Atmos. Chem. Phys. Discuss., 14, C6453–C6458, 2014
www.atmos-chem-phys-discuss.net/14/C6453/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Exploiting simultaneous observational constraints on mass and absorption to estimate the global direct radiative forcing of black carbon and brown carbon" *by* X. Wang et al.

Anonymous Referee #1

Received and published: 3 September 2014

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.

(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?

(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

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the incorrect attribution of BrC absorption to BC"? Together with (1) above, I cannot agree with this conclusion.

(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.

The improvement in representation of BC aging and BrC absorption in GEOS-Chem improves its estimate of BC forcing with better agreement with observations, but these model improvements do not seem to reconcile the large gap in BC forcing between different models and observational studies as suggested in abstract and conclusion.

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.

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

3. Line 5: why emissivity?

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

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

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

7. Line 22: Kahn et al. (2010) is missing from the references. Are these two papers

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discussing issues in AOD or AAOD satellite retrievals ?

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

9. Line 19: “coupled with”

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?

11. Page 17535, line 14: “fossil fuel”

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.

13. Page 17536, line 3: “which find”

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

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

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.

17. Page 17538, line 8: disappear?

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

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.

20. Page 17548, line3: replace “this treatment produces” with “this procedure gener-

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ates”

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.

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

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

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.

25. Line 21: 50% in AAOD at “550nm”?

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

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

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

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”.

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

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

References:

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 17527, 2014.