

We thank the reviewer #2 for providing helpful comments and suggestions for improvement of our manuscript. Below are our responses (in normal fonts) to reviewer comments (in bold and italic) including descriptions how we modify the manuscript.

***TITLE: Can you make it more specific to the work featured in this paper? E.g., I think that it would be appropriate to replace COMBUSTION with PYROLYSIS.***

We replaced corresponding “combustion” with “pyrolysis” in the body of the paper, and added the following paragraph to explain how pyrolysis and combustion are related in P. 20475 Line 8. We did not change the title just in case that some readers from atmospheric chemistry might not be familiar with the difference between pyrolysis and combustion.

Paragraph added: “Wood combustion comprises at least two distinct processes that affect the properties of the resulting aerosol: release of volatile material from the solid, and its condensation or combustion in the atmosphere. Our combustor simulates the release of volatile matter at realistic wood temperatures; there is no oxygen either inside the wood or in the diffusion flame. In a real burn, after this material leaves the solid wood, it may either condense to form “organic carbon” or burn in a diffusion flame to produce black carbon. Our goal in this experiment is to understand the nature of the devolatilizing material generated by wood pyrolysis. This material is, emitted directly.” This point is also stated in the Response to Anonymous Referee #1.

***ABSTRACT: I think it is important to make the Abstract crystal-clear b/c many will read only this part of the manuscript. I note here some things I think could be stated more precisely and/or clearly, without appreciably increasing the total number of words that make up the abstract. I first restate in quotes the author's sentence(s) and then below it provide one or more comments.***

***"While BC is highly absorbing, some organic compounds also have significant absorption, which is greater at near-ultraviolet and blue wavelengths."***

***This sentence could be interpreted as meaning that some organic compounds are more absorbing than BC at near UV and blue wavelengths. It could also be inferred from this sentence that while OC absorption increases at near UV and blue wavelengths, BC absorption does not.***

We changed the sentence to “While BC is highly absorbing, some organic compounds also have significant absorption, especially at near-ultraviolet and blue wavelengths.”

***"To the extent that OC absorbs visible light, it may be a non-negligible contributor to direct aerosol radiative forcing."***

***This sentence seems to neglect the OC scattering of sunlight. Isn't it true that, even if OC does not absorb sunlight, OC may contribute non-negligibly to direct radiative forcing.***

***"To the extent that OC absorbs visible light, it may be a non-negligible contributor to direct aerosol radiative forcing." and subsequently "A simple model suggests that, despite the absorption, both high-temperature and low-temperature carbon have negative climate forcing over a surface with average albedo."***

***Are RADIATIVE forcing and CLIMATE forcing the same? (I think they are not.) I would reverse the usage and say something like this in the first: "It has become widely recognized that OC aerosols absorb ultraviolet and visible light, but the atmospheric science community has yet to reach a consensus on (or evaluate in detail) whether or not this absorption affects climate." In the last sentence, I would say ".... negative RADIATIVE forcing ..." if your are referring only to predicted W/g, since climate is more comprehensive (including feedbacks that influence efficacy, impact on precip, etc).***

We changed the sentence of "To the extent that OC absorbs visible light..."to "To the extent that OC absorbs visible light, it may be a non-negligible contributor to the positive direct radiative forcing. Quantification of that absorption is necessary so that radiative-transfer models can evaluate that hypothesis."

We changed the sentence of "A simple model..."Changed to "A simple model suggests that.....carbon have negative radiative forcing over a surface with average albedo"

***"Higher wood temperature is the main factor creating organic aerosol with higher absorption, causing about a factor of four increase in mass-normalized absorption at visible wavelengths."***

***The first part of this sentence is qualitative while the second is quantitative. I recommend noting the magnitude of the temperature increase (E.g., Increasing temperature from ABC to XYZ degrees) that led to the x4 increase in absorption. This would be meaningful.***

We changed the sentence to "Higher wood temperature is the main factor creating organic aerosol with higher absorption; changing wood temperature from a devolatilizing state of 210 C to a near-flaming state of 360 C causes about a factor of four increase in mass-normalized absorption at visible wavelengths."

***"A simple model suggests that, despite the absorption, both high-temperature and low-temperature carbon have negative climate forcing over a surface with average albedo."***

***In this sentence, I think you should add the word ORGANIC in front of carbon, unless you mean to include BC as well, in which case you should say so explicitly.***

***Minor point: "... have negative climate forcing" the word HAVE sounds odd to me. Should HAVE be replaced with RESULT IN A, or something similar.***

***I reiterate that CLIMATE forcing is not the same as RADIATIVE forcing, at least not to me and others I have spoken with.***

***Noting that the model is simple makes me wonder what a more sophisticated model would predict, and if the results of a simple model are worth much. I know this is mentioned in the discussion section, but it could be handled in the abstract as well. Perhaps instead of saying SIMPLE you might be a bit more descriptive. E.g., "A model that included features A and B predicted that light absorbing OC imparts a negative climate forcing over surfaces with average albedo." And perhaps add "It is recommended (or not?) that including climate-relevant features C and D using a more complex model be investigated."***

***If average albedo is simply a number, replace AVERAGE with the number (or a range).***

***How does this sentence relate to the global dimming phenomena. Isn't it true that both absorbing and scattering aerosols result in dimming at the ground? Is the negative climate (RADIATIVE) forcing to which this sentence refers a forcing that is calculated for the ground, top-of-the-atmosphere, or something else? Can this be clarified in the abstract (and in the body of the paper).***

We agree that both absorbing and scattering aerosols result in dimming at the ground. Based on the review's comments on radiative forcing model, we changed the statement in the abstract with the following paragraph. "A clear-sky radiative transfer model suggests that, despite the absorption, both high-temperature and low-temperature OC result in negative top-of-atmosphere radiative forcing over a surface with an albedo of 0.19 and positive radiative forcing over bright surfaces. "

We added the following NOT in abstract but in text (P. 20494 Line 5): "Global average radiative forcing of absorbing OC, considering its location above and below clouds and relative to bright surfaces, should be assessed with a general circulation model."

***It may not be necessary for most readers, but OC and BC should be defined. Also it may be wise to use fewer terms when referring to the materials on which the paper focuses. In the abstract you have the following: organic compounds, OC, organic carbon, organic aerosol, and high- and low-temperature carbon - all of which apparently refer to the same thing.***

We defined BC and OC in the second sentence in the abstract (P. 20472, Line 2): “While black carbon (BC) is highly absorbing, some organic carbon (OC) also has significant absorption...”

We changed “organic compounds”, “organic carbon”, “organic aerosol” to OC.

***GENERAL COMMENTS: The absorption (and scattering) of UV light by organic aerosols may influence photolysis-driven chemistry in the troposphere. Some have suggested impacts on ozone, for example. Can you add anything new to this discussion in addition to what you mention in the intro? Maybe the OC absorption is more important to include in predicting tropospheric chemistry than in direct-radiative forcing in general. Maybe not.***

We added the following as Sect. 4.4 in the paper:

The absorption of UV light by OC may influence photolysis-driven chemistry in the troposphere. Although this is not a focus of our study, we briefly discuss the potential impact. Jacobson (1997) investigated the effect of absorbing aerosols on photolysis ratio coefficient and ozone mixing ratio with the GATORM model, by applying the refractive index of liquid nitrobenzene as a surrogate in model simulations. It was concluded that absorbing organic aerosols may cause the reduction in UV photolysis coefficients, resulting in up to 5-8% decrease in near-surface ozone mixing ratio during the Southern California Air Quality Study period. The UV absorption of OC obtained from our samples generated at 360C is lower than that in the Jacobson (1997) study by 30-40%, depending on wavelength. It can be inferred that the significant UV absorption by total OC from wood pyrolysis could affect photolysis rates and thus ozone mixing ratio by a few percent.

***“... although this method measures the bulk liquid refractive index and not particulate absorption” - What if the interaction of light with the material in the particle phase is appreciably different than in the extracted phase? I think it is important to discuss this at least at some level, even if only to explicitly say that it is a source of uncertainty, or to say that the comparisons with other studies (e.g., in Fig 9) suggest that it is not a major caveat. Also, would the real-time 3-wave PSAP data be helpful to address this?***

The bulk liquid and particulate absorption might differ for two reasons: (1) Bulk absorption differs from particulate absorption; and (2) Absorption by a dilute suspension differs from absorption by a concentrated material. We address both of these below.

(1) We had originally developed a section in which the difference between liquid and particulate absorption was compared. That discussion was based on the presentation in Sun et al. (2007), where a relationship for the ratio between the two values  $\xi$  was presented for the small-particle limit.  $\xi$  is a function of both real and imaginary parts of

the refractive index. Depending on particle size, absorption per mass by particles can be double that of bulk liquid if the particles are in the resonance region where surface currents contribute to absorption. Such high values of enhancement occur for less absorbing particles. However, we removed that discussion because of length considerations. The paper as it stands provides values of refractive index that can be used in Mie calculations to produce values of particulate absorption and a longer discussion would detract from the main point of the paper.

(2) Dilution studies showed no concentration-dependent differences of absorption per mass. We acknowledge that our dilution studies did not reach the high concentrations in atmospheric aerosol and we have added the following statement in the paper (P. 20476, Line 17): "Our procedure may yield inaccurate results if very concentrated material has different absorbing properties than diluted material, and this possibility should be investigated in future work."

Finally, we agree that the PSAP measurements might shed additional light on the comparison between papers. However, filter-based absorption measurements for liquid aerosol are prone to artifacts (Subramanian et al, 2009; Lack et al, 2009) and our measurements are no exception. The PSAP measurements do not match Mie calculations. This must be explored in a separate paper, and we have decided to delay that exploration until we have measurements that do not rely on filters. These measurements are being pursued now.

***I find that the last sentence in the abstract does not have the same impact as the following sentences in the discussion, "Unless absorption by real ambient aerosol is higher than that measured here, representing it is probably not important for global average clear-sky forcing." and "The contribution of absorbing organic material could be important in energy balances over bright surfaces." I.e., perhaps the abstract misses the punchline.***

We added the following to the abstract.

"Unless absorption by real ambient aerosol is higher than that measured here, it probably affects global average clear-sky forcing very little, but could be important in energy balances over bright surfaces."

**Reference:**

Jacobson, M. Z., Studying the effects of aerosols on vertical photolysis rate coefficient and temperature profiles over and urban airshed, J. Geophys. Res., 103, 10593-10604, 1998.