution of BrC to the AAOD at 350 nm to results from Kirchstetter and Thatcher.

P2806, L19: The authors state: “When OC is absent (OC/BC= 0), the [calculated] AAE for pure BC is approximatedly [sic] 0.53.” This should tell them that they are doing something wrong. The AAE for pure BC is not 0.53. It is much closer to 1. This has been shown experimentally many times. The calculation of a lower value is simply an artifact of applying spherical particle theory to non-spherical particles. The AAE = 1 comes about theoretically from consideration of (i) a wavelength independent refractive index for BC and (ii) consideration of small spherules, < 50 nm, that sum to give the total absorption. Obtaining a smaller value from calculations simply means that a larger particle size was used. Unfortunately, it is entirely unclear what size distribution was used in this study for BC and OC. The authors state: “Carbonaceous aerosols, BC, and OC were considered in one size bin only (radius<0.63 μm).” Looking at the Liu et al. (2005) reference, it appears that fossil fuel BC and OC actually have a distinct size distribution from biomass burning BC and OC. This does not seem compatible with the statement that these were only considered in one size bin. Further, Liu et al. (2005) indicate that the FF and BB particles actually have multiple distributions within each class with different diameters. So it is impossible to tell what the authors have done here. Since they calculate an AAE = 0.53 for pure BC, it would seem that

they must have, on average, relatively large particles in terms of the mass weighting (see again Gyawali et al., Atmos. Chem. Phys., 9, 8007–8015, 2009). If the model particles were, on average, ∼150 nm in diameter then one would calculate an AAE ∼ 0.5. But this does not make this value relevant to the atmosphere! Again, a variety of lab and near roadway studies clearly demonstrate that "pure" BC does not have an AAE this low. Instead, it is closer to 1. Consider, for example, the Kirchstetter et al. (2004) paper from which the BrC properties in this work come from. They find AAE = 1 for measurements made right next to a roadway. Or Schnaiter et al. (JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 110, D19204, doi:10.1029/2005JD006046, 2005) who find an AAE of ∼1.19 for fresh Diesel soot. These are but two examples. That the model gives AAE = 0.53 is a fundamental problem.

Related to the above comment, the authors never clarify how the pure BC AAE = 0.53, yet even the non-absorbing OC case gives a global mean AAE = 0.9. This needs to be stated.

P2806, L18: The authors use a 440 nm-675 nm pair here. But above they use 440 nm – 880 nm. Why the sudden change? They should use a consistent definition throughout, since when BrC is present the curvature in the AAOD vs. wavelength curve leads to variations in the AAE depending on the wavelength pair selected.

P2806, L21: Again, the authors do a relatively poor job of clarifying that this is for strongly absorbing BrC. I strongly suggest that they adopt some clarifying terminology, such as AAEBrC=0 and AAEBrC=strong (or something like that) so that the reader can quickly see what is being referred to.

P2806, L24: The authors state that they can estimate the "enhancement of AAOD due to BrC" relative to models that only consider BC from consideration of AAE vs. OC/BC. However, they do not include the important reference case of the same situation but for non absorbing OC. Non-absorbing OC will, by itself, lead to an "enhancement" in the absorption because the authors have used core-shell Mie theory to calculate

optical properties. Thus, their analysis is not actually providing direct information on the "enhancement of AAOD due to BrC" alone. This must be revised to include the appropriate comparison with the non-absorbing OC case.

Fig. 7 and associated text: It is entirely unclear what functional form is assumed for the "best-fit" and whether it has any physical relevance (I suspect not). Further, it is really difficult to tell from looking what the density of points is. Obviously, the density of points must be highest towards the larger AAE regions, since the fit is biased towards the highest AAE values. Regardless, the fact that the intercept is 0.53 is without meaning (except to say that their model formulation is insufficient to allow for robust comparison with observed AAE values).

Section 3.3: Given all of my above concerns, I have little faith that the radiative forcing estimates provided here for BrC are robust. For example, it is clear from Fig. 6 that the AAE for strongly absorbing BrC in S. America/S. Africa is completely wrong. So why should I put stock in an estimate of the TOA forcing in this region. And again, it is often not clear when the authors are referring to the weakly absorbing case and when the strongly absorbing case (for example, the 0.25 W m 2 value on P2807, L23).

Table 4: The authors should update the AeroCom numbers to the latest work.

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