Anonymous Referee #2:

This manuscript presents a global simulation of primary and secondary brown carbon (BrC) from biomass burning and biofuel emissions. The primary BrC emissions are related to different vegetation types through combustion efficiency based on a fire inventory, while secondary BrC formation is simulated by oxidation of anthropogenic aromatics. The predicted aerosol optical properties are compared with absorption measurements at surface and AERONET data. Direct radiative impact on photochemistry and shortwave radiation is examined with the inclusion of BrC absorption. The content is original and interesting to the community.

We thank the referee for careful reading and valuable comments.

1-1. The method that relates BrC absorption to MCE is largely based on McMeeking (2008), which appears to be a thesis work not published. It would be helpful to provide a link to its electronic or print version for readers' reference since some information about the experiments referred to McMeeking (2008) might be important to understand the limitation and uncertainties of the method presented here.

➔ ***We added a link to McMeeking (2008) reference as follows:***

McMeeking, G. R.: The Optical, Chemical, And Physical Properties Of Aerosols And Gases Emitted By The Laboratory Combustion Of Wildland Fuels, Dissertation, Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado Fall 2008. Available at: http://chem.atmos.colostate.edu/Thesis/McMeeking%20dissertation.pdf

1-2. I asked if the Equation 2 from McMeeking (2008) assumes that all the OC from the biomass burning samples are absorbing BrC. The authors responded "No: : :because the colorless OC does not contribute to CAs absorption, the AAE of Equation 2 is only contributed by BrC and BC". I disagree with the second half of the statement. In fact, BC with clear coatings (i.e., non-absorbing OC) could result in AAE (380nm- 750nm) greater than 1.4 due to the lensing effect, as shown in Gyawali et al. (2009) and Lack and Cappa (2010). So can one assume AAE in Equation 2 due to BrC and BC only? If not, how would it affect the method used to drive the BrC emissions? Given the known large uncertainties associated with AAE, MAE for BrC and even BC, error bars representing uncertainties are certainly needed in Figure 1. Discussions on the propagated errors in global budgets

Gyawali, M., Arnott,W. P., Lewis, K., and Moosmueller, H.: In Situ Aerosol Optics in Reno, NV, USA During and After the Summer 2008 California Wildfires and the Influence of Absorbing and Non-Absorbing Organic Coatings on Spectral Light Absorption, Atmos. Chem. Phys., 9, 8007–8017, 2009.

Lack and Cappa, Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon, Atmos. Chem. Phys., 10, 4207–4220, 2010.

➔ ***Thanks for the constructive comment. We agreed with the reviewer's comment, but we cannot derive the relationship between MCE and BrC/BC ratio without external mixing assumption. Nevertheless, we agreed with the necessity of uncertainty analysis. To examine uncertainties, we conducted additional simulations with three different absorption ratios of BrC to BC (shown in Figure 1 below) and estimated uncertainties of BrC budgets in our model, which is discussed in Section 5.2. Using the new results, we redrew all figures except Figure 2. We changed the text for new simulations as follows:***

Section 2.1:

Figure 1 shows the estimated BrC/BC absorption ratio at 550 nm as a function of MCE. Different lines indicate different AAEs of BC and BrC according to the Table 1 of Kirchstetter and Thatcher (2012). They calculated BC AAE and BrC AAE using 115 wood smoke samples. For the calculation of BrC AAE, BC AAE had to be decided, and they assumed three different BC AAEs (0.86, 1.00, 1.15) based on their smoke samples and previous studies. Resulting BrC AAEs were 5.00, 5.48, and 6.19. We conduct three simulations according to the AAE values of Kirchstetter and Thatcher (2012) (Figure 1), as described later in this section.

Section 4.1:

Figure 3 shows monthly mean simulated and observed light absorption coefficients of BrC at 365 nm averaged over all sites in the southeastern United States for 2007. Black circles and colored bars indicate the observed and simulated BrC absorption at 365 nm, respectively, and different colors in the bar show contributions from different sources. Each panel represents different model simulations with each case for AAE selections as shown in Figure 1 and Table 1.

Section 4.1:

The model generally captures the observed seasonal variation with high absorption in the winter, having a peak in March and low absorption in the summer (R of 0.93). On an annual mean basis, we find that the model is too high by 46% for case 1, and is too low by -31% for case 3, relative to the observations, respectively. The model for case 2 is in the best agreement with the observations (4%) on an annual mean basis.

Section 4.1:

The BrC source contribution in the model is similar to the observed source contribution. Hecobian et al. (2010) showed that biomass burning was the main

contributor for the winter season, whereas the SOC contribution increased during the summer season. The simulated seasonal variation is consistent with the observation, as shown in Figure 3. The annual mass contribution of SOC to BrC is 38% (in case 2), which is in good agreement with the observed contribution of 32% (Hecobian et al., 2010). Based on the results in Figure 3, the model for the case 2 yields best estimates of BrC emissions.

Section 4.2:

Figure 5 shows comparisons of monthly mean simulated versus observed AOD at 500 nm, SSA at 440 nm. We find that the model captures the observed AOD quite well with a regression slope of 0.86 and a R of 0.88. However, the model tends to overestimate the observed SSA, implying that the simulated aerosol concentrations appear to have too large a fraction of scattering aerosols. We find that the inclusion of BrC in the model reduces the high bias of simulated SSA by 33% and 23% (lower left and lower right panel of Figure 5), indicating a considerable contribution of BrC to aerosol absorption. Although the statistics suggest a greater improvement with the case 1 in terms of the bias, simulated SSA values at sites in Africa with high BrC concentrations, are too low apart from the regression line (discrepancy > 0.1). This result also supports our selection of the case 2 as the best model for BrC emission estimates.

Section 5.2:

Table 3 summarizes our best estimates of the global tropospheric budgets of BrC, along with BC and OC. The global BrC source is 12.5 ± 3.0 TgC yr^{-1}, which accounts for 27% of OC sources. Although the biofuel emission (6.5 TgC yr^{-1}) is three times lower than the biomass burning emission (22.7 TgC yr^{-1}) for OC, the biofuel emission (3.0 ± 1.3 TgC yr^{-1}) becomes significant for BrC, contributing about 43% of primary sources. The secondary source of BrC is 5.7 TgC yr^{-1}, and is comparable to the primary sources (6.8 ± 3.0 TgC yr^{-1}).

Section 6:

Although the DRE of BrC at the top of the atmosphere is nearly zero, the increased DRE of OC after considering BrC absorption (usually considered as scattering OC) is 0.11 W m^{-2}, as shown in Figure 8(c). The DRE of OC without BrC absorption is -0.69 W m^{-2} (Figure 8(d)), and this value is increased to -0.58 W m^{-2} after considering BrC absorption. Consequently, the cooling effect of OC is reduced by 16%.

Section 7:

Finally, we quantify the effects of BrC on global NO_2 photolysis rates and ozone concentrations at the surface. Figure 9 shows changes in annual NO_2 photolysis rates and O_3 concentrations in surface air owing to BrC absorption. Although BrC absorption is included, there are no significant changes (less than 1%) of the global

mean NO_2 photolysis rate and O_3 concentration in surface air. However, the effect of BrC appears to be important for regions with high BrC concentrations. We find a maximum decrease of the annual mean NO_2 photolysis rate by 8% in surface air over Asia where the resulting reduction of O_3 concentration is up to -2 ppbv (6% of annual mean surface O_3 concentration). We also find that the BrC effect has a strong seasonal variation such that it is maximized in the spring when surface O_3 concentration is decreased up to -13% in Asia because of high BrC concentration (55 μgC m^{-3}). This maximum O_3 decrease by BrC (-13%) is similar to the 15% O_3 reductions by the aerosols from fires calculated by Jiang et al. (2012).

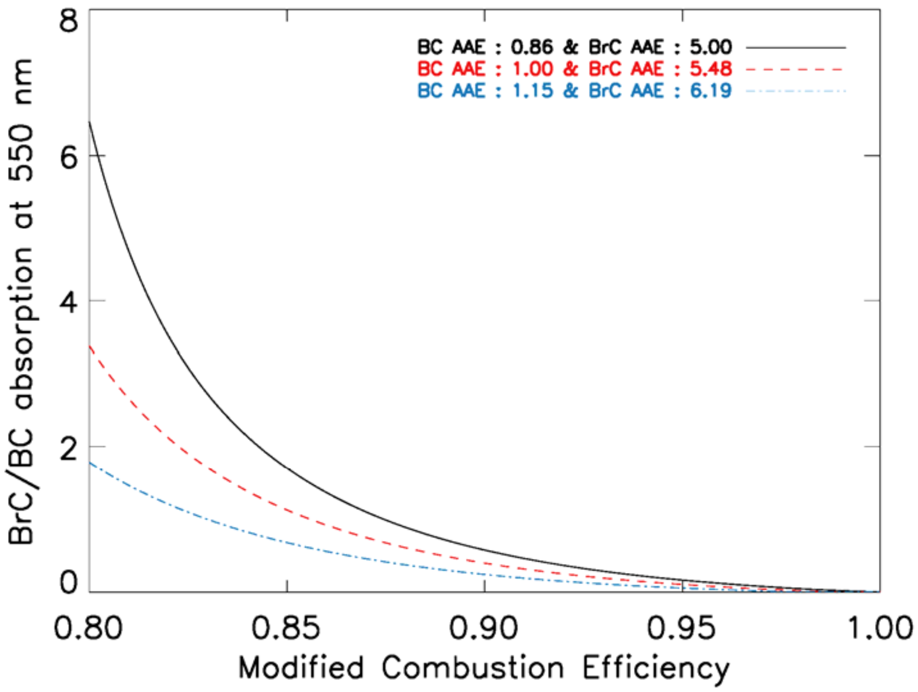

Figure 1. Estimated absorption ratios of BrC to BC at 550 nm as a function of MCE. We assume that the CA absorption is only contributed by BC and BrC absorption. Black solid line indicates case 1, red dashed line represents case 2, and blue dotted line shows case 3.

2-1. In order to best-match the observed absorption at 365nm, 40% lower MAE values than those used to derive emissions have to be used for BrC optics in aerosol optical comparison and radiative transfer calculations. Are MAE values at other wavelengths lowered similarly? Can you use the low MAE values in obtaining the BrC/BC absorption ratio in emission estimating? Would it increase the BrC/OC fractions and primary BrC emissions estimated?

➔ ***Thanks for the constructive comment. We removed our discussion of the use of 40% reduced MAE value, and instead added new discussions for the uncertainty analysis based on three different absorption ratios of BrC to BC as shown in Figure 1 above.***

2-2. The calculated BrC/OC fractions in Table 1 are much lower than those used in previous studies except for cropland, but the estimated direct radiative effect due to BrC absorption is similar to others (_0.12 Wm^{-2}). Can you explain why?

➔ ***The global mean value in figure 8(c) (0.11 W m^{-2}) is a relative increment of BrC DRE assuming between absorbing BrC and scattering BrC. The DRE of BrC itself is -0.018 W m^{-2} (0.010 - 0.028 W m^{-2}). The BrC DRE of Park et al. (2010) is 0.05 W m^{-2}, which is higher than our value. Note that the imaginary refractive index of BrC in Park et al. (2010) (0.27 at 550 nm) is higher than the value used in this study (0.10 at 532 nm), but our value is higher than that of Feng et al. (2013) (0.03 at 550 nm in strongly absorbing BrC case).***

2-3. How do the model-predicted AAEs compare with the AERONET data, in addition to SSA at 440nm? The AAE comparison would give indications about the simulated BrC/OC or BrC/BC fractions, which presumably is one of the main improvements in this study compared with previous studies.

➔ ***As the reviewer suggested, we evaluated the model against AERONET AAE as shown in Figure 2 below. We found that the model result is degraded after including BrC, in part, because the model underestimates BC emissions as discussed in the main text. However, the simulated AAE will be decreased if we increase BC emissions as suggested by the top-down estimate (Cohen and Wang, 2014). We expect that the model with BrC will be improved if BC underestimation of the model is resolved. For example, we further evaluated the model for regions (North America, Central America, South America, Southeast Asia, and Australia) where the difference between our BC emission and the top-down estimate is within a factor of 2, and found that the model with BrC shows a better agreement with the observed AAE (Figure 3). We also found that our SSA evaluation results are improved for these regions (Figure 4).***

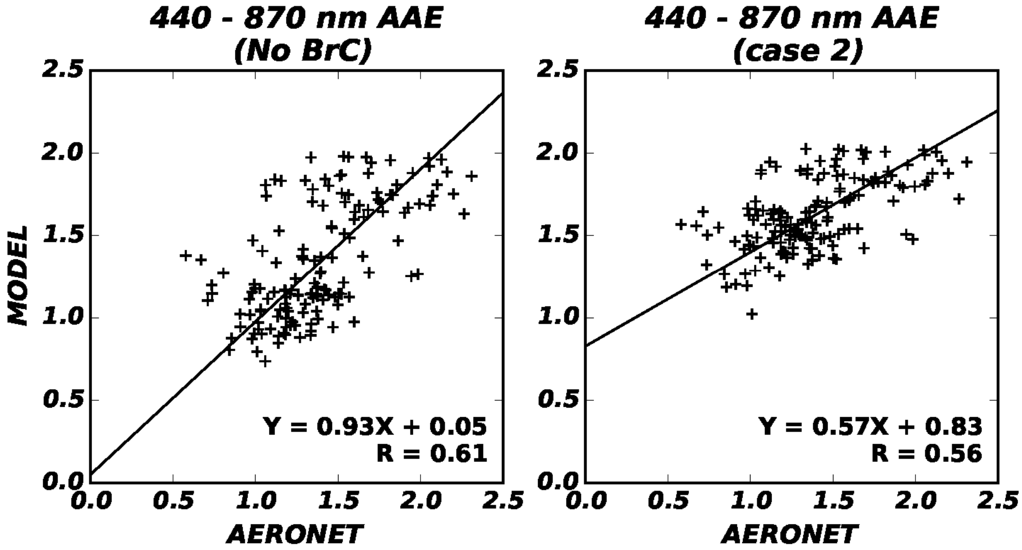

Figure 2. Scatterplots of simulated versus AERONET 440 – 870 nm AAE for the without BrC (left) and with BrC of case 2 (right).

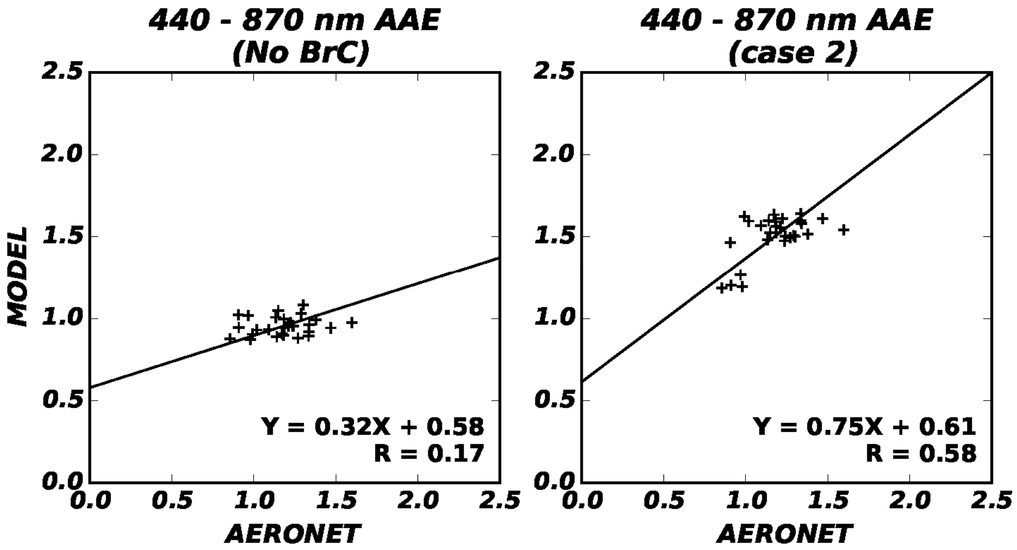

Figure 3. Same as Figure 2 above but for the regions where the BC emission ratios between our model and top-down estimates from Cohen and Wang (2014) are less than 2.

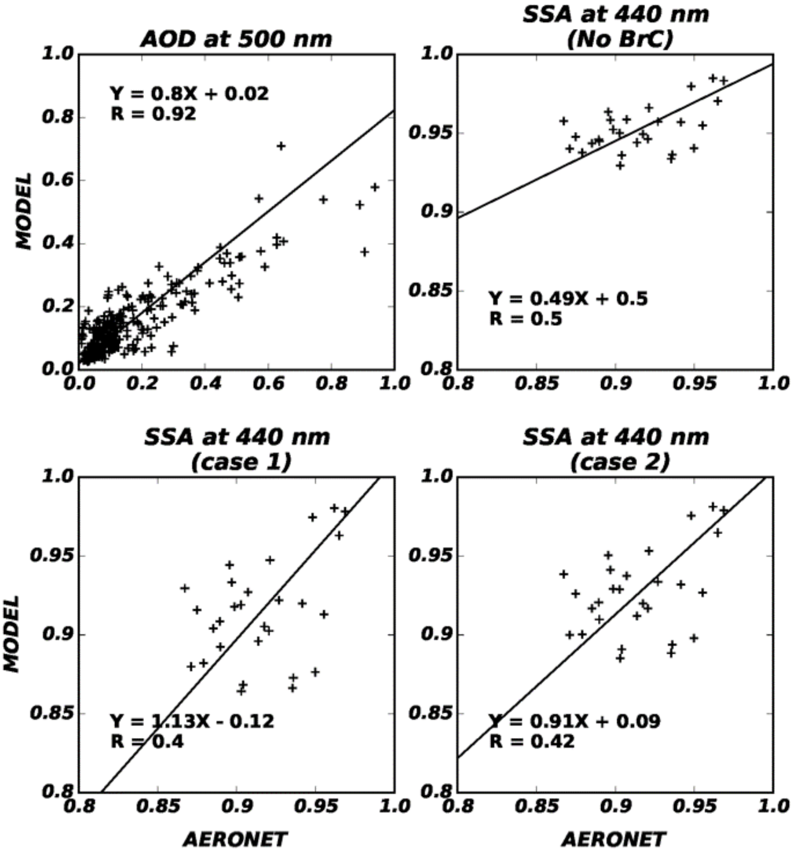

Figure 4. Same as Figure 5 in the main text but for the regions where the BC emission ratios between our model and top-down estimates from Cohen and Wang (2014) are less than 2.

3-1. Description for biofuel BrC emission factors (Table S1) is still a bit confusing. How do the biofuel EFs relate to the MCE? based on the Fire inventories for open biomass burning?

➔ ***Yes, you are correct. We changed the Table S1 for the clarity as shown in Table 1 below. In order to estimate BrC/OC ratio from biofuel, we have to know OC biofuel emissions for each sector. However, we have total biofuel emissions alone. Therefore, we first estimated the emission of each sector by multiplying biofuel consumption by EF. Finally, we calculated BrC/OC ratio for each sector with the same process for biomass burning.***

Table 1. Global biofuel consumption estimates, EFs of OC, and OC biofuel emission estimates for each biofuel category. Base year is 2000.

		Fuelwood	Crop Residues	Dung	Charcoal	Industrial	Total
Biofuel Consumption[1] [Tg]		1351	495	75	39	498	2457
EF [g/kg][2]		2.97[3]	3.3	1.8	1.3	0.91[3]	2.6
OC emission [Gg]		4010.3	1633.5	135	50.7	453.6	6281[4]
BrC/OC	case1	0.653	0.946	0.000	0.000	0.000	
	case2	0.442	0.652	0.000	0.000	0.000	
	case3	0.261	0.400	0.000	0.000	0.000	

1) From Fernandes et al. (2007)
2) From Bond et al. (2004)
3) Global mean value is estimated from Bond et al. (2004)
4) From GEOS-Chem biofuel OC inventory (carbon_200909) by Bond et al. (2007)

3-2. What is the overall BrC/OC fraction for biomass burning? Is it lower than biofuel? How do they compare with the BrC/OC fractions from SOC?

➔ ***Overall BrC/OC fraction for biomass burning is 0.25, which is lower than that for biofuel (0.45). BrC/OC from SOC is 0.18 in this study.***

3-3. Are there any laboratory studies that could support these estimates?

➔ ***We did not find laboratory studies, supporting our BrC/OC ratio yet.***

3-4. I think that Table S1 should be included in the main text instead of the supplementary as biofuel is as important as biomass burning.

➔ ***We agreed with the reviewer's comment and moved Table S1 to Table 2 in the main text.***

3-5. In Table S1, the BrC/OC fraction of 0.663 is mistakenly placed in the row for BrC/Biofuel. Then the total burden of BrC from biofuel should be 0.663*6.281=4.16 Tg, not 4.3 Tg. Please also check other numbers in Table S1.

➔ ***Total biofuel OC emission is increased to 6.536 Tg in 2007 (base year of BrC budget discussion) from 6.281 Tg in 2000, which is base year of the Table S1. 6.536 Tg * 0.663*** *gives 4.3 Tg. We are sorry for the confusion and added the base year information in Table caption and the main text.*

➔ *We added and changed the text as follows:*

Our method of estimating BrC emissions from biofuel use is similar to that of estimating emissions from biomass burning. We estimate BrC/OC ratio using the MCE and BC to OC ratio in the same way as the biomass burning estimates. The only difference is that the biofuel emission of each sector is not known (the biomass burning emission is known for each vegetation type). Therefore, we first estimate OC biofuel emissions from each biofuel category with the information given by previous studies (Bond et al., 2007; Bond et al., 2004; Fernandes et al., 2007). Because there is no clear evidence that BrC is emitted by dung, charcoal, and the industrial sector, here we consider only fuelwood and agricultural residue as BrC sources. Fuelwood burning is the largest contributor to biofuel BrC emission. Our estimate of BrC/OC mass ratio is 0.271 - 0.663 from biofuel use. Overall results are summarized in Table 2. Note that base year of Table 2 is 2000 because previous studies reported their values based on 2000. We scale up the emission for 2007 as described in Section 3.2. Resulting BrC emission from biofuel use for 2007 is 3.0 ± 1.3 TgC yr^{-1}, which is comparable to BrC emission from biomass burning.

Minor comments:

1. Line 1, Page 27807: add “until recently” following “.. as light-scattering aerosols”

➔ ***We added it.***

2. Line 3, page 27807: replace “climate effect” with ”direct radiative forcing”

➔ ***We replaced it.***

3. Line 7, page 27807: replace “shortwave” with “ultra-violet”

➔ ***We replaced it.***

4. Line 2, page 27808: add “and also strongly absorbing” following “. . .HULIS”

➔ ***We added it.***

5. Line 12, page 27810: attenuation means both scattering and absorption. Is it just absorption Angstrom exponent measured?

➔ ***We think that “attenuation” and “absorption” are not significantly different in this case. Because light transmission through particulate matter on filters is predominantly due to particle light absorption rather than particle light scattering when the particulate matter is even weakly absorbing (Kirchstetter and Thatcher, 2012; Arnott et al., 2005).***

6. Line 19, page 27810: delete citation: McMeeking, 2008; isn’t the equation 2 from McMeeking, 2008?

➔ ***We deleted it.***

7. Line 25, page 27813: reference for the secondary BrC MAEs at 365nm and 550nm?

➔ *We added the references.*

8. Appendix A1: line 17, page 27829: 4.1 for the 350-400nm pairing, while 8.0 for the 550-600nm pairing? Or it's reversed?

➔ *4.1 for the 350-400nm pairing, while 8.0 for the 550-600nm pairing. We changed the sentence for the clarity as follows:*

However, the AAE of BrC using just two wavelengths is 4.1 for the 350-440 nm and 8.0 for the 550-600 nm, respectively.

9. Appendix A1, lines 3-4, page 27830: this fitting is not clear to me. Because the slope variable (Angstrom for CA) also appears in the residual term C, which is the intercept B, can the obtained fitting function satisfy the slope (A) and the intercept (B) at the same time? It would be nice to plot an example for the linear regression with one F value.

➔ *Figure 5 shows an example of the linear regression for F = 4.0. In this case, R^2 is 0.99 and Angstrom exponent of CA is 4.44. Y-intercept of the numerical fitting is -29.81, which is consistent with Y-intercept (-29.64) from Eq. (A5) below. We tested results with different F values, and found that the difference between two Y-intercept values are always within 1%. Because this is numerical fitting and R^2 is not 1.0, Y-intercept values from two methods are slightly different, but we think this is acceptable. Therefore, we think the numerical fitting function satisfies both the slope (A) and the intercept (B) at the same time within 1% error.*

$$\text{Y - intercept} = -\mathring{A}_{CA}\log(\lambda_0) - \log(1+F) \qquad \text{(A5)}$$

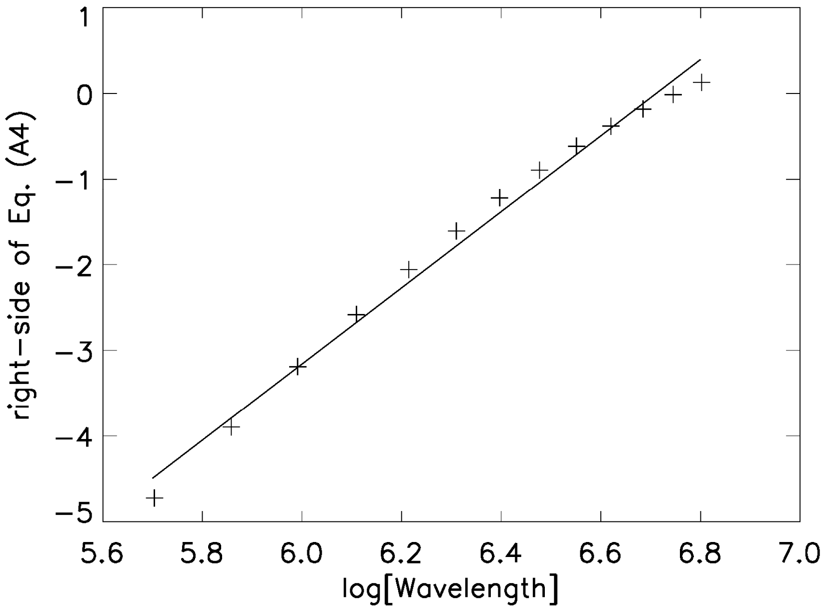

Figure 5. Linear regression fitting of Eq. (A4) when F = 4.0. X-axis is logarithm of wavelength and Y-axis is right side of Eq. (A4).

10. Line 5, page 27830: what wavelengths does the selected Angstrom for BrC correspond to?

➔ ***Over the 360 to 700 nm spectrum range. We added this information in the text as follows:***

We choose the $Å_{BrC}$ values of {5.0, 5.48, 6.19} and the $Å_{BC}$ values of {0.86, 1.00, 1.15}, following Kirchstetter and Thatcher (2012), who estimated mean $Å_{BrC}$ using several wood samples (87 samples) over the 360 to 700 nm spectrum range.