

Response to comments by referee#1:

The authors basically answered all the reviewer's concerns. However, there are some issues or minor concerns that the authors should be aware of:

Response: We would like to thank the referee for the constructive comments and suggestions that helped us improve our manuscript. We have implemented changes based on these comments in the revised manuscript. Please find our point-by-point response below. We repeat the specific points raised by the reviewer in italic font, followed by our response. The pages numbers and lines mentioned are with respect to the acp-2022-544-manuscript-version2. PDF that uploaded on 16.02.2023.

1. A thoroughly check of the manuscript is needed since there are still lot of typos, ill-sentences in the revision.

Response: We have checked and revised all typos and ill-sentences throughout the manuscript.

1) Line 51: quantification of the biomass burning

Response: Many thanks. We have revised this sentence and now they read as:

Page 3 line 48-52: “Levoglucosan aerosol nanoparticles have attracted increasing interest in recent years (Simoneit et al., 1999; Mochida and Kawamura, 2004; Mikhailov et al., 2009; Elias et al., 2010; Lei et al., 2014, 2018; Bhattarai et al., 2019) and is considered as an ideal tracer for characterization and quantification of the biomass burning (Fraser and Lakshmanan, 2000).”

2) L95-98, in the sentence, what is the role of the where clause? It is hard to understand.

Response: Many thanks. We have revised these sentences and now they read as:

Page 5 line 93-98: “Thermodynamic models rely on the concentration-dependent thermodynamic data (such as water activity, liquid-vapor interfacial energy), which are often derived from measurements of large droplet and/or bulk solution (Tang and Munkelwitz, 1994; Tang 1996; Pruppacher and Klett, 1997; Clegg et al., 1998). Nanodroplets can become more highly supersaturated and thus reaching higher solute concentration compared to bulk solution, which makes it difficult for models to predict its hygroscopicity.”

3) L102, *understand new particle formation*

Response: Many thanks. We have revised this sentence and now they read as:

Page 5 line 101-103: “This will further help us to understand the new particle formation, transportation, and their interactions with water molecules.”

4) L61, *HTDMA appears in the main text for the first time, and full name should be spelled.*

Response: Many thanks. We have provided the full name of HTDMA, Hygroscopicity Tandem Differential Mobility Analyzer.

5) L124, *high solution concentrations*

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 6 line 124-127: “In order to avoid blocking the 25- μm capillary tube in the electrospray with high concentration solution, the aerosol nanoparticles with diameters of 60-100 and 20 nm are generated by an atomizer with a 0.05 and 0.01 wt % organic solution (i.e., levoglucosan and D-glucose), respectively.”

6) L237, *it is apparently an ill-sentence, this is not a dependent sentence, and are should be replaced with of*

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 11-12 line 235-239: “For example, the hygroscopic growth factors of levoglucosan nanoparticles at 80 % RH, 87 % RH are 1.16, 1.23, respectively, in the deliquescence mode, very close to the corresponding values in the efflorescence mode at the same RH (shown in Fig. S1), suggesting that growing and shrinking of particles are in equilibrium.”

7) *I think it is ok to say 20 nm particles instead of 20-nm particles, check throughout the whole text*

Response: Many thanks. We have revised in the whole manuscript and now they read as:

Page 12 line 253-254: “E.g., a slight difference in hygroscopic growth factor between 100 and 20 nm levoglucosan nanoparticles is ~ 0.02 at 88 % RH.”

8) L261, *with diameters of 100, 60*

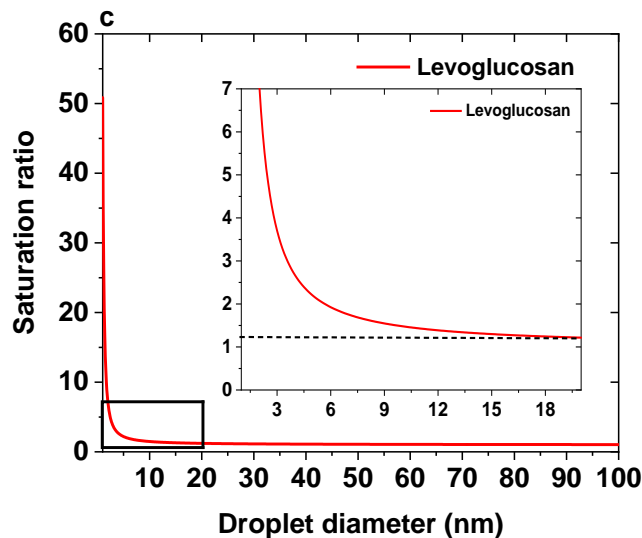
Response: Many thanks. We have revised in the whole manuscript and now they read as:

Page 12-13 line 260-263: “For example, as shown in Fig. 4a, for levoglucosan nanoparticles with diameters of 100, 60, and 20 nm, the thermodynamic equilibrium model (E-AIM (standard UNIFAC)) shows a weak size dependence of the growth factors at low RH but a strong size dependence at RH above 70 %.”

9) L286, *how can you image a particle with 0 nm diameter?*

Response: Many thanks. We have calculated the saturation ratio of levoglucosan particles against droplet diameters from 1 to 100 nm and revised in the following Fig. 5c and sentence and now they read as:

Page 14 line 285-286: “Figure 5c shows the vapor saturation ratio of levoglucosan as nanodroplet diameter increases from 1 to 100 nm.”



10) L290, *why use thus here, there is no cause relation according to the context*

Response: Many thanks. We have deleted “thus” in the sentence.

11) L298, *evaporation is not countable*

Response: Many thanks. We have revised in the following sentences and now they read as:

Page 14 line 297-298: “Therefore, there is the obvious partial levoglucosan evaporation from DMA1 to DMA2 within several seconds.”

12) L324, where clause, there is nowhere that can be referred to.

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 14 line 322-324: “Estillore et al. (2017) observed a slightly amorphous structure of D-glucose particles under ambient conditions using an atomic force microscopy and D-glucose particles grow through gradual water uptake.”

13) L363, an awkward sentence, I think it should be rewritten

Response: Many thanks. We have revised in the following sentence and now they read as:

Page 17 line 363-364: “The ideal solution theory is used to predict the hygroscopic curve of D-glucose nanoparticles with diameters of 6-100 nm, shown in Fig. 9b and Fig. S3.”

2. L337-340, I still don't get it where RH matters here and why it is 80%?

Response: Many thanks. As shown in the following Fig.8a, at about 80 % RH, we observed that an obvious difference in hygroscopic growth factor of D-glucose nanoparticles in the size range from 6 to 100 nm, while a small difference in the hygroscopic growth factor is observed at RH below about 80 %. The reason for the obvious difference in water absorption at high RH ($\geq \sim 80\%$ RH) still needs to be investigated.

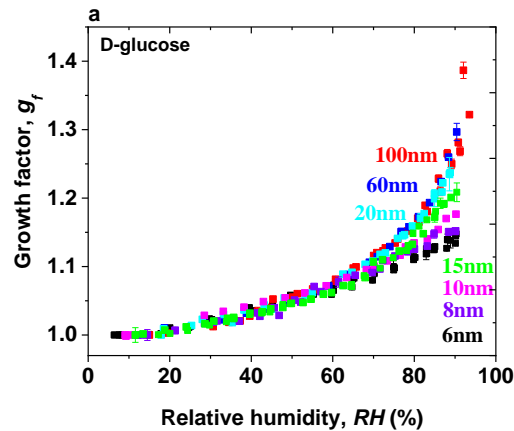


Figure 8: (a) Hygroscopic diameter growth factor (G_f) of D-glucose nanoparticles with dry diameters of 100 nm (red square), 60 nm (blue square), 20 nm (cyan square), 15 nm (green square), 10 nm (pink square), 8 nm (royal square), and 6 nm (black square), respectively.

3. L370-371, what do you mean “the unfavorable assumption of ideal solution theory”?

Response: Many thanks. For ideal solution, water activity of liquid droplets can be simply estimated from the mole fraction of water. With from 20 down to 6 nm, D-glucose nanodroplets can be highly supersaturated, and the water activity is not equal to mole fraction of water. Thus, with the assumption of idea solution, the model failed to predict the observed growth factors of 6-nm D-glucose nanoparticles at RH above 30 %.

We have revised this sentence and now they read as:

Page 17 line 370-372: “For ideal solution, water activity of liquid droplets can be simply estimated from the mole fraction of water. With from 20 down to 6 nm, D-glucose nanodroplets can be highly supersaturated, and the water activity is not equal to mole fraction of water. Thus, with the assumption of idea solution, the model failed to predict the observed growth factors of 6-nm D-glucose nanoparticles at RH above 30 %.”