Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-781-RC2, 2020

Atmospheric Chemistry and Physics Discussions

Interactive comment on "The effect of biological particles and their ageing processes on aerosol radiative properties: Model sensitivity studies" *by* Minghui Zhang et al.

Anonymous Referee #2

Received and published: 19 November 2020

This work explores the radiative effects of biological aerosol particles (BAPs). The authors conduct a literature review on the physicochemical properties associated with scattering and absorption of radiation, cloud condensation nuclei (CCN) activation and ice nucleation efficiency of BAPs. From this they establish plausible ranges for different BAPs properties, then perform several sensitivity studies to roughly assess the possible impacts on radiation and cloud evolution, hence on climate. This is a well written paper that lies within the scope of ACP. However, it is also very speculative. It is not clear that enough data has been reported to establish a possible impact. The sensitivity studies are also conducted in a very idealized and simplified way, particularly those related to cloud formation. Thus, some clarification of the approach taken, as well as

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Discussion paper

the considerable limitations of this study, are required before it can be accepted for publication.

General Comments:

The authors carry out a somehow extensive literature review to select a plausible range of parameters to carry out sensitivity studies. This is commendable; however, the process studies sprung from it may be too limited and too idealized to be meaningful regarding the effect of BAPs on radiative forcing. For example, a parcel model is not at all appropriate to make conclusions on the prevalence of the Bergeron-Findeisen (BF) process. This is further discussed below. The process studies thus lead to somehow obvious conclusions, which are not necessarily unique to BAPs and that are already well-known, i.e., higher kappa value leads to easier CCN activation, lower contact angle to more efficient ice nucleation, in a parcel model ice grows at the expense of liquid, higher refractive index leads to enhance absorption, and so on. Thus the authors must modify the language with a honest and thorough assessment of the limitations of their study and also emphasize differences/similarities with the typical behavior of other aerosols.

Detailed comments:

Line 11. Are biological fragments considered here?

Line 37. Delete “the”

Line 41. Maybe “in the urban area of Mainz” is more appropriate.

Line 68. In Figure 1 would absorption of solar radiation lead to a semi-direct effect?

Line 165. Must be “agglomerates”

Line 240. It is not clear what the maximum frozen fraction means here. If the temperature is lowered to -40 C the bacteria won’t freeze at all? Why is -10 C the temperature of choice?

Line 244. This is factually wrong. All real materials show stochastic behavior during ice nucleation. Please rephrase.

Line 250. The application of the contact angle approach to ice nucleation in biological materials is fraught with problems, since all the assumptions of classical nucleation theory break, and depends strongly on the values selected for other very uncertain parameters like for example the ice-liquid interfacial tension and the activation energy. Please add an explanation on the limitations of describing ice nucleation in biological materials.

Line 258. INAS is obtained by fitting freezing experiments neglecting the time dependency of ice nucleation. Please rephrase. I would suggest the authors refrain from discussing deterministic vs stochastic behavior since it is distracting and not at all clear what they mean, particularly for BAPs.

Line 273. Are these changes due to denaturation or are they reversible?

Line 286. Is there a reason to consider BAPs externally-mixed and monodisperse?

Line 301. What are the properties of the "other" aerosol. Is there any sensitivity of the results to this assumption?

Line 311. If the BAPs freeze by immersion, shouldn't they be inside the droplets? Are the results sensitive to Nother?

Line 313. This is a crude approximation that only works to make an assessment on droplet/ice formation, but would be very misleading to estimate LWC and IWC. Once ice is formed a whole set of other microphysical processes rapidly take place. Please justify why this approach is used at all.

Line 330. This "Nother" is different from the "Nother" of line 309, which is also different to the one in line 301.

Line 438. All of these values change strongly with location, so it is not clear why this

estimate is not given with a range of uncertainty, down in line 450.

Line 459. This would only be true if BAPs were uniform in the globe and isolated from other aerosols.

Line 479. There is certainly not data to support this “independence” assertion. The authors could probably make this assumption but clarify that it is in the absence of better data.

Line 509. I am not sure what is shown here. This caption needs more information, Table 2 does not even say what Senv or Sc are.

Line 531. I don’t think this is a buffer effect, or at least explain what that means in this context.

Line 561. This conclusion is short-sighted. D influences droplet activation hence where freezing could occur. Mixed-phase clouds are CCN limited as well, so the effect may not be negligible.

Line 570. Please explain how the authors pin the BF effect to a particular Delta_T (also what Delta_T means). Is the T shift related to a later onset of freezing?

Line 574. No, this is not clear at all. Early freezing may result in early scavenging of available BAP and actually limiting instead of enhancing BF processes. There is a myriad of other things that can negate the onset of BF process, none of which can be represented in a parcel model: high subgrid scale vertical velocity, the presence of other efficient ice nucleating particles (for example feldspards can freeze at very high T as well), preferential spatial concentration of liquid and ice particles, to name a few. I would accept a much more cautious language like for example, “has the potential to affect the BF process” followed by a list of all the things that need to be addressed before this conclusion can be asserted with any degree of accuracy.

Line 584. Please show here where the BF process is initiated.

Line 596. Figure 11 must be removed. To start it is confusing since clearly the different aging processes affect more than one variable at a time. More fundamentally it presents a misleading, “final” assessment of something that is highly uncertain. The data is still too scarce and the studies way too idealized to support this figure.

Line 615. What about the semi-direct effect?

Line 630. See comment on Line 479.

Line 636. See comment on Line 240.

Line 654. This is speculation, since the authors do not perform any studies on cell generation.

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