We are thankful to Reviewer #2 for his/her comments.

Reply to Reviewer 2:
In the following, the comments from Reviewer 2 are in italics followed by our responses.

1. *This paper is a good study, but I believe Fig 4 demonstrates that the strongly absorbing BrC is too absorbing in comparison with observations. Hence, the paper needs to de-emphasize this case. There are other smaller points and points of clarification that should also be corrected (see below). i.e., the paper does not compare their derived refractive indices with those in the literature.*

   Thanks for the suggestions. We have revised the manuscript to emphasize more on the range of estimated BrC absorption, rather than just the strongly absorbing case. We have also included the comparison of refractive index in literature in the Table 2. Please see below for the one-by-one responses to the specific comments below.

2. *Page 2797: Lines 10, 11 "Although absorbing aerosols amplify the atmospheric forcing due to GHGs, their dimming effect compensates for the GHG warming at the surface." As shown in Penner et al. 2003 (JGR), whether an absorbing aerosol warms or cools depends on it's vertical placement in the atmosphere. If close to the surface, there is a negative surface forcing, but the aerosols warm the surface temperature.*

   Revised as "Although absorbing aerosols amplify the atmospheric forcing due to GHGs, their dimming effect may compensate for or enhance the GHG warming at the surface depending on the vertical placement in the atmosphere (Penner et al., 2003)"

3. *Page 2799 line 3: carbonaceous aerosols above 0.63 um radius are not considered? How much increase in forcing would there be if these were included?*

   No we did include emissions and mass concentrations of all the carbonaceous aerosol particles in one bin. The size cut of aerosols in the IMPACT model is based on consideration of the mass size distribution and thermodynamics of these aerosols as well as the CCN activation spectrum under typical supersaturations (Liu et al., 2007). Since this version of IMPACT model does not treat aerosol dynamics, aerosol size distributions are assumed to follow those derived from measurements. Fossil fuel carbonaceous aerosols have a mean diameter of 100nm and standard deviation of 1.9, and biomass burning BC/OC aerosols have a mean diameter of 160nm and standard deviation of 1.65, following Liu et al. (2007) and Penner et al. (2001). Based on these size distributions, the surface area of carbonaceous aerosol particles is dominated by the submicron particles. So the impact on forcing estimate is negligible, because the surface-weighted effective radius calculated in the sub-micron size bin is representative of all carbonaceous particles in the optical calculations.

   To avoid the confusion, this sentence is now revised as:
   "Total carbonaceous aerosols (OM and BC) are represented in a single submicron size bin."

4. *Page 2800: line 8-9: "In the IMPACT model, OC is converted to particulate organic matter at a ratio of 1 : 1.4 to account for secondary aerosol formation from volatile organic compounds." This sentence made me go back and find your definition of OC. On page 2798, line 7, it appears that OC is defined as "absorbing organic carbon". So then a ratio of 1:1.4 would be the mass ratio between total organic matter (OM) and absorbing organic matter? This is unfortunate. Usually OC only refers to the carbon within organic matter. Often a ratio of 1 to 1.4 is the ratio*

between OC and OM. And this ratio, while partly explained by secondary aerosol formation, is not entirely due to this formation, since primary emissions of OM can be emitted with a ratio of OM to the carbon-only mass that is larger than 1. Please clarify what is meant here and on page 2798.

The ratio of 1:1.4 is still the mass ratio between OC (both non-absorbing and absorbing) and total OM. And only 66% of total OM and 92% of OC is considered as absorbing, as described on line 11, page 2800. The sentence on line 7, page 2798, does not mean to define all OC as "absorbing organic carbon". The breakdown of total OM (1.4) for biomass burning and bio-fuel sources is: 0.92 BrC, 0.08 non-absorbing OC, and 0.4 for all non-carbon mass and SOA. For fossil fuel OM, the breakdown is 1 non-absorbing OC and 0.4 for all-carbon mass and SOA.

The sentence on line 7, page 2798, is revised as,
"…for the existence of some organic carbon as light-absorbing. This fraction of absorbing organic carbon, known as brown carbon, …"

5. *Line 13: state whether you refer to the primary emissions of fossil fuel here or are including secondary produced fossil fuel aerosol (I assume the former).*

Yes, it is for primary emissions of fossil fuel. Revised as,
"no BrC is considered in the primary emissions of fossil fuel-produced organic matter…"

6. *Line 15: is any BrC considered in the natural OM? Are these externally mixed from other aerosols*?

BrC is not considered in the natural OM. Yes the natural-emitted OM is externally mixed with other aerosols. We added the external mixing of natural-emitted OM on line 18, page 2802, as "Dust, sea salt, and naturally emitted OM are assumed to be mixed externally."

7. *Line 27: what is the source for the natural organic matter emissions? Are the organic emissions Tg/yr or Tg C/yr?*

The source of the natural organic matter emissions is now added on line 26, page 2800, as,
"BC and OC from open biomass burnings and naturally emitted OC are based on the AeroCom emissions (Dentener et al., 2006)".

The organic emissions are in Tg per year, not Tg C/year.

8. *Page 2801: line 19: Kirchstetter et al. also derived the refractive index as did Chen and Bond. Please compare their values in Table 2. (differences may be due to different size distributions and/or densities).*

The refractive index from the two reference papers are now included in the Table 2 as shown below:

Table 2. The imaginary refractive index, absorption cross section, and absorption Angstrom exponent (AAE) calculated for brown carbon (BrC) at various wavelengths, compared with those from Chen and Bond (2010) and Kirchstetter et al. (2004).

	Imaginary refractive index at indicated wavelength				Absorption cross section at indicated wavelength (m^2 g^{-1})				AAE
Wavelength λ	350	450	550	650	350	450	550	650	400-700

(nm)									
Moderately absorbing BrC	0.075	0.02	0.003	0.0003	2.25	0.63	0.08	0.006	11.1
Strongly absorbing BrC	0.168	0.063	0.03	0.005	3.4	1.6	0.7	0.1	6.6
Chen and Bond (2010)[1]	0.1	0.02	0.006	1.e-4	2.25	0.4	0.08	0.001	7.5
Kirchstetter et al. (2004)	0.168	0.063	0.03	0.005	5.0	1.5	0.6	0.1	-

[1] Based on the absorption cross section for methanol-soluble OAK_L_360; refractive index is derived based on $\rho\lambda\sigma/4\pi$, and ρ is the density of BrC, 1.65 g cm^{-3}

The following sentence is added on line 3, page 2802:
"The differences in derived refractive indices from Kirchstetter et al. (2004) and Chen and Bond (2010) are due to different size distributions and/or densities used in the Mie calculations"

9. *Page 2802: Line 11: how is absorbing BrC and non-absorbing OC handled? Are these also a core-shell treatment, or do you do a mixing rule for the refractive indices?*

Absorbing BrC and non-absorbing OC are internally mixed as shell substances coating on BC. Their refractive indices are volume-averaged.

10. *Figure 4: this is an interesting figure and deserves more discussion than it is given on page 2804. In particular, are you able to say anything about whether the high absorbing OC is too absorbing? Certainly in S. and C. America and S. Africa, the slope through the green full dots appears to indicate that there is too much absorption.This is the region where biomass burning is most prevalent, and thus where most of your absorbing BrC is. This might indicate that the higher estimate leads to a forcing by BrC that is too high. I cannot tell whether the slope through the open green symbols might be better. The red and blue dots (Europe, N. America) appear to have too little absorption (as does the dust regions, which you discuss).*

For all AERONET sites shown in Figure 4, the linear fitting functions between model results and data 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:

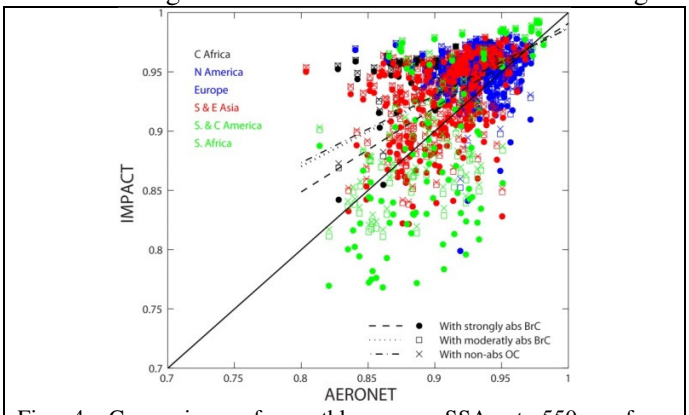

Fig. 4. Comparison of monthly mean SSA at 550nm from AERONET (1992–2012) and three model simulations (NON, MOD, and STR). The solid line indicates the 1 : 1 ratio. Data points in central Africa (C. Africa), Europe and North America, South Asia and East Asia (S and E Asia) and regions dominated by biomass 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).

We also include a new table (Table 3) 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*

Both the linear fitting in Figure 4 and Table 3 suggest that over all AERONET sites, the average SSA at 550nm for strongly absorbing BrC agrees the best with the observations. In S & C America and S. Africa, the best agreement is for non-abs OC, and strongly absorbing BrC overpredicts by 3% than the AERONET data. In addition to refractive index/absorption cross section of BrC, the overestimation of SSA in biomass region could also be because the assumed fractional BrC in total OC (92% based on a solid fuel analysis) might be too high (Lines 1-4, page 2806). So we cannot say that "*the high absorbing OC is too absorbing*", due to lack of constraints on the mass loading of BrC. Being aware of the uncertainty of using one configuration in global simulations, we have done simulations with 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.

11. *Figure 5: It would seem that the strongly absorbing BrC should be replaced by the moderate BrC, since the former is an overestimate (see above).*

Both moderately and strongly absorbing BrC are now shown in Figure 5:

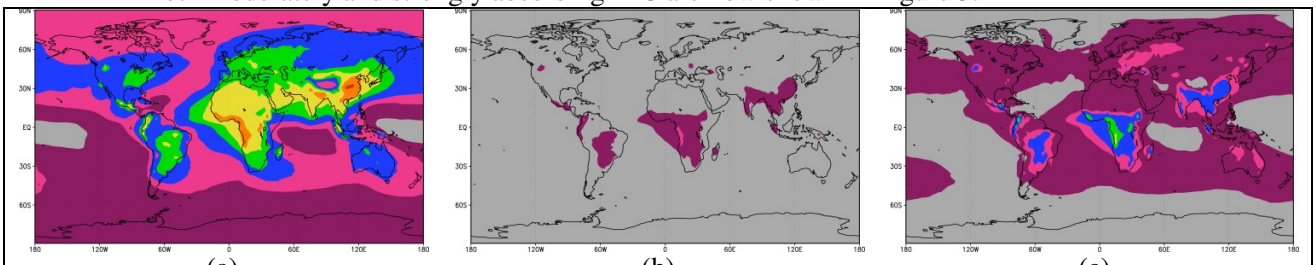

Figure 5. (a) Annual mean absorption aerosol optical depth (AAOD × 100) calculated with the NON absorbing OC; and increases in AAOD (× 100) due to the inclusion of the (b) STR and (c) MOD BrC

12. *Fig 6: the strongly absorbing case appears to overestimate AAE in biomass burning cases also.*

The strongly absorbing case overestimates AAE in biomass burning region, and the best case is moderately absorbing BrC. We have thus included moderately absorbing BrC in both Figure 5 and Figure 7.

13. *Fig 7: consider also presenting the moderate BrC for this figure.*

Both moderately and strongly absorbing BrC are shown in Figure 7:

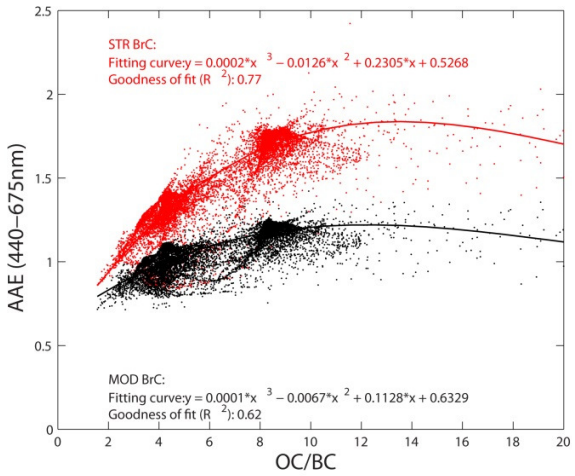

Figure 7. The simulated wavelength dependence of AAE (440-675 nm) as a function of the ratio of OC/BC column burden, for the STR and MOD BrC. The fitted curves are valid for ratios of OC to BC between 1.5 and 20

14. *Page 2807, lin 13: Is the forcing really with and without each aerosol type? Normally one calculates the TOA flux with all aerosols minus the TOA flux with all aerosols except the aerosol type of interest. Please correct this if done incorrectly.*

Revised as, “Aerosol radiative forcing is estimated as the differences in the calculated radiative fluxes with all aerosols and with all aerosols except the aerosol type of interest.”