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Interactive comment

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"Biomass-burning-derived particles from a wide variety of fuels: Part 2: Effects of photochemical aging on particle optical and chemical properties" by Christopher D. Cappa et al.

Anonymous Referee #1

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This manuscript by Cappa et al. presents results of the evolution of biomass-burning smoke due to photochemical aging in a mini chamber. The study features a wide range of fuel types and combustion conditions and derives general trends / conclusions from averaging different burns grouped into 6 categories based on combustion conditions and consequent aerosol optical properties. The averaged experimental results are then used to develop a model for SOA formation and processing and the associated evolution of brown carbon optical properties. The major strengths of this manuscript are the 1) analysis of averages of large data sets which enables the derivation of gen-

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eral trends based on the average behavior (this is not possible to achieve from a small number of burns due to the high variability in combustion conditions), and 2) measurement/modeling analysis that enabled decoupling the effects of vapor-to-particle conversion versus heterogeneous oxidation on the evolution of optical properties (granted by using some simplifying assumptions, which is justified given the high complexity of the problem).

I believe this paper presents interesting and very useful insights on the evolution of biomass-burning smoke, and the data analysis and presentation are of high quality. I have only a few minor comments.

General comments:

- 1) Section 3.3: The discrepancy in observed evolution in BrC absorptivity between the experiments here and ambient observations is interesting. One possible explanation is related to the molecular-size dependence of absorptivity, solubility, and susceptibility to photobleaching (Saleh [1] and references within). Wong et al. [2] showed that larger BrC molecules are less susceptible to photobleaching. These larger BrC molecules are more absorptive [3,4] and less soluble in water/methanol [5]. It is plausible that the ambient studies referenced in Section 3.3 (that used solvents) overestimated the effect of photobleaching compared to this study, which relies on airborne particle measurements. I suggest including a discussion along these lines in Section 3.3.
- 2) Section 3.2.6: It is not clear that the analysis in Line 766-786 is useful for the discussion of the results. The authors argue that the May et al. volatility distributions are not expected to provide good predictions for the concentrations in their experiments, so why do the calculations / comparison to begin with? I suggest taking this part out.

Specific comments:

1) Line 95: Feng et al. 2013 and Wang et al. 2014 are not appropriate here, as these studies did not address BB evolution / photochemical aging.

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- 2) Line 218-223: in McClure et al., MAC_BC_pure values were obtained by extrapolating MAC vs OA/BC fits to zero. This should be mentioned here.
- 3) Line 227-229: Can this statement be more quantitative?
- 4) Figure 1: panel (g) is not described in the caption and the corresponding plots for individual burns are not given in the SI.
- 5) Line 455 (Reaction scheme): I might be missing something, but shouldn't the reaction be: NMOG + OH -> SOA + NMOG_MG, where SOA = alpha * NMOG (since by definition, the yield is SOA/NMOG) and NMOG_MG = (1- alpha) * NMOG?
- 6) Line 546: specify wavelength for MAC.
- 7) Line 768-771: This part is a bit confusing. Are you saying that using May et al. volatility distribution, OA/BC decreases from 19.2 to 11.2 assuming a dilution factor of 33 (i.e. assuming the particles on the walls do not contribute to particle-gas equilibrium) and to 10.5 with a dilution factor of 21? One should expect lower dilution ratio to result in less decrease in OA/BC, no?

Some typos:

Line 68: particulate.

Line 217: in equation 2, MAC_BC_ref is inconsistent with the text (MAC_BC_pure).

Line 437: units of rate coefficient (cm⁻¹ should be s⁻¹).

Line 450: delete "occurs"

References:

- 1. Saleh R. From Measurements to Models: Toward Accurate Representation of Brown Carbon in Climate Calculations. Curr Pollut Reports. 2020; https://doi.org/10.1007/s40726-020-00139-3
- 2. Wong JPS, Tsagkaraki M, Tsiodra I, Mihalopoulos N, Violaki K, Kanakidou M, et

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- al. Atmospheric evolution of molecular-weight-separated brown carbon from biomass burning. Atmos Chem Phys 2019;19:7319–34.
- 3. Saleh R, Cheng Z, Atwi K. The Brown–Black Continuum of Light-Absorbing Combustion Aerosols. Environ Sci Technol Lett. 2018; 5:508–13.
- 4. Di Lorenzo RA, Washenfelder RA, Attwood AR, Guo H, Xu L, Ng NL, et al. Molecular-Size-Separated Brown Carbon Absorption for Biomass-Burning Aerosol at Multiple Field Sites. Environ Sci Technol. 2017;51:3128–37.
- 5. Corbin JC, Czech H, Massabò D, de Mongeot FB, Jakobi G, Liu F, et al. Infraredabsorbing carbonaceous tar can dominate light absorption by marine-engine exhaust. npj Clim Atmos Sci. 2019; 2:12. https://doi.org/10.1038/s41612-019-0069-5

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