

Interactive comment on “Short wave radiative impact of liquid-liquid phase separation in “Brown Carbon” aerosols” by Mehrnoush M. Fard et al.

Anonymous Referee #2

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This is a very interesting and useful manuscript that should be suitable for publication in ACP after the flawed discussion of direct radiative forcing in section 3 (Atmospheric Implications) has been redone correctly. Detailed comments are as follows:

P1L29-31: Add the reference of Bond et al. (2013) and change the sentence also referring to the surface albedo (e.g., Chylek and Wong, 1995) and changing the unclear wording “may also contribute to warming”. Does this mean that in addition to cooling they also cause warming?

P3L9: “Brown Carbon is referring to the light-absorbing fraction of the organic carbon that has a wavelength dependent absorptivity.” This is a very poor definition of BrC because the key definition is that the imaginary part of the refractive index (not the absorptivity) is wavelength dependent and increases toward shorter wavelengths (e.g.,

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Moosmuller et al., 2011).

P3L13: “We use simple volume mixing...”. This needs some explanation of effective medium theories, why the volume mixing rule was chosen, and what its accuracy is. A good starting point would be Chylek et al. (1988).

P7 Fig. 2: (1) Explain exactly what is meant here with random location and how it is realized computationally; (2) Give the complex refractive index both for the particle and the inclusion here and elsewhere; (3) “100 nm particle”: Does “100 nm” refer to particle radius, diameter, circumference or something else; please state explicitly here and elsewhere!

P12L22 – P17L12: “3 Atmospheric Implications”. This section is flawed and in need of major revision! The reference Charlson et al. (1991) discusses only radiative forcing by non-absorbing (i.e., sulfate) aerosols; the reference Nemesure and Schwartz (1998) is in the “grey” literature and should be replaced with a peer-reviewed reference such as Chylek and Wong (1995). In addition, the authors pick the wrong equation from Nemesure and Schwartz (1998) that doesn’t account for the albedo of the underlying surface. In reality, the radiative forcing in the optically thin aerosol layer case depends on one extensive aerosol parameter (AOD), two intensive aerosol parameters (SSA and upscatter fraction), and the albedo of the underlying surface or scene. The equation for this can be found in Nemesure and Schwartz (1998) p. 532, left column just above the right column header “Results” or in the peer reviewed literature (Chylek and Wong; 1995; eq. 8), with further discussion of validity and assumptions to be found in Hassan et al. (2015), Moosmuller and Ogren (2017), and Moosmuller and Sorensen (2018). Of specific interest would be to plot the ratio (LLPS/homogeneous) of the dominating intensive aerosol forcing parameter SSA as function of particle diameter such as done in Fig. 6 for Qscat and Qabs.

References

Bond, T. C., S. J. Doherty, D. W. Fahey, P. M. Forster, T. Berntsen, B. J. DeAngelo, M. G.

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Flanner, S. Ghan, B. Kärcher, D. Koch, S. Kinne, Y. Kondo, P. K. Quinn, M. C. Sarofim, M. G. Schultz, M. Schulz, C. Venkataraman, H. Zhang, S. Zhang, N. Bellouin, S. K. Guttikunda, P. K. Hopke, M. Z. Jacobson, J. W. Kaiser, Z. Klimont, U. Lohmann, J. P. Schwarz, D. Shindell, T. Storelvmo, S. G. Warren, and C. S. Zender (2013). Bounding the Role of Black Carbon in the Climate System: A Scientific Assessment. *J. Geophys. Res.*, 118, 5380-5552.

Chylek, P., V. Srivastava, R. G. Pinnick, and R. T. Wang (1988). Scattering of Electromagnetic Waves by Composite Spherical Particles: Experiment and Effective Medium Approximations. *Appl. Opt.*, 27, 2396-2404.

Chylek, P., and J. Wong (1995). Effect of Absorbing Aerosol on Global Radiation Budget. *Geophys. Res. Lett.*, 22, 929-931.

Hassan, T., H. Moosmuller, and C. E. Chung (2015). Coefficients of an Analytical Aerosol Forcing Equation Determined with a Monte-Carlo Radiation Model. *J. Quant. Spectrosc. Radiat. Transfer*, 164, 129-136.

Moosmuller, H., R. K. Chakrabarty, K. M. Ehlers, and W. P. Arnott (2011). Absorption Ångström Coefficient, Brown Carbon, and Aerosols: Basic Concepts, Bulk Matter, and Spherical Particles. *Atmos. Chem. Phys.*, 11, 1217-1225.

Moosmuller, H., and J. A. Ogren (2017). Parameterization of the Aerosol Up-scatter Fraction as Function of the Backscatter Fraction and Their Relationships to the Asymmetry Parameter for Radiative Transfer Calculations. *Atmosphere*, 8, doi:10.3390/atmos8080133.

Moosmuller, H., and C. M. Sorensen (2018). Small and Large Particle Limits of Single Scattering Albedo for Homogeneous, Spherical Particles. *J. Quant. Spectrosc. Radiat. Transfer*, 204, 250-255.

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