Responses to Referee Comments #1 and #2

We would like to thank both referees for their very helpful comments regarding our manuscript. Below we have included our responses. The referee comments are in black text while our responses are in blue.

Referee Comment #1

This paper estimates the effective absorbing optical property of “brown carbon” (BrC), which is a part of the bulk primary organic carbon (POC) emitted from biomass burning, by adjusting the spectral dependence of imaginary refractive indices of BrC and BrC/POC ratio in the GEOS-Chem simulation of UVAI to match the UVAI from OMI and reported AAE from literature. Then, the consequences of BrC absorption on tropospheric OH and aerosol direct radiative effects (DRE) are calculated with the GEOS-Chem model, which show significant reduction of OH and changes of DRE over major biomass burning regions/seasons.

1. The term “primarily non-absorbing” or “primarily scattering” for OC is somewhat misleading. The imaginary refractive indices (k) of OC from OPAC is 0.005 – 0.008 in the 300-550 nm wavelength range, which means OC is absorbing, at least weakly absorbing. In fact, the k value of OC increases with the wavelength from 350 to NIR (about 0.4 at 9000 nm), therefore OC should not be defined as non-absorbing aerosol even with the OPAC values. The problem seems mostly in the UV and shortwave VIS range where the absorption from OPAC is likely to be too weak and the AAE too low.

We now avoid the terms “primarily non-absorbing” or “primarily scattering”.

2. The relationship between k and BrC/POC ratio shown in Table 2 and Figure 3 is quite confusing. For example, at 350 nm, k=0.005 at BrC/POC=0, and it increases to 0.077 (0.11) at BrC/POC=0.5, but then it decreases to 0.037 (0.051) when the BrC/POC ratio further increases to 1. That sounds counterintuitive - should k increase with the fraction of BrC? Why should it go the opposite direction?

Thank you for identifying this source of confusion. For the base case, k was for all POC. For case 2, k was for BrC alone (not all POC). To eliminate this confusion, we have separated Table 2 into two tables, have modified the caption of Figure 3, and have modified the text.

Table 2: Imaginary part of the refractive index (k) values for the base case with weakly absorbing “colorless” primary organic carbon.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Base Case</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>0.008</td>
</tr>
<tr>
<td>350</td>
<td>0.005</td>
</tr>
<tr>
<td>400</td>
<td>0.005</td>
</tr>
<tr>
<td>450</td>
<td>0.005</td>
</tr>
<tr>
<td>500</td>
<td>0.005</td>
</tr>
</tbody>
</table>
Table 5: Imaginary part of the refractive index ($k$) values for brown carbon (not total organic carbon) inferred for case 2. We include $k$ values associated with multiple densities ($\rho$), and multiple fractions of brown carbon to primary organic carbon (BrC/POC). All four columns for case 2 yield the same absorption.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Case 2 with BrC ($\rho = 1.3$ g cm$^{-3}$)</th>
<th>Case 2 with BrC ($\rho = 1.8$ g cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>BrC/POC = 0.50</td>
<td>BrC/POC = 1.0</td>
</tr>
<tr>
<td>300</td>
<td>0.11</td>
<td>0.051</td>
</tr>
<tr>
<td>350</td>
<td>0.077</td>
<td>0.037</td>
</tr>
<tr>
<td>400</td>
<td>0.052</td>
<td>0.025</td>
</tr>
<tr>
<td>450</td>
<td>0.035</td>
<td>0.014</td>
</tr>
<tr>
<td>500</td>
<td>0.023</td>
<td>0.009</td>
</tr>
<tr>
<td>550</td>
<td>0.015</td>
<td>0.007</td>
</tr>
</tbody>
</table>

I understand that you use the UVAI as a constraint; you have to choose a combination of $k$ and BrC/POC ratio, i.e., if you use a higher $k$ then you would have to use a lower BrC/POC ratio and vice versa, in order to match the same UVAI with OMI. However, this also means that you cannot independently obtain the $k$ or BrC/POC ratio (and BrC density as well) from UVAI, since the four columns of case 2 in Table 2 give you exactly the same UVAI. What is the catch here? This question really should be explicitly discussed in the manuscript.

The catch is that this method eliminates the need to know the BrC/POC ratio. The effective value of $k$ can be inferred for the assumed BrC/POC ratio. We added on line 610 of the revised manuscript, “This effective refractive index of BrC eliminates the need to know the BrC/POC ratio to model the radiative effects of BrC.”

To obtain the $k$ values for BrC through our sensitivity simulations, we assume a fixed spectral dependence and BrC/POC fraction, then we adjust the magnitude of the $k$ values in order to reproduce the absorption observed by OMI for the different BrC/POC cases.

In order to clarify our approach, following lines 431-433:

“Through sensitivity simulations, we derive the effective $k$ values for BrC and the BrC/POC fraction required to reproduce the observed absorption by the OMI UVAI.”

We add:

“This is accomplished by conducting the sensitivity simulations for several cases of BrC/POC fraction and assuming the same fixed spectral dependence for each case. We then adjust the magnitude of the effective $k$ values to match the OMI UVAI.”
At line 448 we also add the sentence: “The effective $k$ values increase with decreasing BrC/POC fraction because as BrC concentration decreases, BrC must be more absorbing to match the absorption observed by OMI.”

These changes should make our method clearer to the reader.

3. Related to the question above, what BrC/POC ratio is used in computing UVAI in Figure 4? If it does not matter as far as you use the consistent pair of $k$ and BrC/POC (and density) to reproduce the UVAI, what is your recommendation to other models to use, especially not every model will be able to compute the UVAI in order to select the pair of $k$ and BrC/POC?

The reviewer is correct that the choice of BrC/POC ratio is irrelevant as long as a consistent pair of $k$ and BrC/POC ratio is chosen. The relationship between $k$ and BrC/POC ratio was derived so that others will be able to choose which fraction makes the most sense to them scientifically, and will then be able to calculate the $k$ value associated with that fraction. We leave the choice of BrC/POC ratio to the user’s discretion. We added to line 612, “Rather, an effective refractive index can be chosen (eq. 6) to represent an internal mixture of BrC and colorless POC.”

We also add to line 471, “The columns with effective $k$ values for BrC/POC fraction of unity offer the convenience of representing the effective absorption by BrC without needing to assume an arbitrary BrC/POC fraction, or to introduce a separate BrC tracer. The effective $k$ values for unity BrC/POC fraction can be thought of as the effective imaginary refractive index for an internal mixture of BrC and colorless POC.”

Other comments:

Page 27411, line 23, “where k is the intercept”: Intercept of what? I think you meant that log($k$) is the intercept of linear fitting of log(AAOD) and log(lambda).

The exponential relationship relating AAOD and AAE is: $\text{AAOD} = k\lambda^{-\text{AAE}}$, where $k$ is just an arbitrary constant that is not important for the calculation of AAE from the AAOD vs $\lambda$ relationship in log-log space.

Page 27411, line 23 (line 281 in the revised manuscript,) we have changed “where k is the intercept.”

To:

“where $k$ is a constant.”

**Referee Comment #2**

This paper uses satellite measurements and model simulations to constrain the optical properties of absorbing organic aerosol (brown carbon), and estimate changes to photo-oxidation and the aerosol direct radiative effect. The authors first show that in a model simulation with only weakly/non-absorbing organic carbon, the ultraviolet aerosol index is biased low relative to satellite measurements. Next, the authors use satellite observations to estimate the wavelength-dependent imaginary index of refraction for brown carbon. This is then used in GEOS-Chem to estimate changes to OH production and the direct radiative effect. The paper is suitable for publication with some minor comments.

It is stated that Figures 1-3 are monthly mean UVAI. I am unclear, though, if you supplied monthly-mean aerosol fields to VLIDORT and calculated the UVAI or calculated UVAI at a higher temporal resolution (daily?) and then took the average. If the former do you expect much difference (perhaps from cloud or
On page 12 line 328 we now state that we use daily aerosol fields at satellite overpass time.

Section 4.2 contains a good deal of information; however, I found it a little hard to follow. The first two paragraphs of Section 4.2 contain background information and could possibly be moved to the introduction. It’s not clear how the effective imaginary index of refraction is determined. I think a few more sentences elaborating on the methods of estimating the imaginary index of refraction for brown carbon are needed (this could perhaps be moved to the ‘methods’ - Section 3, and the results remain in Section 4?). The background and methods described here (in my opinion) tend to disrupt the flow of the results.

Helpful suggestion. We have moved paragraphs from the start of sec. 4.2 to the introduction.

It is difficult to tell the difference between the color of the filled circles and background colors in Figure 3 (I realize this is partly the point). A thicker edge color may help.

We have added a thicker edge color to the circles in Figure 3.

I believe GC-RT assumes externally mixed aerosols. As you are considering an absorbing (and hydrophilic) species, an internal mixture may lead to slightly more absorption. I doubt this would change your general point, but it may be useful to state.

On page 12 line 341 we have added: “Aerosols are treated as externally mixed.”

Tables 1 and 5 are a little difficult to compare. Is there a way to combine them? The paragraph in the text is clearly written (and could possibly stand on its own). You could consider moving the tables and Section 2.2 to the supplement, but that’s just a suggestion.

Table 1 has the headings of each wavelength region we considered in our Absorbing Angstrom Exponent (AAE) analysis, then lists below the literature references falling within that wavelength range. We provide the mean and standard deviation of the literature references for each wavelength region, to compare with our AAE analysis in Table 5 (Table 6 in the revised manuscript). We do not wish to include a supplement with this paper.

In addition to addressing the reviewer comments, we also correct two minor errors in the calculation of the change in OH and DRE, which affected numerical results alone and did not change the structure of the paper.