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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Experimentally measured morphology of biomass burning aerosol and its impacts on CCN ability" *by* M. Giordano et al.

Anonymous Referee #1

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The manuscript, "Experimentally measured morphology of biomass burning aerosol and its impacts on CCN ability" investigates morphology of fresh and aged biomass burning aerosol in terms of the dynamic shape factor (χ) and the fractal-like dimension (Df'). The hygroscopicity parameter, κ, of biomass burning aerosol (flaming regime, chamise and mazanita) was accurately determined by accounting for χ. The authors suggest that the observed range of χ (1.06 $\sim$ 1.42) leads to significant overestimation of particle volume, and hence underestimation of κ.

Although it is reasonably expected that some fresh biomass burning aerosol can be non-spherical (especially in the flaming regime), quantitative analysis of particle shape is rarely available and thus the analysis employed in this study is a useful addition of our understanding of physical/chemical transformation of biomass burning aerosol in the

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atmosphere; however, the discussion needs significant improvement. I recommend the manuscript for publication in ACP after the following concerns have been addressed.

Major comments:

1. The shape effect observed in this study seems to be nearly an upper limit since the experiments focus on the flaming regime, and also evaporation during TEM analysis (leaving behind fractal-like backbones) can enhance shape factor as mentioned in section 4.2. This point needs to be more clearly mentioned. Since aged particles are reasonably spherical, the focus of this study is biomass burning aerosol "near source". It appears that the relevance to "near source" is mentioned only in conclusion. This clarification should be a part of introduction or another early part of the paper.

2. The results and discussion on dark (fresh) and aging experiment should be more clearly separated. If I understood correctly, the TEM was operated during the beginning and the end of the experiment (Page 12565, Line 2), but only results during the beginning was used (Page 12568, Line 5: "The TEM was not used in this section because saturation of the TEM grids required extensive time. . ."). Since the shape factor, χ, was only available for the dark condition, relevance of sections 4.3 and 4.4 are limited to fresh aerosol. Currently, sections 4.1, 4.3, and 4.4 are about fresh aerosol; section 4.2 is about aged aerosol. In my opinion, this organization makes the discussion hard to follow.

3. A very relevant paper, Martin et al. (2013) is not cited. Martin et al. carried out chamber experiments of biomass burning aerosol and observed collapsing of non-spherical aerosol when exposed to a high relative humidity, resulting in underestimation of κ. Similarities and differences between this study and Martin et al. should be discussed.

4. Petters et al. (2009a) is cited as an example of in-situ sampling of possibly non-spherical CCN. However, Petters et al. preconditioned particles by humidifying the sample flow to >95% RH and then drying to RH < 5%, in order to collapse particles prior to the size-resolved CCN measurement (section 2.3 in Petters et al.). This point

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must be clarified.

5. Section 4.4: Sensitivity of κ to χ is shown by applying χ = 1 $\sim$ 1.5 to two biomass burning aerosol (Black Spruce and Sage Brush) observed by Petters et al. (2009a). However, as mentioned above, it is very unlikely that χ of particles observed by Petters et al. is as high as 1.5 because of the particle preconditioning step. Therefore, the calculated maximum κ of 1.1 (even higher than pure KCl) (Carrico et al., 2010) is not reasonable. For the purpose of sensitivity analysis, I think a systematic analysis of synthetic data would be better than using Petters et al. with an unreasonable assumption ($\chi \sim 1.5$). When slip correction factors are ignored, the impact of (constant) χ on κ is simply χ^3 (1.50^3 = 3.38), which reasonably explains the reported change of κ from 0.07 to 0.24 and 0.33 to 1.1 (factor of 3.3 $\sim$ 3.4). I believe it would be much more informative to illustrate to what extent slip correction matters in the impact of constant χ on κ.

6. Page 12572, Line 18: Are you suggesting BC may be sparingly soluble? That seems unreasonable. Please add more description. You could just say sparingly soluble species could contribute to deviation from Köhler theory. Size-dependent chemical composition can also contribute to changing κ as a function of size (or sc).

7. Conclusions: "SOA condenses on the fractal particle and the volume changes (measured through electrical mobility) are small compared to changes in particle mass." The data is not shown in this paper. Please provide relevant data such as time-series of effective density and particle volume (possibly as supplement).

8. Page 12573 Line 25-28: How can you make this statement about the impact of chemical and physical transformation on non-ideality since χ is only available for fresh aerosol?

9. I think it is important to emphasize that when material density (rho_m) is known (e.g., pure inorganic salt), measurements of mobility diameter (Dm) and the effective density (rho_eff) using the APM system is enough for acquiring accurate Dve, from the

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following equation: mp=π/6* rho_m * Dveˆ3=π/6 * rho_eff * Dmˆ3 which was utilized by Kuwata and Kondo (2009). The uniqueness of this study is experimental observation of χ (as a function of size) to compensate for the unknown rho_m of biomass burning aerosol.

Minor comments:

Page 12556, Line 9: “microscopy” should be “microscope” to be consistent with aerosol particle mass analyzer and scanning mobility particle sizer (instrument, not technique)

Page 12557, Line 14: “Particles formed . . . non-uniform” is an unnecessarily repetitive sentence.

Eq. (1), Page 12558, Line 21: sc and Sc are mistaken (sc = Sc – 1). It can be lnSc or sc. Note lnSc = ln(1+sc) $\sim$ sc

Page. 12559, Line 10: κ-Köhler theory does not necessarily assume ideal solution. κ includes the effect of non-ideality typically in the form of osmotic coefficient (Φ) or van’t Hoff factor (i) (e.g., Rose et al., 2008). When ideality is assumed, κ simply follows Raoult’s law and previous studies presented it as κ_Raoult (Petters et al., 2009b).

Page. 12559, Line 18: “. . .not experimentally measured, especially as a function of particle aging.” needs to be rephrased. Martin et al. (2013) experimentally measured hygroscopicity of non-spherical biomass burning aerosol as a function of particle aging, although they did not acquire χ.

For clarity, please add description of the relationship between effective density and material density, such as mp=π/6* rho_m * Dveˆ3=π/6 * rho_eff * Dmˆ3

Page 12560, Eq. (2): Please show what formula was used for the Cunningham slip correction factor.

Page 12562, Line 14: There are two “fractal”. Aren’t they “fractal-like” since the APM was used?

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Page 12563, Line 26: “similar”, in terms of what?

Page 12566, Line 12: I don’t understand how primary particle diameter was calculated.

Page 12567, Line 9 - 10: There are duplicate sentences. “Over 80% of. . .”.

Page 12567, Line 19: “. . .overestimation” of primary particle size? Please specify.

Page 12568, Line 1: The impact of χ (=1.06 $\sim$ 1.42) on particle volume should not be linear (1.06 $\sim$ 1.42), but approximately cubed, although slip correction factor may also matter.

Page 12568, Line 8: Figure 4 $\rightarrow$ Figure 5

Page 12568, Line 11: please add reference for “not quite spherical” ammonium sulfate. E.g., (Kuwata and Kondo, 2009)

Page 12569, Line 14: diameter cubed is not equal to the volume. There is π/6.

Page 12571, Line 4-5: How can shape factor contribute to a lower κ of the spherical reconstituted particles? Isn’t that the opposite direction?

Page 12572, Line 8-10: “The data shown is data from the chamise experiments shown in Fig. 7”. Are triangles in Fig. 9 actual measurements or interpolation/extrapolation of Ddˆ-3/2 curve fit? It appears to be the latter since there are four sc settings in Fig. 7 and eight in Fig. 9. Please clarify.

Page 12572, Line 13-16: The reference to mineral dust seems to be out of place since the mechanisms are likely to be very different (adsorption vs. absorption).

Page 12573, Line 6: “to ensure a mobility diameter derived volume”. I don’t understand this. Do you mean to ensure accurate determination of volume-equivalent diameter?

Page 12573, Line 15: “Biomass burning derived aerosol experience a decrease in hygroscopicity with aging” is a too general statement. You can only say this for those two fuels used in this study.

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Page 12573, Line 16: How can morphological changes (approaching sphere) explain decrease in hygroscopicity? It should increase the apparent hygroscopicity because non-sphericity leads to underprediction of κ.

Fig. 4 Please add fuel-type and burn-regime (Flaming)

Fig. 7 Does this data include both fresh and aged aerosol? Please specify. If aged aerosol is included, you cannot determine volume equivalent diameter since χ is only measured for fresh aerosol.

Reference

Carrico, C. M., Petters, M. D., Kreidenweis, S. M., Sullivan, A. P., McMeeking, G. R., Levin, E. J. T., Engling, G., Malm, W. C., and Collett Jr, J. L.: Water uptake and chemical composition of fresh aerosols generated in open burning of biomass, Atmos. Chem. Phys., 10, 5165-5178, 2010.

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Martin, M., Tritscher, T., Jurányi, Z., Heringa, M. F., Sierau, B., Weingartner, E., Chirico, R., Gysel, M., Prévôt, A. S. H., Baltensperger, U., and Lohmann, U.: Hygroscopic properties of fresh and aged wood burning particles, Journal of Aerosol Science, 56, 15-29, 2013.

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