We are thankful to Reviewer #1 for his/her comments.

Reply to Reviewer 1:
In the following, the comments from Reviewer 1 are in italics followed by our responses.

1 .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.

We would like to clarify two points of this paper:

(1) First, our base model simulation is similar to most of the global aerosol model calculations, in which organic carbon is included but as scattering or weakly-absorbing aerosols only (page 2798, lines 22-25). The main goal of this paper is to examine the changes in global aerosol forcing due to change of the refractive index for a fractional organic carbon that should be considered absorbing (as BrC). As only the scattering of solar radiation by BrC is included in current global models, we investigate what effect the absorption of BrC has on the carbonaceous aerosol forcing, and to what extent.

There seems to be a misunderstanding about the lowest AAE calculated by the model. The global mean AAE (440-870nm) calculated in our control run is 0.9 for core-shell configurations with BC as core and non-absorbing OC and/or sulfate as shell, as in Table 3, and all discussions related to BrC absorption in this paper are referred to this reference value.

The following is added at the beginning of the section 3 Global results:
"3 Global results
In order to investigate the impact of BrC absorption on aerosol forcing, we conducted three global calculations of aerosol optical properties and radiative transfer, with non-absorbing OC, moderately absorbing BrC, and strongly absorbing BrC. Model results from these three simulations are referred hereafter as "NON", "MOD", and "STR", respectively. The control case is the NON case, similar to most of the current global aerosol models. The two sensitivity calculations (MOD and STR) are used to estimate the low and high limits of the resulting aerosol forcing by converting some non-absorbing OC to absorbing BrC."

(2) Secondly, the value AAE (440-675nm) of 0.53 is not calculated from the Mie theory for pure BC. It is an asymptotic value extrapolated from the model calculations in Figure 7. This value of AAE =0.53 represents for an extreme case when the OC content is zero for a population of biomass burning or fossil fuel aerosols with prescribed climatological size distributions (please refer to the response to the comment #7 regarding the size distributions used). It is certainly not used as AAE for pure BC. We don't account for BC and OC optical properties as individual species, instead, we calculate aerosol optical properties for BC and OC mixtures (as well as sulfate for fossil fuel aerosol).

Figure 7 shows AAE for 440-675nm, in order to be compared with the observational study by Bahadur et al., PNAS, 17366–17371, PNAS | October 23, 2012 vol. 109 | no. 43. Bahadur et al. (2012) suggest in their Figure 2 that "the mean values of AAE1 calculated by averaging different fractions of the total frequency distribution at the different AERONET sites. We find that the mean AAE1 value asymptotically converges to 0.55 ± 0.24…" and AAE1 in Bahadur et al. (2012) refers to the absorption angstrom exponent between 440 and 675nm.

As for using spherical particle theory to describe particles that are non-spherical, we agree that it is problematic; however, it is also a common practice in global and large-scale models, and beyond the scope of this paper to address this deficiency. We now add the "assuming spherical particles" explicitly on page 2801, line 14: "Aerosol optical properties (specific extinction, single-scattering albedo (SSA), and asymmetry factor) are calculated from the Mie theory assuming spherical particles…". Recently Gaywali et al., (2013) using measurements from CARES have shown that Mie theory with spherical particles explains approximately 90% of the measured scattering. Thus, the error due to this assumption is probably small and within the uncertainty of all the other measurements discussed here from AERONET.

In S. America and S. Africa, compared to the control run with the NON absorbing OC, we did obtain better agreement with AERONET data for the MOD absorbing BrC (Figure 6b), although the inclusion of the STR absorbing BrC overestimates the AAE. Being aware of the uncertainty of using one configuration in global simulations, we use simulations with strongly abs BrC to represent the upper limit for the BrC absorption, while using moderately abs BrC to represent the lower limit. We have thus revised the manuscript to emphasize more on the range of estimated BrC absorption, rather than just the STR absorbing BrC case.

2. 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.

We improved the references to different model simulations in the revised manuscript using "NON", "MOD", "STR" for non-absorbing OC, moderately and strongly absorbing BrC, respectively. Below are the one-by-one responses to the Reviewer's specific comments.

3. 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?

This is a very common assumption made in global models to convert OC to OM, i.e., the AeroCom model inter-comparison uses the conversion factor (Kinne et al., ACP, 2006, Table 4, footnotes 4 and 6), Liu et al., JGR, Vol. 110, D18206, 2005, and many other papers in literature.

Revised as: "In the IMPACT model, the conversion factor between OC and particulate organic matter is 1:1.4 (Liu et al., 2005), to account for the non-carbon mass and secondary aerosol formation from volatile organic compounds."

4. 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).

This is corrected: "The fossil fuel and bio-fuel emissions of BC and OC are from Bond et al. (2007). BC and OC from open biomass burnings and naturally emitted OC are based on the AeroCom emissions (Dentener et al., 2006)."

5. 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.

The map of the BrC/BC ratio is added to the Figure 1:

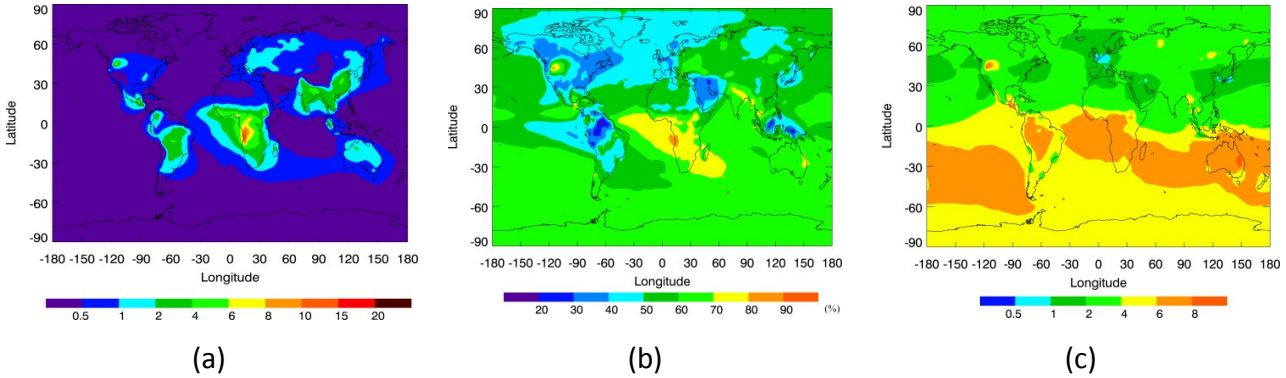

Figure 1. The estimated (a) annual mean atmospheric burden (mg m^{-2}) of BrC, (b) percentage (%) of BrC in total OC, and (c) ratio of BrC to BC

6. 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.

The value of 1.8+i0.74 is from Bond and Bergstrom (2006), Aerosol Science and Technology, 40:27-67, Figure 9, in the caption, which states: "…Central curve shows "void-fraction" line in Figure 7 (approximately 1.8–0.74i)…". And in their section 9.2, second paragraph, "It should be assumed that most LAC lies on the void-fraction line in Figure 7 and Table 5. Our best guess is that the high values in the table are most promising, but Figure 9 shows that the values are not distinguishable in terms of absorptive properties." Since Bond and Bergstrom (2006) suggests, most LAC lies on the void-fraction line in Figure 7, we decided to use the 1.8-0.74i, which approximates the "void-fraction line" in Figure 7 as indicated in the caption of Figure 9. Furthermore, Bond and Bergstorm (2006) recommend "the high values" in their Table 5, not the highest value, 1.95+i0.79.

7. 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.

On page 2802, lines 16-18, we described "Climatological aerosol size distributions are used for fossil fuel and biofuel aerosols (Radke et al., 1988) and for biomass burning aerosols (Anderson et al., 1996), respectively". And it is mentioned earlier in the same paragraph that optical properties of BC, OC and sulfate are calculated as core-shell mixtures for fossil-fuel, biofuel and biomass burning aerosols (Page 2802, lines 6-11). So it means that fossil-fuel and biofuel BC is distributed following Radke et al., 1988; and biomass burning generated BC follows size distribution from Anderson et al., 1996. In optical

calculations for global models, aerosols are assumed aged and mixed together, so their mass concentrations are distributed to source-dependent climatological size distributions (fixed in the model), rather than individually fresh-emitted sizes.

We revise the text on page 2802, lines 16-18, as:

"Climatological aerosol size distributions are used for fossil fuel sulfate/BC/OC and biofuel BC/OC aerosols (Radke et al., 1988), with a mean diameter of 100nm and standard deviation of 1.9, and for biomass burning BC/OC aerosols (Anderson et al., 1996), with a mean diameter of 160nm and standard deviation of 1.65, following Liu et al. (2007) and Penner et al. (2001)". And also on page 2802, discussions related to the Figure2:

"Since source-based aerosol size distributions are prescribed and do not change in the IMPACT model, decrease of BC mass fraction in aerosol mixture means thicker coatings on smaller BC cores. The simulated enhancement in absorption as a function of the decreased BC mass fraction in Figure 2 reflects not only the coating effect from increasing shell substances but also from the size reduction of BC particles as core. It may not be used to directly compare with the lensing effect for coating on BC particles with the constant sizes."

8. 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.

In Bond and Bergstrom (2006), Aerosol Science and Technology, 40:27-67, section 8.3, second paragraph, it states "First, a lower bound of about 5 m2/g for submicron MAC particles is observed, The particle collapse observed by Schnaiter et al. (2003), even after just two hours, reduced absorption by just over 20%, corresponding to a change from our fresh value of 7.5 m2/g to about 5.8 m2/g. Clarke et al. (2004) reported values of 6–8 m2/g by normalizing absorption to the non-volatile fraction of the aerosol."

And they also state that "We suggest that the lowest values of 5 m2/g are collapsed but uncoated. They may also represent large agglomerates formed in the thick plumes of open biomass burning, as suggested by Liousse et al. (1993)", although" These low values are less commonly observed than other values, and we suggest that the uncoated aerosol does not remain in that state for very long." We think that the above statements justifies the sentence on line 29, Page 2802 "Bond and Bergstrom (2006) and Bond et al. (2006) suggested that lower bounds of absorption are 5m2 g−1 for observed submicron BC particles and 7.5m2 g−1 for freshly emitted aggregates".

9. 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.

In addition to our response to the comment #8, we also would like to clarify that in our model, BC is always coated with OC (we assume that OC/BC are mixed once emitted, since this global model does not simulate the aging process), as shown in Figure 7, in which there are no points near OC/BC = 0. Therefore, in global simulations there are no occurrences of uncoated BC particles with low absorption, for the reviewer's concern.

10. 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.

Please refer to our response to the comment #1. Our study focuses on the impact due to direct absorption of brown carbon, because the enhancement in absorption due to the lensing effect has been treated in our base model simulation (as in most of the current global models) that assumes internal mixtures of BC and non-absorbing OC.

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

We have plotted satellite and model results on the same scale to make them comparable. It would be better than the difference plot, because there are many missing points in the satellite data due to no retrievals over high-reflected surface (noted in the caption) over which model has values.

12. 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.

Revised as: "The global model calculations of AOD_NON capture approximately the observed aerosol hot spots identified by the local maximum in the MODIS AOD"

13. 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.

We did not include/discuss any AERONET AOD data products explicitly. In the calculation of the AAOD and AAE, the AOD and SSA data that we used are from the same AERONET V2 Inversion productions Level-2 daily files, as clearly described on page 2804, lines 2-5. According to the "AERONET's Version 2.0 quality assurance criteria" on the AERONET website, it does not indicate "AERONET can only extract SSA when the AOD > 0.2", but suggest that the Level 2 Inversion product uses AOD (440nm) > =0.4 as criteria in the inference of single scattering albedo, and refractive indices.

A sentence is now added on line 6, page 2804: "Note that the Level-2 SSA retrievals may be biased for large aerosol optical depth based on their quality assurance criteria (http://aeronet.gsfc.nasa.gov/new_web/data.html)"

14. 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.

The calculated correlation coefficient of 0.51 is the Pearson's correlation coefficient. The derived linear fitting functions are y = 0.71x + 0.3 for strongly abs BrC, y = 0.58x + 0.4 for moderately abs BrC, and y = 0.57x +0.42 for non-absorbing OC. The fitted lines are added to the Figure 4 shown below:

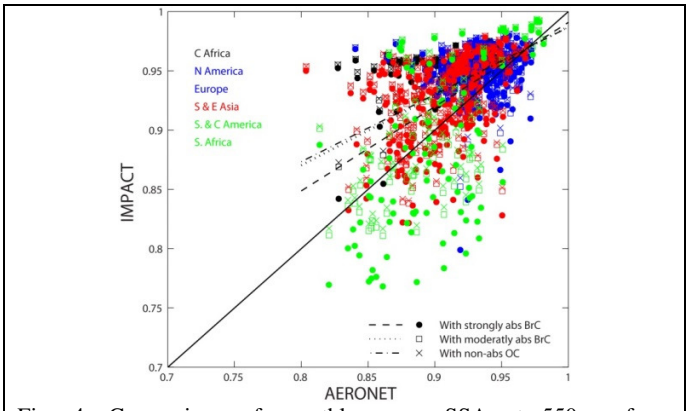

Fig. 4. Comparison of monthly mean SSA at 550nm from AERONET (1992–2012) and three model simulations (NON, MOD, and STR). The solid line indicates the 1 : 1 ratio. Data points in central Africa (C. Africa), Europe and North America, South Asia and East Asia (S and E Asia) and regions dominated by biomass burning (South and Central America (S and C America) and southern Africa (S. Africa)) are colored in black, blue, red, and green, respectively. The linear fitting functions are y = 0.71x + 0.3 for the STR BrC (dashed line; solid circles), y = 0.58x + 0.4 for the MOD BrC (dotted line; open squares), and y = 0.57x +0.42 for the NON absorbing OC (dash-dotted line; cross symbols).

Because the differences in SSA at 550nm between different model runs (with the STR and MOD BrC, and NON absorbing OC) are really small at about 1% to 2% of the absolute SSA values (line 20, page 2804), we have focused on the AAOD changes at 550nm and at smaller wavelengths (section 3.2, and Table 3). We include a new table (Table 3) now showing the calculated global and regional average SSAs at 550nm from different model simulations compared with the AERONET data:

SSA at 550nm	N America & Europe	S & E Asia	C Africa	S & C America & S Africa	Global average
Data points	228	233	224	92	842[a]
AERONET	0.933±0.021	0.912±0.03	0.928±0.024	0.899±0.041	0.917±0.030
Non-abs OC	0.953±0.017	0.936±0.031	0.957±0.011	0.898±0.054	0.940±0.033
Moderately abs BrC	0.952±0.017	0.936±0.032	0.957±0.011	0.895±0.056	0.939±0.034
Strongly abs BrC	0.949±0.021	0.932±0.035	0.956±0.014	0.871±0.070	0.932±0.042

[a] *In order to compare with the model results, a total of 1061AERONET data points are averaged into 842 data points on 2x2.5 degrees model grid cells*

For reviewer's interest, also shown below are the median, standard deviation and maximum of bias (absolute differences) and error (differences) in SSA between model results and AERONET observations for different regions. It is consistent with our statement in the manuscript that "The mean percentage error and bias between the modeled SSA_NON and observed SSA values are small, at about −2% and 3 %, respectively".

	N America & Europe	S & E Asia	C Africa	S & C America & S Africa
	Bias: abs[SSA(model) – SSA(obs)]			

	Median	Std	Maximum	Median	Std	Maximum	Median	Std	Maximum	Median	Std	Maximum
Non-abs OC	0.021	0.019	0.129	0.026	0.025	0.150	0.031	0.021	0.128	0.028	0.026	0.1
Moderately abs BrC	0.021	0.019	0.129	0.025	0.024	0.150	0.031	0.021	0.127	0.029	0.027	0.1
Strongly abs BrC	0.019	0.021	0.128	0.026	0.024	0.146	0.029	0.021	0.125	0.04	0.034	0.127
	Error: SSA(model) – SSA(obs)											
	Median	Std	Max/Min	Median	Std	Max/Min	Median	Std	Max/Min	Median	Std	Max/Min
Non-abs OC	0.017	0.025	0.129/-0.059	0.022	0.032	0.15/-0.099	0.031	0.021	0.128/-0.006	0.006	0.042	0.1/-0.078
Moderately abs BrC	0.016	0.026	0.129/-0.067	0.021	0.033	0.15/-0.101	0.031	0.021	0.127/-0.007	0.004	0.044	0.1/-0.084
Strongly abs BrC	0.013	0.028	0.128/-0.12	0.017	0.035	0.146/-0.123	0.029	0.021	0.125/-0.012	-0.017	0.056	0.099/-0.127

The calculated Pearson's correlation coefficients are similar between different model simulations, as shown below for different regions:

Pearson's correlation coefficient with AERONET data	N America & Europe	S & E Asia	C Africa	S & C America & S Africa
Non-abs OC	0.12	0.44	0.45	0.63
Moderately abs BrC	0.12	0.44	0.45	0.63
Strongly abs BrC	0.08	0.44	0.46	0.61

The sentences on P2804, L7 are revised as

"The monthly predictions of SSA_NON correlate with the AERONET observations (a total of 1,061 data points) with a calculated Pearson's correlation coefficient of 0.51. The mean percentage error and bias between the modeled SSA_NON and observed SSA values are small, at about -2% and 3%, respectively."

And also on P2804, the first paragraph in section 3.2 is revised as:

"The consideration of BrC increases total aerosol absorption. In Figure 4 and Table 3, the two model simulations with BrC, SSA_STR (solid circles) and SSA_MOD (open squares), predict lower SSA values at 550 nm than the SSA_NON (cross symbols). Averaged over all the AERONET sites, the SSA_STR (0.93) agrees slightly better with the observations (0.92) than the SSA_MOD (0.94) and SSA_NON (0.94). In S & C America and S. Africa, the best agreement is for the non-absorbing OC case (SSA_NON = 0.9), while the SSA_STR (0.87) is lower than the AERONET data (0.9) by 3%. In addition to refractive index/absorption cross section used for BrC in the STR case, the overestimation of SSA in biomass region might also be because the assumed fraction of BrC in total OC (92% based on a solid fuel analysis) is too high for these sources. Nevertheless, the changes in SSA at 550nm between the NON, MOD, and STR cases are at about 1-2%."

15. 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.

Please refer to the response to the comment #14.

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

The citation is deleted.

17. 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.

The fractional increases due to BrC can be estimated from comparing the two panels in the Figure 5, since they are plotted on the same color scale. We have included discussions about the contribution by BrC in percentage to AAOD, such as lines 25-26, on page 2804, and lines 3-4, on page 2805. In addition, we include the changes of AAOD from the MOD BrC in Figure5:

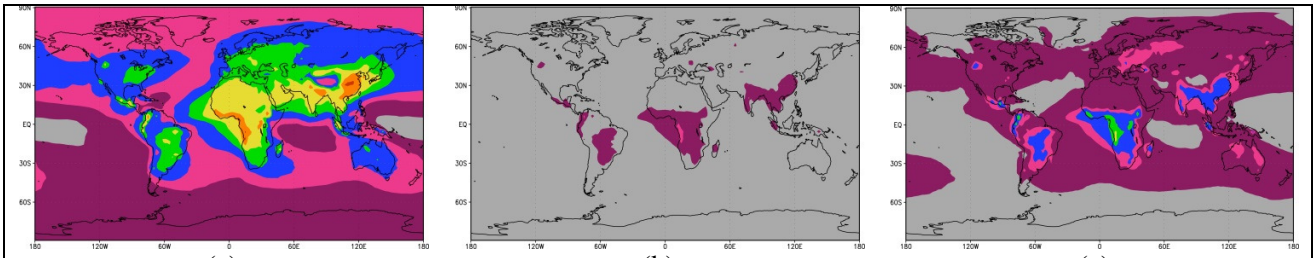

Figure 5. (a) Annual mean absorption aerosol optical depth (AAOD × 100) calculated with the NON absorbing OC; and increases in AAOD (× 100) due to the inclusion of the (b) STR and (c) MOD BrC

18. 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.

Please refer to the response to the comment #7 regarding the size distributions used in the model simulations. Same sizes distributions are used in all three model simulations. The only difference between those model simulations is the imaginary part of the refractive index. We have also explained that our control case includes BC and non-absorbing OC, so the increase in AAE values from the two sensitivity model simulations can be solely attributed to the inclusion of direct absorption of BrC.

In spite of the uncertainty in aerosol size distribution and sphericity of particles, there are better agreement in AAE between the strongly or moderately absorbing BrC and AERONET data than between the control run and AERONET data. And the better agreement in AAE at least would lead to better calculations of radiative forcing, which is the main point of this paper.

19. 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.

The following sentence is added on line 20, page 2805, after "…low biased": "The width of the AAE frequency distribution implies the variability in aerosol size and composition. For non-absorbing OC case, it indicates the variations in the coating thickness on BC."

20. 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.

Revised as, "Although no BrC is explicitly assumed to be associated with fossil fuel combustion, the AAE_STR distribution when BrC from bio-fuel combustion and biomass burning is included is in very good agreement with the AERONET data over Europe and North America."

21. 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.

The following is added at the beginning of the section 3 Global results:

"In order to investigate the impact of BrC absorption on aerosol forcing, we conducted three global calculations of aerosol optical properties and radiative transfer, with non-absorbing OC, moderately absorbing BrC, and strongly absorbing BrC. Model results from these three simulations are referred hereafter as "NON", "MOD", and "STR", respectively. The control case is the NON case, similar to most of the current global aerosol models. The two sensitivity calculations (MOD and STR) are used to estimate the low and high limits of the resulting aerosol forcing by converting some non-absorbing OC to absorbing BrC."

22. 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.

Although the AAE_STR is overpredicted in biomass burning regions, average over all sites, it agrees the best with AERONET spectral retrievals. We think discussions of both global and regional mean values are meaningful, because the global values would give rise to the importance of BrC absorption in global aerosol forcing and motivate further research on BrC. Being aware of the uncertainty of using one configuration in global simulations, we did simulations using strongly abs BrC to represent the upper limit for the BrC absorption, and using moderately abs BrC to represent the lower limit for the BrC absorption. In addition, the calculated AAE_MOD for the moderately absorbing BrC in Figure 6b agrees better with the AERONET AAE than the AAE_NON but is still low-biased, implying there is stronger BrC absorption than the MOD case from biomass burning sources.

The sentence comparing with results from Kirchstetter and Thatcher (2012) is deleted.

23. 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.

We have addressed the issue about the AAE of BC =0.53 and aerosol size distributions in our responses to comments #1 and #7. Please refer to those responses.

The authors state: "Carbonaceous aerosols, BC, and OC were considered in one size bin only (radius<0.63 m)." 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.

Please see our response to the comment #7, regarding the BC and OC size distributions used.

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.

Please refer to our responses to the comment #1 regarding the AAE = 0.53. and comment #7 regarding the size distributions of BC and OC. Again, AAE = 0.53 is not calculated from the Mie theory for individual BC particles. It is asymptotic for aerosol mixtures with size distributions of fossil fuel and biomass burning aerosols while the OC content is zero.

24. 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.

Please see our response to the comment #1: this is to compare with Bahadur et al. (2012).

25. 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.

This is addressed in our response to the comment #21.

26. 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.

Our base model simulation has included the enhancement due to non-absorbing OC. Please refer to our response to the comment #1.

27. 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).

Figure 7 is revised as below:

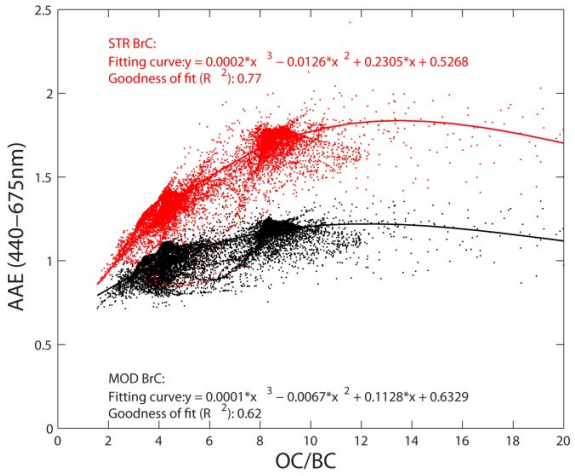

Figure 7. The simulated wavelength dependence of AAE (440-675 nm) as a function of the ratio of OC/BC column burden, for the STR and MOD BrC. The fitted curves are valid for ratios of OC to BC between 1.5 and 20

As explained in the response to the comment #1, the intercept AAE value of 0.53 for strongly absorbing BrC represents for an extreme case when the OC content is zero for a population of biomass burning or fossil fuel aerosols with prescribed climatological size distributions. It is not used as AAE for pure BC. We don't account for BC and OC optical properties as individual species, but calculate optical properties for BC and OC (and sulfate for fossil fuel) mixtures. As Figure 7 shows, the smallest ratio of OC to BC is 1.5, so OC/BC=0 or AAE=0.53 never occurs. Because the AAE value of 0.53 causes lots of confusion from the reviewer, we deleted those lines without affecting our main point. The revised discussions are:

"Figure 7 demonstrate the simulated AAE_STR for strongly absorbing BrC and AAE_MOD for moderately absorbing BrC between 440 nm and 675 nm as a function of OC:BC burden. The OC/BC ratios in model results are concentrated between 2 to 6 and 8 to10, corresponding to fuel combustion and biomass burning source regions, respectively. The minimum value of the AAE_STR in global calculations is about 0.825 at the OC/BC ratio of 1.5; as the ratio of OC/BC increases, the calculated AAE_STR increases up to values around 1.5 to 2, while the maximum AAE_MOD is about 1.2. Because the absorption by BrC is close to zero near 675 nm, the fitting curves for the strongly or moderately

absorbing BrC simulations (solid lines) could be useful to approximate the enhancement of AAOD due to BrC absorption at the smaller wavelengths for models that consider BC and non-absorbing OC only, based on the simulated OC/BC ratios and aerosol absorption at 675 nm."

28. 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).

Being aware of the uncertainty in AAE, we did the simulations of strongly abs BrC to represent the upper limit for the BrC absorption, and the simulations of moderately abs BrC to represent the lower limit for the BrC absorption. We have revised the manuscript to emphasize more on the range of estimated BrC absorption, rather than just the strongly absorbing case. We have added "_NON", "_MOD", and "_STR" to distinguish simulations with non-absorbing OC, moderately, and strongly absorbing BrC, respectively.

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

We compared with those AeroCom forcing numbers in Table 4, because we have matched the same emissions used by those AeroCom studies.