

Atmos. Chem. Phys. Discuss., referee comment RC2 https://doi.org/10.5194/acp-2022-581-RC2, 2022 © Author(s) 2022. This work is distributed under the Creative Commons Attribution 4.0 License.

Comment on acp-2022-581

Anonymous Referee #2

Referee comment on "Physicochemical properties of charcoal aerosols derived from biomass pyrolysis affect their ice-nucleating abilities at cirrus and mixed-phase cloud conditions" by Fabian Mahrt et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-581-RC2, 2022

The authors present measurements of the ice nucleation ability (immersion and pore condensation freezing modes), water sorption profiles, and limited chemical composition measurements using a laser ablation mass spec of charcoal-derived particles from one grass and one wood charcoal sample which they resuspend. The methods used are sound, as are the conclusions made. Connecting this work to atmospheric conditions and possible effects is a bit more challenging due to the nature of the charcoal samples used and the limited chemical analysis performed. Still this work is a notable contribution to our understanding of the possible particle types emitted from biomass burning and their effects on cloud microphysics. It should be of interest to the readers of ACP and suitable for publication after some aspects are better clarified and discussed in the manuscript. Since pyrolysis-derived charcoal particles are studied here I do not think using "biomass burning" in the title is appropriate, however. This could be a bit misleading since combustion-derived particles were not studied here and this is a very different process from pyrolysis (which is not directly relevant to actual biomass burning that emits particles into the atmosphere).

The charcoal particles were produced by pyrolysis under an N2 environment. Clearly this will produce very different particle properties and compositions that occurs in actual wildfires and prescribed burns that proceed over a large range of combustion conditions in oxygen-containing air. There was not much discussion of how the charcoal particles studied here might relate to components of biomass-burning particles that are actually emitted to the atmosphere. The authors should really add a discussion of this so that it is clear that their objective is to focus specifically on the properties of charcoal particles that were produced through pyrolysis and not combustion processes. This is fine and provided some valuable insights, it will just make it difficult to generalize to atmospheric conditions and properties.

Along with this, what contribution organic aerosol components make to the charcoal

particle studied should be better clarified. In combustion-derived BBA there is a variable and significant amount of organic aerosol material that spans a range of volatility down to quite low saturation vapor pressures. This often includes tar like material that produces tar ball particles that can also mix with and coat other BBA components. I imagine these organics could also fill or conceal the pores needed for pore condensation freezing. Since pyrolysis and not combustion-produced particles were studied here, and a N2 purge gas was used, it seems that there is little contribution from such organic aerosol components here? This is important to clarify, and again makes the charcoal particle studied here rather poor mimics of actual combustion-derived BBA.

Related, Jahl et al. (2021) reported the enhancement of INA in BBA from grass fuels through simulated atmospheric aging. This was attributed to the removal of organic coatings that concealed the mineral ice active surface sites. I imagine similar processes likely occur that alter the availability or properties of the pores involved in PCF also. While the pyrolysis charcoal particles studied here are too simplistic to provide this sort of behavior, this is why it is important to discuss the contribution of organic carbon in these particles, and what role this might play in the artificial charcoal particle studied here, and realistic combustion-derived BBA.

In the ATOFMS results I did not understand why m/z + 26 and +27 were attributed to markers/fragments of nitrogen oxides. You often do get an NO+ fragment at m/z + 30. What do the authors propose are the ions at +26 and +27?

It is not clear what the purpose of the ATOFMS analysis is. What about the particles is being learned here that informs their ice nucleation or water uptake properties? This section did not add much to the paper as it is currently presented.

The authors could also try to apply the OC/EC mass ratio estimates that have been demonstrated using laser ablation mass spec analysis, e.g.:

Spencer, M. T., & Prather, K. A. (2006). Using ATOFMS to determine OC/EC mass fractions in particles. *Aerosol Science and Technology*, *40*(8), 585–594. https://doi.org/10.1080/02786820600729138

Ahern, A. T., Subramanian, R., Saliba, G., Lipsky, E. M., Donahue, N. M., & Sullivan, R. C. (2016). Effect of secondary organic aerosol coating thickness on the real-time detection and characterization of biomass-burning soot by two particle mass spectrometers. *Atmospheric Measurement Techniques*, *9*(12), 6117–6137. https://doi.org/10.5194/amt-9-6117-2016

It is a bit odd that when discussing the ions likely derived from mineral species that the authors did not draw a connection to the recent idea that combustion-derived minerals are the main source of ice nucleants in some types of biomass-burning aerosol first presented by Jahn et al. (2020) and then supported by the field measurements of Adachi et al. (2022).

The discussion of the lower INA observed for these pyrolysis charcoal particles compared to some other types of BBA reported such as by Jahn et al. was confusing. The authors propose that this is because of lower particle surface area in the droplets studied. But they use the widely-used n _s metric that normalizes to surface area, so such differences should be largely normalized for. It seems far more likely that as these pyrolysis charcoal particles were produced through a very different process that combustion derived-BBA is that the resulting particles and their INA/n_s are just very different. Jahn et al. (and Adachi et al.) attributed the higher INA observed in their BBA to mineral-containing particles. The pyrolysis method may not have produced the right conditions for volatilization and then recondensation of much of the inorganic/mineral components. Charcoal particles produced by pyrolysis will be different in many important ways from combustion particles. This is the aspect of this work that I find is the least well justified. It seems much more likely that a lower INA/n s was observed for these charcoal particles simply because they are entirely different from combustion BBA studied in the other reports. This is why I think the manuscript title should be changed as biomass burning particles were not actually studied here.