We thank both reviewers for there time and comments. We have made efforts to improve the manuscript accordingly, please find response for corresponding points below.

Reviewer #1

No comments.

Reviewer #2

Major Comments:

This manuscript presents a global estimate of direct radiative forcing by black carbon (BC) and brown carbon (BrC) using a chemical transport model (GEOS-Chem) coupled with a radiative transport model. The GEOS-Chem model is improved with an aging parameterization of BC and enhanced BC absorption accounting for the coating effect and inclusion of BrC absorption. Sensitivity simulations are conducted to estimate uncertainties in the estimated radiative forcing due to uncertainties associated with size, optical properties, emissions, and vertical placement of BC. The content is interesting to the community and the manuscript is overall well written. However, I have concerns about some of the conclusions drawn from the comparison of this study with other models and observationally-based study (Bond et al. 2013). Specifically, I have the following comments:

(1) It is not that surprising that GEOS-Chem assuming externally mixed BC results in smaller forcing than models that assume internally mixing. For example, CAM5.1 (Myhre et al., 2013) predicts a column burden of FF and BF BC (0.07 mg/m2), which is lower than total BC (FF+BF+BB) in this study from either GC-RT baseline (0.11 mg/m2) or "best" (0.10 mg/m2) models, but still gives a larger RF (+0.2 W/m2) than that of this study (+0.07_0.08 W/m2). Aging of BC is considered in CAM and the BC lifetime is also about 4 days (Liu et al., GMD, 2012). The difference is that CAM5 assumes internally mixed BC. So it seems like external mixing is more likely the reason why GEOC-Chem has a low estimate of BC forcing compared to CAM5 and other bottom up models, instead of the aging treatment or shorter lifetime of BC as inferred here. Although a scaling factor is used to account for coating, it may still be underestimated. Discussions about mixing state between models and the impact on the estimated forcing compared to other factors such as aging should be added.

We agree with the author that the mixing state and other factors may influence the comparison with other models. However, the majority of this difference is addressed in our study by the inclusion of the absorption enhancement in our "best" estimate. This is a proxy for the lensing effect that is produced by internal mixtures of BC and scattering aerosols. As a result, the simulated AAOD in our "best" estimate (0.0014) is only slightly lower than the CAM5.1mean AAOD (0.0015). Thus the larger difference in DRF (+0.13 Wm^{-2} in our "best" vs. 0.20 Wm^{-2} in CAM5.1) is more likely to be from the attribution of BC/BrC in AAOD and the vertical profile of BC, than the mixing state. We have added a sentence to explicitly indicate that the absorption enhancement is included in the model to address the effect of internal mixing in Section 2.3, page 17537, line 25:
"It is not possible to model this effect explicitly in the externally-mixed bulk aerosol scheme of GEOS-Chem, therefore we use absorption enhancement (AE = MAE with coating / MAE without coating) to describe this influence from internal mixing. "

(2) In the comparison with BC forcing from Bond et al. (2013): after scaling the AAOD to match the AERONET retrievals, why is the obtained AAOD (0.002) almost 3 times smaller than that (0.006) given by Bond et al. 2013? Would it generate a larger BC forcing, close to Bond et al. (2013) if the AAOD is scaled up to the 0.006 levels?

The scaled AAOD is associated with both the AAOD originally simulated by the model and the scale factor, which describes the difference between model and AERONET. It should be noted that the AERONET sites only cover a small fraction of the earth and are usually located at high AAOD regions. As discussed in section 5, the bias of our model at these AERONET sites is smaller than the AeroCom models which are used in Bond et al., 2013. As a result, we have smaller scale factors. However, in other regions that AERONET does not cover, especially the broad ocean areas, our simulated AAOD are much smaller than AeroCom models due to the shorter lifetime. As there are limited ocean sites, our low AAOD values over these regions are not scaled up over the oceans, unlike Bond et al. (this is discussed in Sections 6.1 and 6.2), thus resulting in a lower AAOD.
If the AAOD were scaled up to 0.006, it would generate a large BC forcing, but still smaller than Bond et al., 2013 (0.5 vs. 0.7). This is because the AAOD alone does not determine the DRF, the vertical distribution of BC is also important. This is discussed in Section 6.1.

(3) Also, Bond et al., 2013 scaled the BC AAOD at 550nm only to match the AERONET retrieved values. So most of the BrC absorption occurring in the UV bands is not attributed to BC forcing. According to this study, only about the 25% enhancement in absorption at 550nm due to BrC would contribute to the overestimation of BC forcing. But the differences in BC AAOD and forcing ("best" and scaled GC-RT) between this study and Bond et al. 2013 are much more significant (as shown in Figure 11). The authors acknowledge the +100% uncertainty in AERONET AAOD retrievals, which is in fact larger than any of the other factors listed in Table 3, but still "suggest that the DRF of BC has previously been overestimated due to the overestimation of BC lifetime and the incorrect attribution of BrC absorption to BC"? Together with (1) above, I cannot agree with this conclusion.

Yes, we agree with the reviewer, Bond et al., 2013 scaled the BC AAOD at 550nm only to match the AERONET values, and we estimate that 25% of the AAOD at this wavelength is from BrC. However, this underestimates the full-spectrum absorption of BrC which contributes to the forcing estimate (~35%), but the absorption from BrC is not the main cause of the difference between Bond et al. and our study. Rather the lifetime and vertical profile of BC are more important factors in this comparison, as discussed in Section 6.1.
We acknowledge the +100% uncertainty in AERONET AAOD retrievals, which is large but may be a relatively small source of uncertainty in the overall analysis. The overall difference between Bond et al., 2013 and our estimate is about 300%. This demonstrates that large differences in DRE/DRF may result from factors independent of AAOD, including the BC lifetime (which determines the BC vertical profile) and the BrC absorption.

(4) Estimate of BrC forcing in this study is based on a simple treatment of BrC optics. Since BrC is often co-emitted with BC from BB and BF and other OA, how would it change the estimated BrC and BC forcing if BrC is coated on BC? The BrC absorption coefficient is highly variable depending on sources, burning conditions, etc, as shown in Figure 1. It would be useful to compare BrC global burdens, optical properties and forcing calculated in this study with previous studies such as Arola et al. (2011), Feng et al. (2013) or Lin et al. (2014), as the latter adopt different BrC fraction in OA and absorption coefficients.

Since the effect of BrC coating BrC on BC is not well understood, we treat it the same as BC coated by other OA. In our "most absorbing" simulation, we use an absorption enhancement factor of 2 for these coatings. To our knowledge, the absorption enhancement observed in field studies (where the coating material can be BrC or non-absorbing OA) are always smaller than 2. So our discussion of the "most absorbing" simulation incorporates the uncertainty associated with the coating of BC by BrC

Given the uncertainties in BrC optics, we do not think that the effect of BrC coating is a dominant source of uncertainty in our simulation of BrC forcing. Based on our simple calculations, we estimate that BrC in coating material is less than 5% of the total BrC mass, even we assume a thick coating condition observed in field studies. This effect is therefore likely negligible.

We thank the reviewer for their suggestion to more thoroughly compare our BrC simulation to previous studies. These studies were introduced in the introduction and model description, and we have added the following dissection to Section 6.1, page 17552, line 9:

"We can compare of BrC simulation and resulting forcing to previous studies. Feng et al., (2013) base their BrC optical properties on Chen and Bond (2010) (smaller than the pink triangles in Fig.1a, since they choose a different burning condition) and Kirchstetter et al. (2004) (light green circles in Fig.1a) and treat 66% of the POA from biofuel and biomass burning as Br-POA. They do not include any secondary sources of BrC. The biomass burning emissions employed in this study , are about 40% higher than our biomass burning source based on the 12-years GFED3 average. This results in a 170% larger global BrC source (30.3Tgyr^{-1}), but a similar global BrC DRF (+0.04 to +0.11 Wm^{-2}). Notice that our DRF value is scaled to meet AERONET data. In contrast, Lin et al. (2014) estimate a much larger DRF for BrC (+0.22 to +0.57 Wm^{-2}). Their optical properties for primary BrC are also based on Chen and Bond (2010) and Kirchstetter et al. (2004). However they also treat all SOA as BrC and apply these same absorption properties to the Br-SOA, resulting in an unrealistically high estimate of BrC DRF."

Other comments:

1. Page 17529, lines 24-26: although IPCC-AR5 models and observationally-based methods such as Bond et al. (2013) all give higher BC forcing than the estimates in this study, they are for different reasons. It's confusing here as it seems to attribute same reasons to the overestimation.

As our response to major comment (1) and (3) above, the largest difference between all of these studies and our estimate is due to AAOD-independent processes: the BC lifetime (vertical profile) and BrC absorption.

2. Page 17530, lines 2-6: add references

We have added the references.

3. Line 5: why emissivity?

The absorption of BC will change the shortwave absorption of BC-including clouds, thus change the cloud emissivity.

4. Line 28 : replace "assumed (5-10 days)" with "simulated (5-10 days)"

This is replaced.

5. Page 17531, line 3: "Model estimates" of ?

This is changed to "Model estimates of BC DRF".

6. Line 8: add "asymmetry factor" after AAOD

This is added.

7. Line 22: Kahn et al. (2010) is missing from the references. Are these two papers discussing issues in AOD or AAOD satellite retrievals ?

It should be Kahnert et al. (2010). Yes, Li et al (2009) is a study on the uncertainties of satellite retrievals. Kahnert et al. (2010) is a model-observation comparison study, but also includes relevant discussion of observational uncertainties.

8. Page 17532, lines 10-11: "all aerosols" in Feng et al. (2013) exclude dust and naturally-emitted aerosols.

This is added.

9. Line 19: "coupled with"

This is added.

10. Page 17534, line 10: is the forcing of individual species estimated as difference in flux between with single component and without aerosols, or difference in flux between with all components and with all but the single component?

It is the difference in flux between with all components and with all but the single component.

11. Page 17535, line 14: "fossil fuel"

This is changed.

12. Lines 27-28: is it justifiable to use coagulation e-folding time derived from the Arctic study to urban conditions? Obviously, this constant b is one or two orders of magnitude smaller than the condensation term typically in urban area, and can be ignored.

Although the b value is applied to the Actic in the study of Liu et al. (2010), the b value itself is not derived specifically for these conditions. Yes, b is not important and can be ignored in urban areas.

13. Page 17536, line 3: "which find"

Both of the two references find.

14. Section 2.2: should the condensation rate in equation (2) also depend on the surface area of particles?

Yes, it should also depend on the surface area. However, the bulk aerosol mechanism in GEOS-Chem does not simulate a changing size distribution so the rate we use can be seen as an average value for different particles.

15. Page 17537, line 5: add reference to the Mie code

We have added the reference.

16. Section 2.3: are these MEE and MAE for dry aerosols? Do they change as relative humidity increases? The calculated MEE and MAE should be compared with those in AeroCom models, which is helpful to explain the differences in forcing.

The MEE and MAE are for dry aerosols, they will change as relative humidity increases (wet aerosols), which follows the hygroscopic growth from Martin et al. (2003), described in section 2.1.
Unfortunately, the MAE for BC are not diagnosed in AeroCom models (Myhre et al., 2013). Furthermore, mentioned in our response to major comment #1 and #3, the difference in forcing is largely explained by other factors.

17. Page 17538, line 8: disappear?

Sentence is correct as written.

18. Page 17540, lines 8-10: references for this assumption?

There is no specific reference for this, the assumption follows the logic given in the text. As summarized in page 17539, paragraph 2, Br-POA are generally from biomass burning/biofuel, most of which are water-insoluble, Br-SOA are more likely to be associated with aromatic carbonyls, which is water-soluble except at large sizes. Since the absorption of water-soluable BrC is really small and acetone/methanol-soluble BrC covers more than 90% of the total BrC, the absorption of acetone/methanol-soluble BrC is about the same as total BrC.

19. Page 17543, lines 13-14: does it mean the same SSA is used for high (>0.4) and low (<0.4) AOD? at what temporal resolution? Is SSA constant throughout the season or month? This paragraph is quite confusing.

We use the same SSA for all AOD condition in a given season.

20. Page 17548, line3: replace "this treatment produces" with "this procedure generates"

Changed.

21. Page 17550, line 29: need add reference for "AeroCom models do not consider absorption enhancement from BC coating". Internally mixing assumption implicitly treats the coating effect.

We agree, and this was unclear in our text. The AeroCom II experiments include some models with internal mixing, however, the Bond et al. study (which is being discussed here) uses AeroCom I models which do not include internal mixing or absorption enhancement from BC coating. We have clarified this in the text.

22. Page 17551, lines 24- 26: how is the scaling factor for BrC AAOD derived separately from that for BC AAOD?

We assume that the AAOD differences between model and AERONET are from both BC and BrC. The scale factors are the same for both.

23. Page 17552, lines 7: compare the BrC forcing with previous studies

Same as major comment #4, added in section 6.1, page 21, line 3-15.

24. Page 17555, line 16: lower than 4 days or about 4 days? It is 4.4 days in Table 2 for "best" GC-RT.

Although our simulations with a ~4 days lifetime improve the comparison with observations, BC concentrations are still overestimated in remote areas and at high altitude. Thus we suggest a lifetime shorter than 4 days would remedy this overestimation.

25. Line 21: 50% in AAOD at "550nm"?

At 440nm.

26. Table 2 caption: GC-RT (for year 2010);

Changed.

27. Figure 1: a. it is said in the text that the BrC RI is included up to 600nm, so extend the x-axis and plot the solid lines in both panels to 600nm; b. in the left panel, I couldn't find symbols (pink dots) for Chen and Bond, 2010

We have corrected the mislabeled symbols. The x-axis has been extended.

28. Figure 7: Why was the northern India excluded? there are many data points

We did not exclude any data points in the analysis. If the reviewer's question is why was northern India not shown as a specific region in Figure 8, the reason is the mixture of urban and biomass burning of emissions in this area. As mentioned in section 5, we selected regions which were dominated by either urban emission or biomass burning .

29. Figure 8: In the panels (d) and (e), the AAE calculated from the "best" model and "most absorbing" model clearly overpredits compared with AERONET data, even worse than the "baseline" model. It contradicts with the conclusion on page 17555, lines 20-21, "inclusion of brown carbon . . . almost eliminated the bias in AAE".

Have changed the conclusion to “inclusion of brown carbon . . . almost eliminated the bias in AAE in typical urban areas.”

30. Figure 11: could also compare BrC forcing with other estimates.

The goal of this figure is to compare the BC forcing with typical studies. BrC forcing is shown here to illustrate how it may contribute to differences between our estimate and previous BC estimates.

31. References: Bond et al. (2013) is missing

This is added.