

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

### **Anonymous Referee #2**

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Chakrabarty et al present optical measurements and modeling on particles produced from the smoldering of a variety of duffs. The authors assert that “tar balls” are the major particle type emitted from duff smoldering. The authors also state that “tar balls” are composed of “brown carbon”. From this assertion, they have used Mie theory to retrieve optical properties in order to calculate a radiative forcing efficiency. These results are of use, but I think more evidence is needed to support the various assumptions used in this study. Furthermore I would encourage more effort (such as a sensitivity study) by the authors to show that their estimates are reasonable. Having the advantage of being able to read the other reviewer’s comments, I am pleased to see that they hit on many of the same points that I have. Therefore, the points below should be addressed before the manuscript is accepted for publication.

## Detailed Comments:

Title and introduction: Personally, I do not like the use of the non-scientific terms “tar ball” and “brown carbon” because of their poorly defined meanings. “Brown carbon” is slightly better because it connotes weak absorption in the visible region. Even better would be “light absorbing carbon (LAC)”. In the end, there is graphitic carbon (soot) and organic carbon; the organic carbon can have a range of optical properties from non absorbing (traditional OC) to weakly absorbing (brown carbon, tar balls etc. . .). Please give a detailed, precise, and succinct definition of brown carbon and tar balls.

P 6279, Author List: Claudio Mazzoleni’s name needs an “i” at the end of it.

P 6280, line 2: Since these are laboratory studies, I disagree with calling this observation “large scale”. Since this is the field of geosciences, I would think “large scale” refers to global or continental scale. I think it would be unique if tarballs were produced in the laboratory, so changing “large scale” to “laboratory” might help this paper stand out.

P 6280, lines 7-12: The author draws the distinction between “brown carbon” and “traditional” organic carbon. Technically, I don’t believe that there is any difference between organic carbon and brown carbon. Brown carbon is organic carbon. What has changed is the traditional view (by the atmospheric geosciences community) that organic carbon does not absorb light. Therefore, I suggest a re-wording here.

P 6280, lines 24-25: I haven’t seen convincing evidence of brown carbon having  $k > 0.01$ . Compared to soot ( $k = 0.5$ ), I wouldn’t call this “strongly absorbing”. However it may still be important due to the large observed mass concentrations of organic species. Suggest re-wording this sentence.

P 6281, line 1: I don’t get how the 88

P 6281, line 7: I found it surprising that a duff could smolder for more than a month. This is interesting. Please provide a reference for this.

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P 6285, line 2; Figure 1: Since the entire analysis presented in this paper depends on the assumption that all particles were homogeneous, spherical tar balls, I think more evidence is needed to show that all of the particles in these samples were indeed homogeneous spheres. This point could be made by showing that they have examined a statistically relevant number of particles, and that some high fraction of those particles was a “tar ball”. I suggest that the authors show several images of tar balls particles from each sample. I have personally seen images of the pine duff sample and am not convinced that every particle is a homogeneous sphere.

P 6285, line 18: I wonder how chemically similar tarballs are between studies and even between sampling times in the same study. Biomass burning composition has been shown to undergo chemical reactions with plume age as a result of photochemical oxidation, which may change optical properties (Capes et al., 2008). Some discussion of how this relates to the results in this paper should be discussed.

P 6285, line 19: BC particles may also be compact (collapsed).

P 6286 line 4: Suggest the start of a new paragraph here.

P 6286, line 24: The explanation of SSA should be given earlier, along with its definition (lines 18-19).

P 6287, line 13-20 and Figure 4: From Figure 4, it is seen that the magnitude of  $k_{BC}$  is less than 0.01. The refractive index of pure BC is generally around 0.5 (Bond and Bergstrom, 2006). What the difference between these two values? If the value of  $k$  is not the refractive index intrinsic to the material, I suggest denoting this by “effective” or “retrieved”. I understand that the Mie derived refractive index is apportioned to black carbon and brown carbon (eq 3). Are the imaginary refractive indices reported here somehow related to their mass concentration? If so, it would make the analysis much more clear to explicitly state this. Along these lines, it was stated earlier in the manuscript that BC was below the detection limit (line 21-23). Therefore, is it really valid to assume there is BC in the sample? Based on the derived refractive index,

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what is the concentration of BC? If there is BC in the sample, is it really appropriate to assume a homogeneous sphere (or is it even a tarball in the first place)? Is equation 3 even valid for the case of the heterogeneously internally mixed BrC/BC particle? Please show error bars on Fig 4a so the reader can see how significant the refractive index is at the longer wavelengths.

P 6287, line 18: I wouldn't call the refractive index "data" since it was calculated. Please change this.

P 6288, line 1-3: Were there any measurements of inorganic species (such as nitrates or ammonium)? How might these inorganic species affect the optical properties? This may also be related to the statement that the derived refractive index is a function of time (p 6287, line 11-12).

Table 2: I think it would be useful to include the SSA in the table.

Table 2: The real part of the refractive index seems very high (almost 1.9). How do these values compare to literature values of organic carbon (or "brown" carbon, HULIS, tarballs, etc)? These values really make me question how accurate the retrievals are. How sensitive are the results presented in this paper to the refractive index? How sensitive is the refractive index to uncertainties in size distribution, shape and inhomogeneities?

Figure 2 and related discussion: Takahama et al has asserted that tarballs may contain Fe. Was this observed? Would this be expected (from elements in soil?)?. (Takahama et al., 2008)

Figure 3: Since absorption and scattering are more intimately related to surface area, I suggest showing the surface area distributions.

Figure 4: All text on this figure is too small. Why is there no data for Pine duff 1 in figure 4a? Y-errorbars should be shown.

References:

Bond, T. C., and Bergstrom, R. W.: Light absorption by carbonaceous particles: An investigative review, *Aerosol Sci. Technol.*, 40, 27-67, 2006. Capes, G., Johnson, B., McFiggans, G., Williams, P. I., Haywood, J., and Coe, H.: Aging of biomass burning aerosols over West Africa: Aircraft measurements of chemical composition, microphysical properties, and emission ratios, *J. Geophys. Res.-Atmos.*, 113, -, 2008. Takahama, S., Gilardoni, S., and Russell, L. M.: Single-particle oxidation state and morphology of atmospheric iron aerosols, *J. Geophys. Res.-Atmos.*, 113, doi:10.1029/2008JD009810, 2008.

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