Atmos. Chem. Phys. Discuss., 10, C2130–C2135, 2010
www.atmos-chem-phys-discuss.net/10/C2130/2010/

Interactive comment on "Brown carbon in tar balls from smoldering biomass combustion" *by* R. K. Chakrabarty et al.

Anonymous Referee #1

Received and published: 27 April 2010

In this paper, the authors report the observation of spherical tar balls emitted from smoldering combustion of two fuels. The authors experimentally determine the optical properties of these tar balls and infer brown carbon (BrC) as a primary component of the tar balls. The imaginary refractive indices of BrC are estimated and then applied in the sensitivity calculation of the radiative forcing efficiency (RFE) by compared with traditional organic carbon with no absorption at visible and UV wavelengths. They conclude that accounting for the absorption by BrC leads to an increase in aerosol RFE and therefore suggest the importance of the inclusion of optical properties of the tar balls into future radiative forcing models.

This paper takes a useful approach toward understanding the optical properties and radiative forcing of brown carbon from smoldering biomass combustion, however, the

sample size discussed in this manuscript is too small - only includes three samples of two different wood types - to convince the community that tar balls with similar properties also exists in other smoldering biomass combustions. The authors need to provide more data from reproducibility tests and other samples to support their findings and therefore draw the attention from the research community that optical properties of tar balls should be included in the radiative forcing models. Furthermore, the estimation of k_{BrC} has to be verified. Overall I recommend the publication of this paper after consideration of the specific comments as stated below.

- Page 6280, line 2, can the authors be more quantitative about "large-scale production of spherical, carbonaceous particles", i.e., how much percentage of spherical carbonaceous particles were found in their samples?

- Page 6280, line 4, can the authors be more specific on the "real-time measurements", e.g., spectrally varying AAC, refractive index and so on?

- Page 6280, line 9, this manuscript is talking about absorption in the visible and near-visible range (405nm, 532nm and 780nm), but the statement here is that "...accounting for UV absorption by brown carbon leads to a significant increase in aerosol radiative forcing efficiency and increased atmospheric warming". This "UV absorption" is mentioned twice in the abstract and also elsewhere in the conclusion. 405nm could be considered as a near-UV range, but I don't think it is within the UV range, so the UV absorption should not be emphasized in this manuscript.

- Page 6280, line 10, the authors should be careful to say this is a "significant" increase in aerosol radiative forcing efficiency. This sentence is also confusing by saying "increased atmospheric warming" since it seems like brown carbon is warming the atmosphere while actually the net effect of brown carbon is cooling the atmosphere.

- The authors say we should consider brown carbon optical properties or we get the RFE wrong. What are RFE of other substances? How much of a change is this RFE compared to that of prevalent substances such as sulfate and black carbon?

- Page 6280, line 24, brown carbon does absorb some solar radiation in the blue and near UV ranges, however, I am suspicious to say that this absorption is strong.

- There are two main terms in this manuscript as stated in the title: brown carbon and tar balls. The authors should first give a clear definition and introduction of these two terms at the beginning of this manuscript and cite more literatures about the tar ball identification in Page 6281, line 13.

- Page 6281, the third paragraph, the authors need to explain how representative are PPduff and AKduff of general combustion. What type of combustion would this represent and how prevalent is it? Following this point, in page 6282, the first paragraph, the authors need to address the question of to what extent does their experimental methodology actually mimic a "real" burn.

- Page 6282, line 13, the authors may state briefly what the modified combustion efficiency is. Was this also measured during the sampling?

- Page 6285, line 1, did all or most of the single particles analyzed under SEM look like near-sphere? What is the proportion of these near-sphere tar balls of all the particles from the emission? It would be more convincing if the authors could state the proportion of tar balls or provide a SEM image in which a large part of near-sphere particles were observed in addition to the SEM image for a single particle. Whether the particles are spherical or not directly relates to the validity of the use of Eq (1) to retrieve particle size-number concentration and the use of Mie theory to calculate the refractive indices.

- Page 6285, the first paragraph, the authors should clarify which statements are based on the analysis in this study and which are cited from other literatures. For example:

- How did the authors identify that the smoke particles in this study are homogeneous tar balls? Did the author just inferred this from other literatures or there was actually a way to identify this in this study?
- The authors mentioned that the EDX analysis shows that tar ball particles consist

primarily of carbon and oxygen with an average molar ratio of about six. Is this a result from EDX analysis in this study?
- If the EDX analysis was performed in this study, since polycarbonate filters which primarily contain carbon were used for SEM-EDX analysis, how can the authors differentiate the carbon of tar ball particles from the total carbon detected by EDX which also includes the carbon from the filters?

- Page 6282, line 5, can the authors provide the concentrations of particles going through all the instruments? If the concentration is much higher than ambient concentration, some gaseous-phase carbon would condense on particulate phase and thus was taken into account here. However, this carbon may not exist in the particulate phase any more in the ambient atmosphere. How do the authors think the optical properties obtained in this manuscript can represent the real condition when this kind of brown carbon is in the ambient atmosphere? Is the increase of radiative forcing really significant for AMBIENT brown carbon?

- Page 6286, line 13, the authors should cite more original papers which did measure AAC of brown carbon. Moreover, WSOC mentioned here is kind of distracting. How does WSOC relate to brown carbon?

- Page 6287, line 4, in the concept of "SSA Angstrom exponent", the authors are trying to have an Angstrom (which is the ratio of two things) for another ratio (SSA), which is confusing. I suggest the authors present the Angstrom for absorption and scattering separately and compare them for different wavelengths, which has a much more physical meaning rather than having such a convoluted value.

- Page 6287, line 9, was the negative SSA Angstrom coefficients observed for brown carbon in other literatures?

- Page 6287, line 18, the authors need to explain more on Equation (3) and the estimation and k_{BrC}.

- To my understanding, k_{BC} and k_{BrC} is not the actual imaginary refractive indices for black carbon and brown carbon, while they are the refractive indices after considering the volume fraction of BC and BrC in the sample. This means kBC and kBrC are dependent on the volume fraction of BC and BrC. This point should be clarified in the manuscript. Otherwise, the dotted line in Fig 4 representing kBC may confuse the reader with the actual imaginary refractive index of pure BC which is around 0.74.
- Could the author give the estimated fraction of BC and BrC based on the calculation using Eq (3)? k_{BrC} was applied to estimate the RFE, if kBrC is just the imaginary refractive index considering a certain fraction of BrC, I suspect the actual imaginary index of pure BrC would be higher (maybe not that much though). How do the authors think about this part of uncertainty in the estimation of REF?
- The authors state that the BC content in PPDuff2 is less than that in PPDuff1, indicating a higher BrC content in PPDuff2. Could the authors explain why two different BrC contents can result in the same BrC component of the imaginary refractive index?
- Could the authors explain more about the validation of Lorentzian function applied here? Can the authors provide support from the literatures that the Lorenzian function is valid to be applied here?

- As stated at the beginning, the sample size discussed in this manuscript is very small. The measured data and modeled curve of imaginary refractive indices fit very well with each other in Fig. 4, but just for the data from very few samples without any reproducibility tests. This raises the questions that whether the RFE calculated later in this manuscript be representative for the brown carbon emitted from real smoldering biomass combustion in a certain region or even globally, and thus may weaken the statement of the importance of including brown carbon into radiative forcing mod-

els. The authors need to provide data from reproducibility tests and more samples to support their findings.

- Page 6288, line 24, the authors may explain how the calculated k_{BrC} is applied in Eq (4) to calculate the RFE.

- Page 6295, Table 1, are the values of scattering and absorption coefficients averaged over the whole sampling period?

- Page 6295, 6296, Table 1 and Table 2, the authors need to rearrange the layout the tables to make the values of refractive index be easily read. Table 2, column 3, the unit of RFE was separated into two lines.

- Page 6298, Fig. 2, if the EDX analysis is not a result from this research, it may not be appropriate to state here. Or the authors should add the citations.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6279, 2010.