We thank the referee for their positive and constructive comments on our manuscript. Below we provide a point-by-point response to comments. The comments and suggestions are in italics. Our responses and revisions are in plain font; responses are in blue. The original manuscript text is in black. Additions to the manuscript text are in red.

The major changes to the manuscript include the change of the title as both referees commented on the limitations of our model studies in terms of global implications. We also made it clearer at several places in the manuscript that the role of PBAPs in the atmosphere for the global aerosol direct and indirect effects may be limited due to their small number concentration. However, detailed knowledge of PBAP properties that affect their interaction with radiation and water vapor is essential to properly describe their transport, dispersion and lifetime in the atmosphere, which affects the biodiversity.

Consequently, the new title is

Sensitivities to biological aerosol particle properties and ageing processes: Potential implications for aerosol-cloud interactions and optical properties

Referee Comment:

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 #1 Received and published: 28 October 2020

General comments: Overall, this paper is a useful study that investigates the relevant optical properties of biological aerosol particles. They provide some excellent comparison tests of which parameters and processes are important, and provide a framework for understanding these findings.

Major comments:

Referee Comment 1:

The authors frequently talk about how they do not intend this paper to be a comprehensive literature review (e.g., lines 137-140), yet it is still important that they cover the range of values that are found in the literature. Specifically, I would like to see an inclusion of more up to date information on pollen and fungal spore rupture (see next comment)

Responses and revisions 1:

We thank the referee for this suggestion and agree that some discussion on pollen and fungal spore rupture should be included as an additional process. We added information on the ranges of sizes of fragments of pollen and fungal spores to the revised manuscript and indicated these changes in the following comments below.

Comment 2:

The authors do not provide equal weight to the physical ageing via rupture of biological particles such as fungal spores and pollen. Physical ageing processes are noted, but they have not done the appropriate literature review to accurately capture how some types of biological particles may change. This represents an important atmospheric secondary process that can change both the size distribution as well as potentially the optical properties. This should be mentioned in the introduction when discussing "physical transformations" around line 100, and more specifically throughout the paper, particularly for including observed size distributions and their influence the optical properties. Pollen rupture is mentioned briefly on lines 168-169 and as a single referenced line item in Table 1, but this underestimates this process based on the long list of epidemiological literature on this process (e.g., Suphiolglu et al. 1992; Grote et al., 2001; Taylor et al. 2002; Taylor et al. 2004). More recently, fungal spores have been shown to rupture as well (Lawler et al., 2020; China et al., 2017), and this has not been mentioned at all in the text nor in Table 1. Overall, the authors spend a lot of time on the chemical processing (e.g., nitration) and its impacts, but very little on this physical process.

Responses and revisions 2:

We thank the referee for pointing us to these references on the rupture of pollen and fungal spores as it contributes to ageing processes of pollen and fugal spores.

We add the following text:

- in the introduction at line 78:

In particular pollen rupture leads to a huge increase in the number of subpollen particles (SPPs) (Bacsi et al., 2006; Suphioglu et al., 1992; Taylor et al., 2004; Wozniak et al., 2018). By assuming that one pollen grain releases up to 106 SPPs, regional model studies suggested that the resulting SPPs can significantly suppress seasonal precipitation (Wozniak et al., 2018).

- at the end of 'Physical transformations' at line 105:

For example, the break-up of pollen or fungi due to rupture can lead to higher number concentrations by several orders of magnitude (Suphioglu et al., 1992; Wozniak et al., 2018).

- At the end of Section 2.1, we modified the text as follows at line 174:

At high RH and during precipitation or thunderstorms, pollen absorb water and one pollen grain can release ~10^3 SPPs due to osmotic pressure (Grote et al., 2001; Suphioglu et al., 1992). ~~Similarly, a biologically-driven physical processes might lead to enhancement of NBAP as it has been observed that pollen ruptures into~~ This process can result in fragments with diameters of 1-4 µm and number concentrations of N_{SPP} ~0.1 cm^{-3} during thunderstorms (Zhang et al., 2019). These concentrations correspond to ~1 to 25 ng m^{-3} (D_{SPP} < 2 μm) (Miguel et al., 2006). Laboratory chamber measurements have shown that SPPs from rupture of fresh birch pollen or grass pollen have diameters of in the range of 0.03 to 4.7 μm (Taylor et al., 2002, 2004). Recent laboratory measurements suggest that also fungal spores can rupture, resulting in subfungi particles (SFPs) with D_{SFP} of 0.03 to 0.9 μm after exposure to high relative humidity (China et al., 2016). Ambient measurements suggest N_{SFP} of 150 to 455 cm^{-3} (10 nm < D_{SFP} < 100 nm) after rainfall; observed peaks in aerosol size distributions at 20 nm < D_{SFP} < 50 nm which frequently appeared 1.5 days after rain events were ascribed to such rupture events (Lawler et al., 2020).

- We modified the following sentences at the end of Section 2.3.1 at line 225:

The hygroscopity of pollen is similar to that of bacteria: The κ value of intact pollen grains falls into the range of $0.03 \leq \kappa_{pollen} \leq 0.17$ (Chen et al., 2019; Pope, 2010; Tang et al., 2019); ~~in agreement with κ of pollen kitts on the surface of pollen~~ pollenkitts (which are parts of pollen surface) and SPPs (which are fragments after rupture) are slightly more hygroscopic ($0.14 \leq \kappa_{pollenkitt} \leq 0.24$, $0.1 \leq \kappa_{SPP} \leq 0.2$) (Mikhailov et al., 2019; Prisle et al., 2019; Mikhailov et al., 2020) than intact pollen grains, which can be explained by the nonuniform composition of pollen (Campos et al., 2008).

- We added the above numbers to Table 1 (please see our response to Comment 6).

Comment 3:

Overall, the sensitivity studies described are useful, but there was little discussion of box model results. Specifically, more detail on the following would enhance the paper

Author response: We provided more details on the model results as specified below.

: a. lines 367-369 – why does the absorption coefficient increase at the higher wavelengths?

Author response (a): We added the following explanation at line 395:

Assuming κ = 0.25 (S_{opt10}) instead of κ = 0.03 (S_{opt9}), leads to an increase of the scattering coefficient by 17 to 90% at RH = 90%. Also the absorption coefficient increases by ~40% at λ > 2 μm. This trend can be explained as the imaginary part of water is higher by three orders of magnitude at λ ~2 μm compared to that at λ ~1 μm (Kou et al., 1993). It can be concluded that the importance of κ_{PBAP} increases at higher RH, as under these conditions PBAP hygroscopic growth is most efficient.

b. Figure 6 – large changes with refractive indices (no surprise) but hardly any discussion in the text of what changes are important

Author response (b): We discussed in more detail what changes are important. The change of optical properties within different species of bacteria (red lines) or different species of fungi (blue lines) can be larger than that between bacteria and fungi. Therefore, the detailed information about the species of PBAPs is important in order to better model the optical properties. These differences in scattering and absorption can induce significant change in radiative forcing and will be discussed in section 4.1.4.

We modified the following at the beginning of section 4.1.3 at line 436:

The complex refractive index of PBAPs can be explained by their building blocks of various functional groups (Hill et al., 2015). Here the complex refractive indices of PBAPs are based on the measurements of *Erwinia herbicola* by Arakawa et al. (2003) and twelve other PBAPs by Hu et al. (2019); the complex refractive indices of 'other particles' in the model are the averaged values based on the volume fractions of ammonium sulfate, soot, and water (***Table 2***). The calculated scattering and absorption coefficients of the total particle population are shown in ***Figure 6***. Scattering coefficients for different PBAPs vary by a factor of up to four and the absorption coefficients by a factor of up to six.

The difference of optical properties between bacteria species or fungi species can be larger than that between these two types of PBAPs. Therefore, detailed information on PBAP species is important in order to estimate their direct interaction with radiation (**Section 4.1.4**).

c. Figure 7: why are the nitrated changes in scattering large at smaller wavelengths?

Response (c): The scattering and absorption coefficients are affected by the real part and the imaginary part in non-linear ways. We modified discussion at line 453:

Due to the lack of data on the change of complex refractive index (Δm) for nitrated proteins in PBAPs, we assume nitrated PBAPs have a similar change in the refractive index to that of SOA (S_{opt12} and S_{opt13}). ~~The scattering coefficient can change by up to 20% and the absorption coefficient by a factor of three at λ = 0.42 µm (***Figure 7***).~~ After nitration, the scattering coefficient decreases by ~20% in the range of 300 nm < λ < 450 nm and is nearly constant in the range of 460 nm < λ < 560 nm (***Figure 7a***). The scattering coefficient depends non-linearly on the real and the imaginary parts. The absorption coefficient of nitrated PBAPs is higher by 14% to 160% in the range of 300 nm < λ < 540 nm (***Figure 7b***) and is nearly constant in the range of 550 nm < λ < 560 nm. The largest difference (~160%) for absorption coefficient is observed at 440 nm and the smallest difference (~6%) is observed at 560 nm, which can be attributed to the wavelength-dependent change of the imaginary part (Δk) (Liu et al., 2015). ~~Thus, the variability in scattering/absorption properties of BAP due to Δm caused by nitration is likely smaller than due to Δm caused by different BAP types.~~

Comment 4:

The ranking in Figure 11 is potentially useful but ultimately confusing. Please revise the accompanying text to make this figure more clear – right now the discussion is scattered and it would help to clarify this figure more, as it is ultimately very useful.

Responses and revisions 4:

We thank the referee for acknowledging the value of the last figure. We modified and extended the discussion of Figure 11 in Section 5. Since Referee #2 had also major concerns about this figure, we frame its discussion now more in the context of our process model results and the need of future studies to characterize PBAP properties, rather than making strong claims about global implications. We also emphasize throughout the revised manuscript that the role of PBAP in the atmosphere for the aerosol direct and indirect effect may be limited due to their small number concentration on a global scale. However, detailed knowledge on PBAP properties that affect their interaction with radiation and water vapor is also essential to properly describe their transport, dispersion and lifetime in the atmosphere, which might affect the global modification of biodiversity and impacts public health.

As the previous title did not imply this, we changed it accordingly:

Sensitivities to biological aerosol particle properties and ageing processes: Potential implications for aerosol-cloud interactions and optical properties

We changed Figure 11 and its caption to make it clearer:

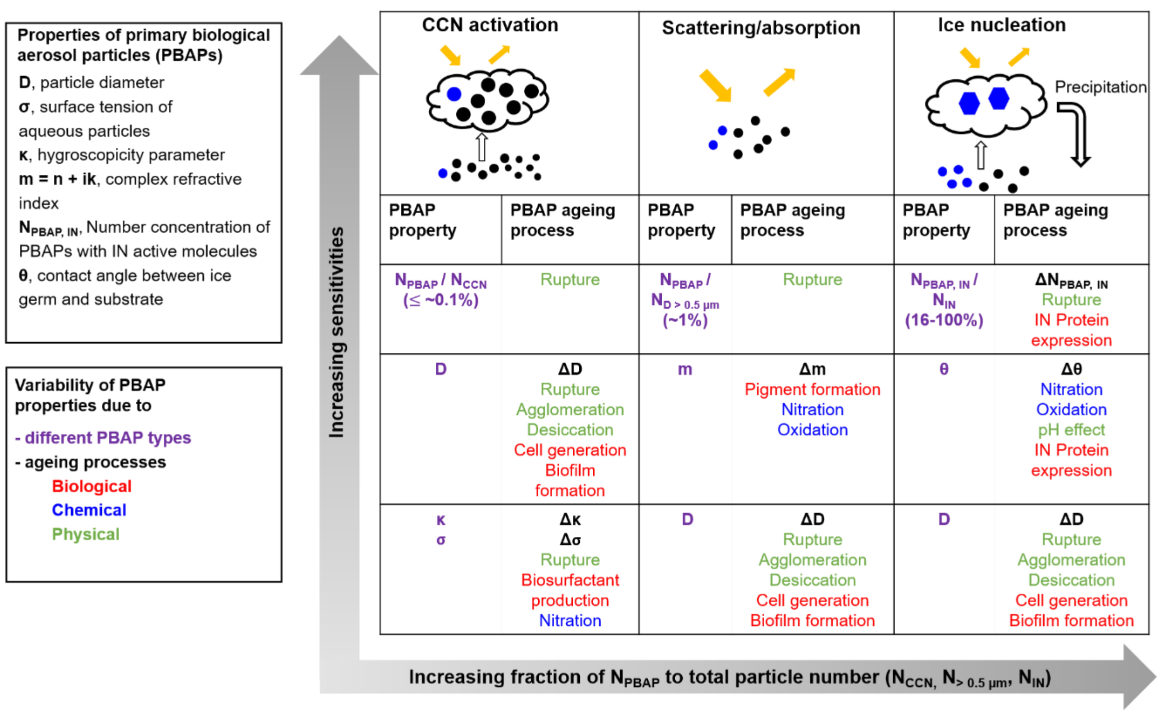

Figure 11. Schematic of PBAP types and ageing processes that affect their aerosol-cloud interactions and optical properties. The bottom arrow shows the increasing fraction of N_{PBAP} to total particles (N_{CCN}, $N_{>5\mu m}$, and N_{IN}, respectively). The left arrow indicates the increasing sensitivity to PBAP properties as predicted based on our process model studies. The various properties might be modified by physical (green), chemical (blue) and biological (red) ageing processes.

We rewrote Section 5 (Conclusions):

Based on our model sensitivity studies, we can rank the relative importance of the PBAP properties and processes in ***Figure 1*** for their aerosol-cloud interactions and optical properties. Given the limitations of our process models in terms of scales, dimensions and parameter spaces, our results should be considered as qualitative, rather than quantitative estimates; the focus of our study is the comparison of relative changes due to various physicochemical parameters. Several findings of our model sensitivity results repeat those that have been drawn previously for other atmospheric particle types (Hoose and Möhler, 2012; McFiggans et al., 2005; Moise et al., 2015). However, in addition, unlike other atmospheric particles, PBAPs may constitute living microorganisms; thus, their properties may not only be modified by chemical and physical processes (marked in green and blue, respectively, in ***Figure 11***), but also by biological processes (marked in red in ***Figure 11***). To date, the extent to which these biological processes affect PBAP properties in the atmosphere is not known due to the lack of suitable data sets for atmospheric models. Our sensitivity studies, in combination with ***Figure 11***, give a first idea on which biological processes could modify relevant PBAP properties.

(1) For any climate-related effect, the number concentration of PBAPs (N_{PBAP}) is the most important parameter. The PBAP number concentrations assumed in our estimates are based on measurements near the ground (Huffman et al., 2012; Jaenicke, 2005; Tong and Lighthart, 2000; Whitehead et al., 2016), which typically decrease with altitude (Gabey et al., 2013; Perring et al., 2015; Ziemba et al., 2016). Thus, processes that affect N_{PBAP} in the atmosphere need to be well constrained; these processes include not only direct emissions but also particle fragmentation (rupture) or possibly new cell generation (multiplication). The number fraction of PBAPs to total CCN is relatively small ($\leq$ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

PBAPs contribute ~1% to large particles with D > 0.5 µm (Zhang et al., 2019), which makes them relatively important for scattering/absorption at a limited range of wavelengths. Only in the presence of high N_{PBAP}, it is expected that they have (local) impacts on the direct aerosol effect.

The number concentration of PBAPs that nucleate ice at T > -10°C is on the order of 10^{-5} to 10^{-3} cm^{-3} (Murray et al., 2012). PBAPs comprise the predominant fraction of atmospheric particles that efficiently nucleate ice at these temperatures, i.e. N_{PBAP}/N_{IN} ~100% at T > -10°C (Hoose and Möhler, 2012). This fraction decreases at temperatures at which more abundant particles (such as dust) are also efficient ice nuclei: For example, at -30 °C, PBAPs contribute 16% to 76% (Prenni et al., 2009) or 33% (Pratt et al., 2009) to total IN in mixed-phase clouds. Lab measurements have shown that up to 100% of pollen grains have IN nucleating macromolecules on their surface, whereas only 0.01 to 10% of bacteria express the proteins or other macromolecules that initiate ice nucleation (Failor et al., 2017; Joly et al., 2013; Pummer et al., 2015).

(2) The size of PBAPs influences the effects in ***Figure 11*** to different extents: While it is likely the most important parameter to determine their ability to act as CCN compared to hygroscopicity and surface tension, its role for PBAPs' optical properties is smaller than that of the refractive index. Also PBAP size plays a less important role than surface properties in the efficiency of ice nucleation. While several biological processes may increase the size of PBAP (e.g. agglomeration, cell generation), these changes are likely not important for the CCN activity of supermicron PBAPs since they will be activated under most conditions and thus an increase in their size does not affect their CCN behavior. However, modifications in the size, hygroscopicity (κ_{PBAP}), and surface tension (σ_{PBAP}) of smaller PBAPs, such as viruses, SPPs and SPFs, can influence their CCN activation. κ_{PBAP} might be modified by physical (e.g., release of inner molecules due to rupture of pollen and fungal spores, condensation of gases), biological (e.g., formation of biosurfactants or other metabolic products), and chemical (e.g., nitration, oxidation) processes. Thus, processes that modify hygroscopic or surface tension properties of these smaller PBAPs might significantly change their ability to take up water vapor and form cloud droplets.

(3) The optical properties of PBAP are mostly determined by their complex refractive index ($m = n + ik$), especially by the imaginary part (k) which varies by three orders of magnitude among PBAPs. Under conditions when PBAPs significantly affect Mie scattering, small variabilities in the refractive index due to PBAP types or ageing processes might enhance (or diminish) their direct interaction with radiation (scattering/absorption). Modification processes include pigment formation as a defense mechanism of bacteria to oxidative stress (Fong et al., 2001; Noctor et al., 2015; Pšenčík et al., 2004; Wirgot et al., 2017) and nitration/oxidation of surface molecules (He et al., 2018; Liu et al., 2015; Nakayama et al., 2018). Additional biological processes such as biofilm formation are also included in ***Figure 11*** although experimental data are lacking to estimate their impact on PBAP optical properties.

(4) The ice nucleation activity of aerosol particles is often parameterized with a single contact angle (θ) between the particle surface and ice. ***Table 1*** shows that θ significantly differs among different PBAP types. In addition, our model sensitivity studies suggest that even a small change ($\Delta\theta_{PBAP}$ ~1°) as caused by chemical processing of surfaces, pH change of the surrounding aqueous phase, or biological processes such as protein expression level might significantly affect this activity. At

temperatures at which PBAPs are the predominant IN (T > -10 °C), such a small change might translate into large changes in the onset temperature of freezing and cloud glaciation can be affected. Thus, in order to comprehensively account for ice nucleation of PBAPs, not only various PBAP types, but also $\Delta\theta_{PBAP}$ due to modification by chemical and possibly biological processes should be considered in models.

Exceeding numerous recent review articles that highlight the importance of PBAPs in general (Coluzza et al., 2017; Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Haddrell and Thomas, 2017; Šantl-Temkiv et al., 2020; Smets et al., 2016), ***Figure 11*** gives more specific guidance on future measurements of the most sensitive PBAP properties in terms of their interaction with radiation and with water vapor. The detailed knowledge of PBAP properties might be of limited importance for global radiative forcing estimates, but is also relevant to properly describe PBAP transport, dispersion and lifetime in the atmosphere, which eventually affects biodiversity (Morris et al., 2014) and public health (Fröhlich-Nowoisky et al., 2016). While previous studies only focused on the physical and chemical properties, we highlight the uniqueness of PBAPs undergoing biological processes to adapt to the harsh atmospheric conditions; such processes might affect the adaption of PBAPs to atmospheric conditions which impacts their survival, transport and dispersion in the atmosphere.

Minor comments: *1. The acronym used in the paper is inconsistent with the literature on *primary* biological aerosol particles (PBAP) not BAP. While they do talk about some secondary processing of the aerosols, the origin of the particles is still primary (as opposed to secondary formation), and consistency with prior work is helpful.*

Responses 1: We agree with the referee that terminology consistent with the literature should be preferred to avoid confusion. We have changed BAP to PBAP throughout the manuscript, including figures.

2. Line 57 – is the Londahl et al. 2014 the correct reference here? This seems to be an error.

Responses 2: We apologize for the confusion. It was indeed a wrong reference. We replaced it by the correct reference by the same author

3. Line 58 – Myhre et al. 2013 is not in the reference list.

Responses 3: The reference was added to the reference list.

4. Line 98 – Pollen can also nucleate ice – see Diehl et al. 2001

Responses 4: We already included pollen in ***Table 1***. We discussed the ice nucleation property of pollen and modified the following sentences:

In addition to acting as CCN, some species of bacteria, fungi, and pollen can nucleate ice at high temperatures (Hoose and Möhler, 2012; Morris et al., 2004, 2008; Pouzet et al., 2017; Diehl et al., 2001, 2002), which makes them unique in terms of ice nucleation to affect the evolution of mixed-phase clouds at these temperatures (***Figure 1c***).

5. Line 181 – missing word between "that" and "might"? Can't tell what this sentence is supposed to say

Responses: Thanks for pointing out this omission. We completed the sentence as follows:

Due to the similarity of the molecular structure of organic macromolecules (e.g. proteins) and secondary organic aerosols (SOA), it can be likely assumed that nitration might alter the BAP refractive index similar to that of SOA.

6. Table 1: missing many references on the rupture of pollen. I actually think that these numbers are incorrect and very much mis-represent the range of potential sizes (see refs Grote, Taylor, Suphioulglu for a few; listed below). Also, you are missing the rupture of fungal spores (China, Lawler; see references below). Also missing the fact that the hygroscopicity of pollen may change on rupture (not just from oxidation).

Responses 6: We extended Table 1:

Table 1. Physicochemical properties of various PBAPs and their changes due to physical, chemical and biological ageing processes based on literature data.

BAP Types	Physicochemical properties						
	Concentration N (cm^{-3})	Diameter D (μm)	Complex refractive index m (λ) = n + ik	Hygroscopicity κ	Surface tension σ (mN m^{-1})	Number fraction of PBAPs with IN active molecules	Contact angle θ (°)
Bacteria	0.001-1 (1)	1 (17); 0.6-7 (18)	n: 1.5-1.56, k: $3\cdot10^{-5}$-$6\cdot10^{-4}$ (24); n: 1.5-1.56, k: 0-0.04 (25); n: 1.25-1.85, k: 0-0.5 (26)	0.11-0.25 (27)	25, 30, 55, 72 (35)	~0.1%, ~1%, ~10% (36)	32-34 (39); 4-20 (40); 28, 33, 44 (41)
Fungal spores	0.001-0.01 (2)	3-5 (4); 1-30 (5)	n: 1.25-1.75, k: 0-0.32 (26)				30-33 (42)
Subfungi particles (SFPs)	150-455 (3)	0.01-0.1 (3); 0.02-0.05 (3); 0.03-0.9 (19)					

Fern spores	10^{-5} (4)	1-30 (4)				
Pollen	0.001 (5)	5-100 (20)	n: 1.3-1.75, k: 0.01-0.2 (26)	0.03-0.073 (28); 0.036-0.048 (29); 0.05-0.1 (30); 0.08-0.17 (31)	~100% (37,38)	14-30 (40); 15, 16.3 (43)
Subpollen particles (SPPs)	0.1 (6)	1-4 (6); 0.03-4 (21); 0.12-4.67 (22);		0.14-0.24 (32); 0.12-0.13 (33); 0.1-0.2 (34)		
Viruses	0.01 (4)	0.01-0.3 (4) 0.04-0.2 (23)				
Ambient PBAPs	0.1-1 (7); 1-8 (8);	> 0.4 (7,8)				
Ambient PBAPs	0.2-1.2 (9); 0.04-0.13 (10); 0.012-0.095 (11); 0.01-1.4 (12); 0.57-3.3 (13); 0.1-0.43 (14); 0.02-0.09 (15); 0.005-0.5 (16)	> 1 (9-16)				

Ageing processes of PBAPs			
	Physical ageing	Chemical ageing	Biological ageing
Bacteria	Agglomeration: $\Delta D > 0$, $\Delta N < 0$ (18)	Nitration: $\Delta n > 0$, $\Delta k > 0$ (44) ; Nitration: $\Delta\theta$ ~1° (41); pH changes: $\Delta\theta$ ~ 1.5° (41).	Biosurfactant production: $\sigma < 0$ (35); Biofilm formation: $\Delta D > 0$ (45); Endospore formation: $\Delta N > 0$ (46); Cell generation: $\Delta D > 0$ (47); Desiccation: $\Delta D < 0$ (48); Pigment formation: $\Delta k > 0$ (49,50); IN protein expression: $\Delta\theta < 0$ (no data yet)
Fungi	Rupture: $\Delta D < 0$, $\Delta N > 0$ (3,19)		Biosurfactant production: $\sigma < 0$ (35); Germination: $\Delta N > 0$ (49) ; Desiccation: $\Delta D < 0$ (48).
Pollen	Rupture: $\Delta D < 0$, $\Delta N > 0$ (6,21,22)	Oxidation: $0.5 \leqslant \Delta\theta \leqslant 0.8°$ (43)	

(1) Total bacteria, Tong and Lighthart et al., 2000; (2) Elbert et al., 2007; (3) After rainfall, Lawler et al., 2020; (4) Després et al., 2012; (5) blooming times, Huffman et al. 2010; (6) thunderstorm times, Zhang et al., 2019; (7) Based on protein dyes, Lake Baikal, Russia, Jaenicke, 2005; (8) Based on protein dyes, Mainz, Germany, Jaenicke, 2005; (9) In the Amazon, Whitehead et al., 2016; (10) In the Amazon, Huffman et al., 2012; (11) Puy de Dôme, Gabey et al. 2013; (12) In megacity Beijing, China, Wei et al., 2016; (13) In Megacity Nanjing, China, Yu et al., 2016; (14) High altitude, Ziemba et al., 2016b; (15) High altitude, Perring et al. 2015; (16) High concentration observed during and after rain, Huffman et al., 2013; (9) to (16) are based on autofluorescence of PBAPs; (17) Burrows et al., 2009a; (18) Lighthart 1997; (19) China et al., 2016; (20) Pöhlker et al., 2013; (21) Taylor et al., 2004; (22) Taylor et al., 2002; (23) Verreault et al., 2008; (24) Arakawa et al., 2003; (25) Thrush et al., 2010; (26) Hu et al. 2019; (27) Lee et al., 2002; (28) Pope et al. 2010; (29) Tang et al., 2019; (30) Chen et al., 2019; (31) Griffiths et al., 2012 ; (32) pollenkitt, Prisle et al., 2019; (33) Mikhailov et al., 2019; (34) Mikhailov et al., 2020; (35) Renard et al., 2016; (36) T ~-10 °C, immersion freezing,

Pseudomonas syringae bacteria, *Pseudoxanthomonas* sp., *Xanthomonas* sp., Joly et al., 2013; (37) deposition freezing for pollen, Diehl et al., 2001; (38) immersion and contact freezing for pollen, Diehl et al., 2002; (39) Hoose and Möhler, 2012; (40) Chen et al., 2008; (41) immersion freezing for *Pseudomonas syringae*, and *Pseudomonas fluorescens*, Attard et al., 2012; (42) immersion freezing for fungi, Kunert et al., 2019; (43) deposition freezing of silver birch and grey alder pollen, Gute and Abbatt, 2018; (44) nitrated SOA (toluene as precursor) to represent nitrated BAP, Liu et al., 2015; (45) Morris et al., 2008; (46) Enguita et al., 2003; (47) Ervens and Amato, 2020; (48) Barnard et al., 2013; (49) Pšenčík et al., 2004; (50) Fong et al., 2001.

We added some texts in section 2.3.1 at line 225:

The hygroscopicity of pollen is similar to that of bacteria: The κ value of intact pollen grains falls into the range of $0.03 \leq \kappa_{pollen} \leq 0.17$ (Chen et al., 2019; Pope, 2010; Tang et al., 2019), pollenkitts (which are parts of pollen surface) and SPPs (which are fragments after rupture) are slightly more hygroscopic ($0.14 \leq \kappa_{pollenkitt} \leq 0.24$, $0.1 \leq \kappa_{SPP} \leq 0.2$) (Mikhailov et al., 2019; Prisle et al., 2019; Mikhailov et al., 2020) than intact pollen grains, which can be explained by the nonuniform composition of pollen (Campos et al., 2008).

In addition, in the conclusion section at line 720, we modified the following sentences:

κ_{PBAP} might be modified by physical (e.g., release of inner molecules due to rupture of pollen and fungal spores, condensation of gases), biological (e.g., formation of biosurfactants or other metabolic products), and chemical (e.g., nitration, oxidation) processes.

7. Lines 283-284: The text should more clearly state that certain classes of PBAP are excluded based on the 0.5-2.8 micron size representation.

Responses 7: We add at line 312:

Thus, the simulations focus on PBAPs in this size range and exclude smaller (e.g. viruses, SFPs or SPPs) and larger (e.g. pollen grains) particles.

8. Line 342: Fungal spores could also be on the order of this size. . .

Responses 8: We added at line 365:

Larger PBAPs (D_{PBAP} = 3 µm, S_{opt6}) such as SPPs and fungal spores lead to an increase in the scattering coefficient by a factor of 1.4-4.7 depending on λ.

9. Lines 360-363: This line downplays the potential importance of non-spherical particles. The true atmospheric range of moisture conditions is not enough to say what is more likely, therefore this speculation should be removed and it would be better to discuss what types of uncertainties non-spherical particles would include.

Responses 9: We removed the speculation at lines 360-363. We found some papers about the uncertainties of non-spherical particles. We added the following to the end of section 4.1.1 at line 384:

Non-sphericity of particles might translate into the same changes as caused by different particles sizes, which might induce uncertainties including optical depth and surface albedo (Kahnert et al., 2007). These uncertainties on scattering and absorption caused by non-spherical shape might be of comparable magnitude to that caused by the complex refractive index (Yi et al., 2011).

10. Figure 3: the caption states that there is an a/b panel to capture scattering and absorption, yet only the scattering is shown.

Responses 10: We apologize for the omission of Figure 3b in the original manuscript. Actually, we added Figure 3b in the supporting information (as Figure S2) We changed the Figure caption accordingly. Its information is rather limited since the absorption for all PBAPs is (nearly) identical, i.e., the absorption coefficient is not affected in the presence of PBAPs.

11. Line 392: "very small PBAP" could also be pollen or fungal fragments. Please see literature suggestions in the major comments.

Responses 11: We have changed the sentences at line 427:

Only for very small PBAPs, i.e. representative for viruses, SPPs or SFPs (***Section 2.1***), the curvature term significantly influences *s* (***Figure 5***).

12. Line 422: I think this is supposed to be S15 and S16?

Responses 12: The referee is correct; we meant to refer to S15 and S16, rather than S13 and S14. As we deleted some simulations in Table 3 to make it shorter, their number changed to S12 and S13.

13. Line 471: what is delta_mBAP? First use, please define. (perhaps including S13, S15 and S16?)

Responses 13: Delta-mBAP means the change of refractive index due to different types of BAP or nitration. We will define Delta-mBAP at line 454. S13 means simulation 13. We will define them at first use at line 316, line 331, and line 343.

14. Table 3: last row – is dm_aged the same as dm_nitrated? Different terminology than Table 2.

Responses 14: They are the same. We use now dm_nitrated for the whole manuscript.

15. Also in Table 3 – what is the dm actually referring to? Hard to tell from comparing with Table 2.

Responses: dm means the change of refractive index. We defined this at line 454. We also included more information in Table 3 (see response to Comment 3b).

16. Lines 495-298: Could be compared with the observed values of Sc from Steiner et al. 2015

Responses: We thank the referee for this additional reference. In addition, we also added data from a recent publication by Mikhailov et al. (2020) who investigated the hygroscopic behavior of various SPPs. We added the following text at line 549:

Steiner et al. (2015) reported critical supersaturations (S_c) of 0.81 (± 0.07)% for 50-nm SPPs and 0.26 (± 0.03)% for 100 nm SPPs. These values are similar to the values discussed above (0.68% to 1.79% for 50-nm particles, 0.24% to 0.69% for 100-nm particles) and are also in agreement with values based on the hygroscopicity ($0.1 \leq \kappa_{SPP} \leq 0.2$) reported by Mikhailov et al. (2019, 2020).

17. Line 500 – The missed rupture literature could also be important here. Physical processes like rupture could create many more hygroscopic particles.

Responses: Many thanks. We have added rupture and modified the sentences at line 547:

Thus, only for fairly small PBAPs such as viruses, SPPs and SFPs ($D \leq 100$ nm), the hygroscopicity κ_{PBAP} may impact their CCN activation.

18. Lines 568-571 – overall, these changes are really hard to see in the figure. Is it possible to overlay Figures 10a/b so they can be more directly compared?

Responses: We intentionally separate figure 10a, 10b and 10c to show that the start of Bergeron-Findeisen process occurs at different temperatures. Namely, the curves in figure 10a, 10b, and 10c are the same whereas the y-axis shows different scales. We make this clearer now in the figure caption at line 643:

Figure 10. Percentage contribution of ice water content (%IWC, dashed lines) and liquid water content (%LWC, solid lines) total adiabatic water content for θ_{PBAP} of (a) 4°; (b) 20°; (c) 40° and (d) 37° and 38°. The curves in the first three panels exhibit similar shapes for different temperature ranges, i.e. the Bergeron-Findeisen process starts at different temperatures. The last panel shows that even when the contact angle increases by 1°, the temperature, at which the LWC fraction starts decreasing, differs significantly.

19. Lines 630-632: What is the reference for this sentence "However, as it has been shown that at many locations NBAP/Ntotal is approximately constant. . .." – this is not true for fungal spores *and pollen. The emissions of these types of PBAP are very spatially and temporally heterogeneous, and tend to be more event-based than consistent.*

Responses 19: We reworded this paragraph as follows at line 690:

The number fraction of PBAPs to total CCN is relatively small (≤ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

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Reviewer 2:

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 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-Findeinsen (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.*

Response: We thank the referee for their constructive comments on our manuscript. We agree that some of the conclusions may have been too strong given the limitation of our process model studies. We have substantially revised our manuscript; the main changes include

- We changed the title to

Sensitivities to biological aerosol particle properties and ageing processes: Potential implications for aerosol-cloud interactions and optical properties

- We showed more clearly the commonalities between PBAP and other aerosol types
- we discuss in more detail the uniqueness of PBAP in terms of their modification by biological processes.
- Throughout the manuscript, and in particular in the last section, we revised the discussion of the importance of PBAP properties and their modification. We make it clearer now that their importance for radiative forcing may be limited under many conditions; however, the

properties discussed throughout the manuscript (ice nucleation and CCN activity, optical properties) should not only be explored to constrain the climatic effects but also to constrain their transport, survival and dispersion in the atmosphere. While our model framework is clearly not suited to give comprehensive estimates of all these implications, we consider our study, including Figure 11, as a useful guidance to identify the most sensitive PBAP properties and processes.

We give more details on our revisions in our point-by-point responses below.

In the abstract, because our small-scale model cannot quantify the effect of PBAP on CCN, direct radiation, and IN, we have rephrased the abstract as follows:

Primary biological aerosol particles (PBAPs) such as bacteria, viruses, fungi, and pollen, represent a small fraction of the total aerosol burden. Based on process model studies, we identify trends in the relative importance of PBAP properties, e.g. number concentration, diameter, hygroscopicity, surface tension, contact angle, for their aerosol-cloud interactions and optical properties. While the number concentration of PBAPs likely does not affect total CCN concentrations globally, small changes in the hygroscopicity of submicron PBAPs might affect their CCN ability and thus their inclusion into clouds. Given that PBAPs are highly efficient atmospheric ice nuclei at T > -10 °C, we suggest that small changes in their sizes or surface properties due to chemical, physical or biological processing might translate into large impacts on ice initiation in clouds. Predicted differences in the direct interaction of PBAPs with radiation can be equally large between different species of the same PBAP type and among different PBAP types. Our study shows that not only variability of PBAP types, but also their physical, chemical, and biological ageing processes might alter their CCN and IN activities and optical properties to affect their aerosol-cloud interactions and optical properties. While these properties and processes likely affect radiative forcing only on small spatial and temporal scales, we highlight their potential importance for PBAP survival, dispersion and transport in the atmosphere.

In addition, we largely rewrote the conclusion to stress the features that are characteristic for PBAPs (also our response to Comment 4 by Referee #1).

Based on our model sensitivity studies, we can rank the relative importance of the PBAP properties and processes in ***Figure 1*** for their aerosol-cloud interactions and optical properties. Given the limitations of our process models in terms of scales, dimensions and parameter spaces, our results should be considered as qualitative, rather than quantitative estimates; the focus of our study is the comparison of relative changes due to various physicochemical parameters. Several findings of our model sensitivity results repeat those that have been drawn previously for other atmospheric particle types (Hoose and Möhler, 2012; McFiggans et al., 2005; Moise et al., 2015). However, in addition, unlike other atmospheric particles, PBAPs may constitute living microorganisms; thus, their properties may not only be modified by chemical and physical processes (marked in green and blue, respectively, in ***Figure 11***), but also by biological processes (marked in red in ***Figure***

11). To date, the extent to which these biological processes affect PBAP properties in the atmosphere is not known due to the lack of suitable data sets for atmospheric models. Our sensitivity studies, in combination with ***Figure 11***, give a first idea on which biological processes could modify relevant PBAP properties.

(1) For any climate-related effect, the number concentration of PBAPs (N_{PBAP}) is the most important parameter. The PBAP number concentrations assumed in our estimates are based on measurements near the ground (Huffman et al., 2012; Jaenicke, 2005; Tong and Lighthart, 2000; Whitehead et al., 2016), which typically decrease with altitude (Gabey et al., 2013; Perring et al., 2015; Ziemba et al., 2016). Thus, processes that affect N_{PBAP} in the atmosphere need to be well constrained; these processes include not only direct emissions but also particle fragmentation (rupture) or possibly new cell generation (multiplication). The number fraction of PBAPs to total CCN is relatively small ($\leq$ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

PBAPs contribute ~1% to large particles with D > 0.5 µm (Zhang et al., 2019), which makes them relatively important for scattering/absorption at a limited range of wavelengths. Only in the presence of high N_{PBAP}, it is expected that they have (local) impacts on the direct aerosol effect.

The number concentration of PBAPs that nucleate ice at T > -10°C is on the order of 10^{-5} to 10^{-3} cm^{-3} (Murray et al., 2012). PBAPs comprise the predominant fraction of atmospheric particles that efficiently nucleate ice at these temperatures, i.e. N_{PBAP}/N_{IN} ~100% at T > -10°C (Hoose and Möhler, 2012). This fraction decreases at temperatures at which more abundant particles (such as dust) are also efficient ice nuclei: For example, at -30 °C, PBAPs contribute 16% to 76% (Prenni et al., 2009) or 33% (Pratt et al., 2009) to total IN in mixed-phase clouds. Lab measurements have shown that up to 100% of pollen grains have IN nucleating macromolecules on their surface, whereas only 0.01 to 10% of bacteria express the proteins or other macromolecules that initiate ice nucleation (Failor et al., 2017; Joly et al., 2013; Pummer et al., 2015).

(2) The size of PBAPs influences the effects in ***Figure 11*** to different extents: While it is likely the most important parameter to determine their ability to act as CCN compared to hygroscopicity and surface tension, its role for PBAPs' optical properties is smaller than that of the refractive index.

Also PBAP size plays a less important role than surface properties in the efficiency of ice nucleation. While several biological processes may increase the size of PBAP (e.g. agglomeration, cell generation), these changes are likely not important for the CCN activity of supermicron PBAPs since they will be activated under most conditions and thus an increase in their size does not affect their CCN behavior. However, modifications in the size, hygroscopicity (κ_{PBAP}), and surface tension (σ_{PBAP}) of smaller PBAPs, such as viruses, SPPs and SPFs, can influence their CCN activation. κ_{PBAP} might be modified by physical (e.g., release of inner molecules due to rupture of pollen and fungal spores, condensation of gases), biological (e.g., formation of biosurfactants or other metabolic products), and chemical (e.g., nitration, oxidation) processes. Thus, processes that modify hygroscopic or surface tension properties of these smaller PBAPs might significantly change their ability to take up water vapor and form cloud droplets.

(3) The optical properties of PBAP are mostly determined by their complex refractive index (m = n + ik), especially by the imaginary part (k) which varies by three orders of magnitude among PBAPs. Under conditions when PBAPs significantly affect Mie scattering, small variabilities in the refractive index due to PBAP types or ageing processes might enhance (or diminish) their direct interaction with radiation (scattering/absorption). Modification processes include pigment formation as a defense mechanism of bacteria to oxidative stress (Fong et al., 2001; Noctor et al., 2015; Pšenčík et al., 2004; Wirgot et al., 2017) and nitration/oxidation of surface molecules (He et al., 2018; Liu et al., 2015; Nakayama et al., 2018). Additional biological processes such as biofilm formation are also included in ***Figure 11*** although experimental data are lacking to estimate their impact on PBAP optical properties.

(4) The ice nucleation activity of aerosol particles is often parameterized with a single contact angle (θ) between the particle surface and ice. ***Table 1*** shows that θ significantly differs among different PBAP types. In addition, our model sensitivity studies suggest that even a small change ($\Delta\theta_{PBAP}$ ~1°) as caused by chemical processing of surfaces, pH change of the surrounding aqueous phase, or biological processes such as protein expression level might significantly affect this activity. At temperatures at which PBAPs are the predominant IN (T > -10 °C), such a small change might translate into large changes in the onset temperature of freezing and cloud glaciation can be affected. Thus, in order to comprehensively account for ice nucleation of PBAPs, not only various PBAP types, but also $\Delta\theta_{PBAP}$ due to modification by chemical and possibly biological processes should be considered in models.

Exceeding numerous recent review articles that highlight the importance of PBAPs in general (Coluzza et al., 2017; Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Haddrell and Thomas, 2017; Šantl-Temkiv et al., 2020; Smets et al., 2016), ***Figure 11*** gives more specific guidance on future measurements of the most sensitive PBAP properties in terms of their interaction with radiation and with water vapor. The detailed knowledge of PBAP properties might be of limited importance for global radiative forcing estimates, but is also relevant to properly describe PBAP transport, dispersion and lifetime in the atmosphere, which eventually affects biodiversity (Morris et al., 2014) and public health (Fröhlich-Nowoisky et al., 2016). While previous studies only focused on the physical and chemical properties, we highlight the uniqueness of PBAPs undergoing biological processes to adapt to the harsh atmospheric conditions; such processes might affect the adaption of PBAPs to atmospheric conditions which impacts their survival, transport and dispersion in the atmosphere.

Detailed comments: Line 11. Are biological fragments considered here?

Response: Also in response to Referee #1, we added more details and discussion on rupture of pollen and fungi. Accordingly, we added text in the introduction, Section 2.1 (including Table 1), and 2.3.1.

- in the introduction at line 78:

In particular pollen rupture leads to a huge increase in the number of subpollen particles (SPPs) (Bacsi et al., 2006; Suphioglu et al., 1992; Taylor et al., 2004; Wozniak et al., 2018). By assuming that one pollen grain releases up to 10^6 SPPs, regional model studies suggested that the resulting SPPs can significantly suppress seasonal precipitation (Wozniak et al., 2018).

- At the end of Physical transformations at line 105:

For example, the break-up of pollen or fungi due to rupture can lead to higher number concentrations by several orders of magnitude (Suphioglu et al., 1992; Wozniak et al., 2018).

- We modified the text at the end of Section 2.1 at line 174:

At high RH and during precipitation or thunderstorms, pollen absorb water and one pollen grain can release ~10^3 SPPs due to osmotic pressure (Grote et al., 2001; Suphioglu et al., 1992). ~~Similarly, a biologically-driven physical processes might lead to enhancement of NBAP as it has been observed that pollen ruptures into~~ This process can result in fragments with diameters of 1-4 µm and number concentrations of N_{SPP} ~0.1 cm^{-3} during thunderstorms (Zhang et al., 2019). These concentrations correspond to ~1 to 25 ng m^{-3} (D_{SPP} < 2 µm) (Miguel et al., 2006). Laboratory chamber measurements have shown that SPPs from rupture of fresh birch pollen or grass pollen have diameters of in the range of 0.03 to 4.7 µm (Taylor et al., 2002, 2004). Recent laboratory measurements suggest that also fungal spores can rupture, resulting in subfungi particles (SFPs) with D_{SFP} of 0.03 to 0.9 µm after exposure to high relative humidity (China et al., 2016). Ambient measurements suggest N_{SFP} of 150 to 455 cm^{-3} (10 nm < D_{SFP} < 100 nm) after rainfall; observed peaks in aerosol size distributions at 20 nm < D_{SFP} < 50 nm which frequently appeared 1.5 days after rain events were ascribed to such rupture events (Lawler et al., 2020).

- We modified the following sentences at the end of Section 2.3.1 at line 225:

The hygroscopity of pollen is similar to that of bacteria: The κ value of intact pollen grains falls into the range of $0.03 \leq \kappa_{pollen} \leq 0.17$ (Chen et al., 2019; Pope, 2010; Tang et al., 2019); ~~in~~

~~agreement with κ of pollen kitts on the surface of pollen~~ pollenkitts (which are parts of pollen surface) and SPPs (which are fragments after rupture) are slightly more hygroscopic ($0.14 \leq \kappa_{pollenkitt} \leq 0.24$, $0.1 \leq \kappa_{SPP} \leq 0.2$) (Mikhailov et al., 2019; Prisle et al., 2019; Mikhailov et al., 2020) than intact pollen grains, which can be explained by the nonuniform composition of pollen (Campos et al., 2008).

- We also added above numbers to Table 1.

Line 37. Delete "the"

Response: We deleted 'the' before location.

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

Response: We changed the text as follows at line 38, agreement with the original literature:

In the semirural area of Mainz in central Europe, the number fraction was 1-50% for particles with diameter (D) > 0.4 μm (Jaenicke, 2005).

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

Response: The referee is correct that generally the absorption of radiation by absorbing organic molecules ('brown carbon') may contribute to the semi-direct effect. However, given the small amounts of light-absorbing material in PBAPs and small mass fraction of PBAP total absorbing mass, the global effect is likely small. In addition, the semi-direct effect is mostly triggered by light-absorbing material (e.g. soot particles) above clouds. Given the large sizes of most PBAP, their concentrations decrease strongly with altitude (Ziemba et al., 2016), thus their impact near cloud top may be small. We have added at line 469:

While generally, light-absorbing organics ('brown carbon') might contribute to the aerosol semi-direct effect (Brown et al., 2018; Hansen et al., 1997), i.e. the impact of aerosol heating on clouds, it seems unlikely that PBAPs have a significant contribution to it. Given the supermicron sizes of most PBAPs, their concentration decreases strongly as a function of altitude (Ziemba et al., 2016) and thus their concentration near cloud tops is likely negligible.

Line 165. Must be "agglomerates"

Response: We replaced 'agglomerate' to '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?

Response: The term 'maximum frozen fraction' was misleading. We were referring to the fraction of PBAPs that have IN macromolecules. In the study by Joly et al. (2013), experiments were performed at T ≥ -10 °C. As PBAPs are the predominant atmospheric particles that nucleate ice above this temperature, we focussed in our model studies on this temperature range.

We changed the wording as follows at line 268:

For example, only 0.1 to 10% of *Pseudomonas syringae* cells express IN active macromolecules (Joly et al., 2013).

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

Response: We changed the text as follows at line 263:

~~However, to date it is not fully understood why in lab experiments some of the bacteria cells show freezing behaviour while others from the same population do not and why individual cells show stochastic behaviour in repeated experiments (Lukas et al., 2020).~~ However, it has been shown that bacteria of the same species and within the same population often exhibit different ice nucleation behavior (Bowers et al., 2009; Failor et al., 2017; Fall and Fall, 1998; Lindow et al., 1978; Morris et al., 2004). This behavior has been explained by various expression levels of IN-active macromolecules that are located at the cell surface. Under conditions such as phosphate starvation, the expression level might be higher, which is a strategy to reach nutrients after destroying the cells of plants by freezing (Fall and Fall, 1998). For example, only 0.1 to 10% of *Pseudomonas syringae* cells express IN active macromolecules (Joly et al., 2013). Bacteria from the same population without expression of such molecules did not freeze under the experimental conditions.

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.

Response: The referee is correct that the contact angle should be regarded as a fitting parameter, rather than as a physicochemical parameter, exactly describing the IN surface.

The implications of different expressions for the activation energy, germ formation and other factors included in the classical nucleation theory have been discussed in detail before (Hoose and

Möhler, 2012). We added the reference of Ervens and Feingold (2012) where the detailed model description is given.

We modified at the beginning of section 2.4.2 at line 272:

In agreement with previous studies, we base our discussion on the contact angle as a fitting parameter in the classical nucleation theory (CNT) to parametrize the frozen fraction observed in experiments. In agreement with previous studies, we base our discussion on the contact angle as a fitting parameter in the classical nucleation theory (CNT) to parametrize the frozen fraction observed in experiments. If not reported in the respective experimental studies, we assumed a freezing time of 10 seconds to derive θ from experimental data, in agreement with many experimental conditions (Attard et al., 2012; Gute and Abbatt, 2018; Kunert et al., 2019). All CNT model equations and parameters are identical to those as described by Ervens and Feingold (2012); Hoose and Möhler (2012) discussed different assumptions made for the various variables in the CNT in previous ice nucleation studies.

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.

Response: We agree with the referee that the mentioning of deterministic behaviour is rather distracting at this place. We deleted the text in 255 – 258 and reworded the sentence as follows at line 283:

~~INAS implies that freezing occurs deterministically as opposed to stochastic freezing described by CNT. As the sensitivity of ice nucleation to time is generally small compared to other parameters (Ervens and Feingold, 2013), we fitted their data using CNT and obtained a range of 32° ≤ $\theta_{bacteria}$ ≤ 34°, consistent with other bacteria (Attard et al., 2012).~~ Hoose and Möhler (2012) reported the ice nucleation active surface site (INAS) density of various bacteria at -5 °C ($10^{2.5}$-10^{10} m^{-2}). Using CNT, we fitted a contact angle to their data, resulting in the range of 32° ≤ $\theta_{bacteria}$ ≤ 34°.

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

Response: Attard et al. (2012) did not investigate whether or not the observed pH effect was reversible. Based on other studies (Schmid et al., 1997; Turner et al., 1990), it can be concluded that denaturation of IN protein's agglomerates (polymers) occurs at pH below 4.5, indicating that IN activities are supposed to be reversible at least above pH 4.5. We add at the end of section 2.4.2 at line 654:

Denaturation of IN protein's agglomerates (polymers) occurs at pH below 4.5 (Schmid et al., 1997; Turner et al., 1990), suggesting that changes in IN activities due to pH might reversible at least above this pH value.

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

Response: The reason for considering PBAPs as being externally mixed and monodisperse is the simplicity of our model studies. We do not attempt to give quantitative estimates of their radiative forcing in the climate system, but our model sensitivity studies are set up such that we compare results from different model simulations to each other, in order to conclude on the sensitivities to individual aerosol properties. Assuming different PBAP properties such as polydisperse size distributions or internally mixed aerosol might change the numbers shown in our figures but not the relative changes due to the variation of one aerosol parameter at a time. We clarify this at line 134:

By means of process models (***Section 3***), we explore in a simplistic way the relative importance of these PBAP properties and ageing processes for the effects depicted in ***Figure 1*** (***Section 4***). Our model sensitivity studies are set up such that we identify trends and their relative importance to show the sensitivities to individual properties and ageing processes that impact PBAP properties in the atmosphere.

In addition, we frame the discussion now more in the context of our process model results and the need of future studies to characterize PBAP properties, rather than making strong claims about global implications. We also emphasize throughout the manuscript that the role of PBAP in the atmosphere for the aerosol direct and indirect effect may be limited due to their small number concentration on a global scale. However, detailed knowledge on PBAP properties that affect their interaction with radiation and water vapor is also essential to properly describe their transport, dispersion and lifetime in the atmosphere, which might affect the global modification of biodiversity and impacts public health. We added this in Section 5 (Conclusions) at line 666:

Given the limitations of our process models in terms of scales, dimensions and parameter spaces, our results should be considered as qualitative, rather than quantitative estimates; the focus of our study is the comparison of relative changes due to various physicochemical parameters.

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

Response: The detailed properties of 'other particles' are listed in Table 2. We used the typical conditions to represent 'other particles', i.e., the majority of typical atmospheric aerosol populations. Since sensitivities on the properties of CCN activation to other aerosol types have been extensively studied, e.g., (Ervens et al., 2005; McFiggans et al., 2005), we did not consider variation of the properties of 'other aerosol' in the current study. Just as stated in our response to the previous comment, the absolute numbers in our figures may change depending on the type and properties of the 'other aerosol', however, the general conclusions on the relative changes will likely not change.

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

Response: In the parcel model, PBAPs first act as CCN on which droplets form. PBAPs are inside the droplets, and then immersion freezing occurs at freezing temperature. The number of other particles might affect N_{CCN} and supersaturation, which in turn affects ice formation. We performed a sensitivity test of the ratio of IWC/LWC to N_{CCN} in our previous study (Ervens et al., 2011), where we concluded that NCCN has likely a small impact in mixed phase clouds.

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.

Response: We agree with the referee that parcel models are of limited value in describing the full evolution of mixed-phase clouds upon the initiation of the Bergeron-Findeisen process, i.e., the full glaciation process followed by precipitation and demise of the cloud. However, they have been proven as useful tools for sensitivity studies that explored the onset of the Bergeron-Findeisen process for various aspects of ice nucleation (Diehl et al., 2006; Eidhammer et al., 2009; Ervens et al., 2011; Khvorostyanov and Curry, 2005; Korolev, 2007; Korolev and Isaac, 2003).

We add the references above and briefly discuss the limitations of the adiabatiac model franework at line 621:

It should be noted that our adiabatic parcel model framework cannot fully represent the complexity of all processes occuring in mixed-phase clouds, such as complete glaciation followed by precipitation and demise of the cloud. However, we rather demonstrate the relative changes in percentage contribution of ice water content (%IWC, solid lines) and liquid water content (%LWC, dashed lines) to total adiabatic water content near the onset of ice nucleation. Thus, we apply our model in a similar way as in previous parcel model studies that explored the onset of the Bergeron-Findeisen process to various aspects of ice nucleation (Diehl et al., 2006; Eidhammer et al., 2009; Ervens et al., 2011; Khvorostyanov and Curry, 2005; Korolev, 2007; Korolev and Isaac, 2003).

In addition, we also modified texts at line 659:

Overall, ~~it can be concluded~~ our model results suggest that a small change in the contact angle due to different types of PBAPs or due to ageing processes might have a large impact on ice nucleation in clouds ~~that chemical processing of bacteria or other BAP that freeze at relatively high temperatures. in the atmospheric for extended periods of time might sufficiently alter their surface to induce a significant change in their IN ability.~~ These differences might translate into feedbacks on other subgrid and dynamical processes in the cloud that amplify or reduce the efficiency of glaciation. However, such processes cannot be further explored in the adiabatic parcel model framework.

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

Response: We agree that it was confusing to use the identical name 'Nother' in three different contexts. We now distinguish the three values of Nother and indicate that they are used in the simulations of CCN, IN and optical properties, respectively:

In line 328: The dry aerosol size distribution covers a size range of 5 nm < $D_{other,\ S(CCN)}$ < 7.7 µm with $N_{other,\ S(CCN)}$ = 902 cm^{-3}, as being typical for moderately polluted continental conditions.

In line 336: We consider an aerosol size distribution with 46 nm < $D_{other,\ S(IN)}$ < 2.48 µm in nine size classes and $N_{other,S(IN)}$ = 100 cm^{-3}, as found in Arctic mixed-phase clouds. The aerosol population includes one additional PBAP size class, which is the only one that includes potentially freezing IN under the model conditions.

In line 354: Note that the concentration of other particles ($N_{other,\ S(opt)}$) would usually increase under haze conditions while we keep $N_{other,\ S(opt)}$ as a constant in the above model (1.4 cm^{-3});

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.

Response: The referee is correct that all values in Eq-5 are strongly time and location dependent. However, we clarify that this estimate is only intended to compare in a relative sense the RFE due to differences in optical properties. We adapted this approach including all values in Eq-5 from Dinar et al. (2007). We also clarified that our results are obtained for relative comparisons, rather than for general or global of radiative forcing calculations at line 500:

The RFE values in ***Table 3*** only represent radiative forcing of a small range of particle sizes and a constant composition and number concentration of other particles; however, the differences (ΔRFE) allow evaluating the relative importance of the various PBAP parameters (N_{PBAP}, D_{PBAP}, m_{PBAP}) in terms of their direct interaction with radiation. A negative ΔRFE implies more scattering and a positive ΔRFE implies more absorption due to the presence of PBAPs.

Note that in the above simulations relatively high concentrations of PBPAs were assumed and should only be used to compare the relative importance of PBAP size and complex refractive index for their optical properties. The properties of PBAPs can vary depending on species of PBAPs and ageing processes. Given that the number concentration of PBAPs is generally small, the direct radiative effect of PBAPs is likely restricted to small spatial scales.

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

Response: See our response to the previous comment. We hope that our text changes and additions above are sufficient to clarify that our intention not to simulate the global effect of PBAPs. Our main idea is to see the difference of RFE (ΔRFE) induced by the addition of PBAPs in a relative sense.

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.

Response: We found more data, added more references, and reworded this paragraph as follows at line 690 (also response 19 to reviewer #1):

The number fraction of PBAPs to total CCN is relatively small (≤ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

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.

Response: We clarified the caption as follows at line 585:

Figure 8. Comparison of the environmental supersaturation within the cloud (S_{env}) as predicted by the parcel model for different updraft velocities (w) to the critical supersaturation (S_c) of PBAPs based on Köhler theory. Results are shown as a function of (a) hygroscopicity parameters κ_{PBAP} and (b) surface tension σ_{PBAP}. Input parameters to the parcel model are listed in ***Table 2***.

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

Response: We removed the word buffering as it may require more definition in this context and may cause confusion as we mostly focus on physicochemical aerosol properties. We intended to use it in the same context as by Feingold and Stevens (2009) who introduced this term to describe the lower sensitivity of cloud properties to aerosol characteristics in the complex aerosol-cloud systems than it is usually suggested if individual aerosol processes or properties were considered separately. We changed the text as follows at line 580:

Our sensitivity studies show ~~once more that under dynamic conditions in clouds buffering reduces the feedbacks of particle composition on supersaturation (Ervens et al., 2005; Stevens and Feingold, 2009).~~ relatively lower sensitivity of cloud properties to particle composition than that predicted based on equilibrium conditions, in agreement with previous sensitivity studies (Ervens et al., 2005). Therefore, previous estimates of surfactant effects on cloud properties that are based on a simplified assumption of equilibrium conditions in clouds (Facchini et al., 1999), led to an overestimate of the role of surfactants on CCN.

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.

Response: We will rephrase the sentence and also refer to the discussion of CCN properties in order to make it clear that immersion freezing is both a function of CCN and IN properties at line 617:

~~Based on these trends, it can be also concluded that processes that change the BAP size (e.g. ΔD_{BAP} by cell generation) are not critical to be included in models to represent the variability of IN property effect on mixed-phase clouds.~~For SPPs and SFPs with D $\leq$ 100 nm, immersion freezing may be limited by the droplet formation on these particles (***Figure S3***). As ice formation is less efficient on non-activated particles ('condensation freezing'), the onset temperatures of freezing is significantly lower. As supermicron particles likely act as CCN under most conditions, this limitation might be smaller for large PBAPs.

In addition, we add a Figure S3 to the supplement:

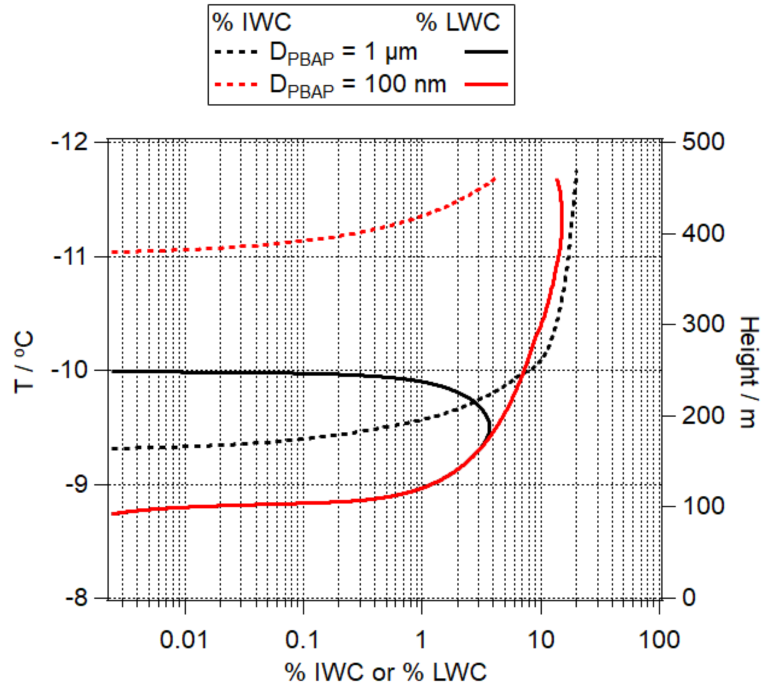

Figure S3. Percentage contribution of ice water content (IWC, dashed lines) and liquid water content (LWC, solid lines) to total adiabatic water content as a function of D_{PBAP}.

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?

Response: We define the onset of the Bergeron-Findeisen process as the point at which LWC% starts to decrease. Accordingly, we can compare the temperatures at which this occurs between the different simulation and ΔT means the change of temperatures of the onset of BF processes due to the change of contact angle.

The T shift is related to the point at which LWC% starts to decrease.

We have added the following at line 606:

We define the onset of the Bergeron-Findeisen process as the temperature, at which the liquid water content fraction starts to efficiently decrease.

We have changed the texts as follows at line 636:

PBAPs exhibit a wide range of contact angles of 4° < θ_{PBAP} < 44° (***Table 1***). ***Figure 10*** compares the predicted relative contributions of %IWC and %LWC to the total adiabatic water content. The comparison of Figures 10a and 10b shows that the onset temperatures of the %LWC decrease are at ~ -7.7 °C (θ_{PBAP} = 4°) and ~ -8.3 °C (θ_{PBAP} = 20°), respectively, i.e. resulting in a difference of ΔT ~0.6 °C. This difference is predicted to be larger (ΔT ~3.3 °C) for PBAPs with θ_{PBAP} = 40°.

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.

Response: We changed the language to more cautious and talked about limitations of adiabatic parcel models (see also our response to the previous comment). Although the feldspars can freeze at very high T, the nucleation site density of feldspars is much lower than the bacteria. We modified the discussion as follows at line 646:

As discussed in ***Section 2***, chemical (e.g., nitration, oxidation, adjustments due to pH) or physical processing of IN surfaces might lead to $\Delta\theta_{PBAP}$ ~1°. In ***Figure 10d***, we show %IWC and %LWC by comparing S_{IN2} and S_{IN9}. The results show that even such a small change of 1° in θ can cause a significant difference in the predicted IWC and LWC evolutions. The temperature, at which the %LWC starts decreasing differs by ΔT ~1.3 °C. Such a change in θ may be induced by pH changes; for example, it was found that Δθ is ~1.5° for bacteria such as *Pseudomonas syringae* when the cells were exposed to solutions of pH 7.0 and 4.1 at temperatures of T > -10 °C. Denaturation of IN protein's agglomerates (polymers) occurs at pH below 4.5 (Schmid et al., 1997; Turner et al., 1990), suggesting that changes in IN activities due to pH might be reversible at least above this pH value.

Similar differences in θ could be also caused due to other processes, such as the oxidation of pollen that lead to Δθ ~1.5° at T ~ -39 °C (Gute and Abbatt, 2018). However, at this much lower temperature, the sensitivity of the frozen fraction to Δθ decreases (Ervens and Feingold, 2013). Overall, our model results suggest that a small change in the contact angle due to different types of PBAPs or due to ageing processes might have a large impact on ice nucleation in clouds. These differences might translate into feedbacks on other subgrid and dynamical processes in the cloud that amplify or reduce the efficiency of glaciation. However, such processes cannot be further explored in the adiabatic parcel model framework.

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

Response: We rephrased the texts about BF process initiation at line 643:

Figure 10. Percentage contribution of ice water content (%IWC, dashed lines) and liquid water content (%LWC, solid lines) total adiabatic water content for θ_{PBAP} of (a) 4°; (b) 20°; (c) 40° and

(d) 37° and 38°. The curves in the first three panels exhibit similar shapes for different temperature ranges, i.e. the Bergeron-Findeisen process starts at different temperatures. The last panel shows that even when the contact angle increases by 1°, the temperature, at which the %LWC fraction starts decreasing, differs significantly.

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.

Response: We agree with the referee that our conclusions based on our model results and describing this figure may have been too strong as our limited process model studies should not be extended to the global scale. However, we would like to keep this figure as it is to our knowledge the first overview of the potential role of biological processes that may affect PBAP properties in the atmosphere. Instead of framing it in the context of radiative forcing, we now focus more on the measurement needs of PBAP properties and processes and their potential (limited) influence on radiative forcing.

We frame its discussion now more in the context of our process model results and the need of future studies to characterize PBAP properties, rather than making strong claims about global implications. We also emphasize throughout the manuscript that the role of PBAPs in the atmosphere for the aerosol direct and indirect effect may be limited due to their small number concentration on a global scale. However, detailed knowledge on PBAP properties that affect their interaction with radiation and water vapor is also essential to properly describe their transport, dispersion and lifetime in the atmosphere, which might affect the global modification of biodiversity and impacts public health.

We rewrote Section 5 (see reply to general comments).

We also modified Figure 11 and caption as follows:

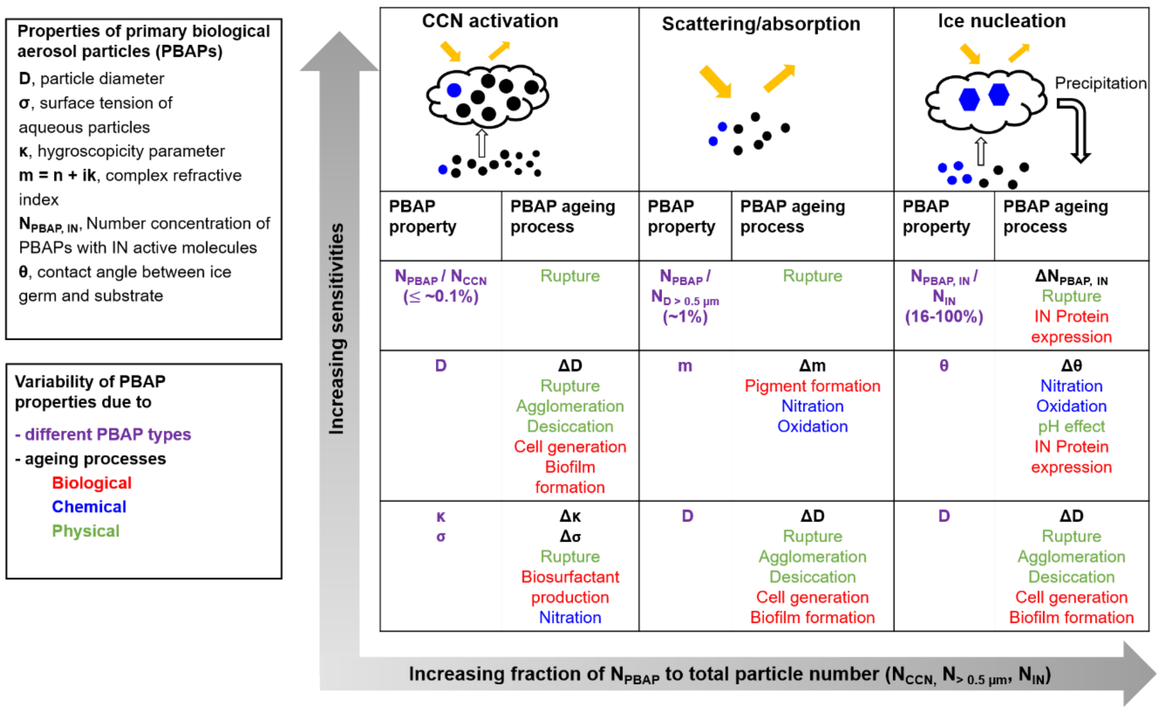

Figure 11. Schematic of PBAP types and ageing processes that affect their aerosol-cloud interactions and optical properties. The bottom arrow shows the increasing fraction of N_{PBAP} to total particles (N_{CCN}, $N_{> 5\ \mu m}$, and N_{IN}, respectively**)**. The left arrow indicates the increasing sensitivity to PBAP properties as predicted based on our process model studies. The various properties might be modified by physical (green), chemical (blue) and biological (red) ageing processes.

Line 615. What about the semi-direct effect?

Response: The referee is correct that generally the absorption of radiation by absorbing organic molecules ('brown carbon') may contribute to the semi-direct effect. However, given the small amounts of light-absorbing material in PBAPs and small mass fraction of PBAP total absorbing mass, the global effect is likely small. In addition, the semi-direct effect is mostly triggered by light-absorbing material (e.g. soot particles) above clouds. Given the large sizes of most PBAP, their concentrations decreases strongly with altitude (Ziemba et al., 2016), thus their impact near cloud top may be small. We have added at line 469:

While generally, light-absorbing organics ('brown carbon') might contribute to the aerosol semi-direct effect (Brown et al., 2018; Hansen et al., 1997), i.e. the impact of aerosol heating on clouds, it seems unlikely that PBAPs significantly have this effect. Given the submicron sizes of most PBAPs, their concentration decreases strongly as a function of altitude (Ziemba et al., 2016) and thus their concentration near cloud tops is likely negligible.

Line 630. See comment on Line 479.

Response: We found more data, added more references, and reworded this paragraph as follows at line 690 (also response 19 to reviewer #1):

The number fraction of PBAPs to total CCN is relatively small ($\leq$ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

Line 636. See comment on Line 240.

Response: The maximum frozen fraction is misleading. We were referring to the fraction of PBAPs that have IN macromolecules. We modified the sentence at 709:

Lab measurements have shown that up to 100% of pollen grains have IN nucleating macromolecules on their surface, whereas only 0.01 to 10% of bacteria express the proteins or other macromolecules that initiate ice nucleation (Failor et al., 2017; Joly et al., 2013; Pummer et al., 2015).

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

Response: We use the term 'cell generation' here in the same way as in our previous study where we referred to it as the combination of cell growth and multiplication (Ervens and Amato, 2020), in agreement with the literature on bacterial processes (Marr, 1991; Price and Sowers, 2004; Si et al., 2017). In this previous exploratory study, we performed an estimate of the potential role of cell generation (i.e. focusing on increase of cell size as we were only concerned with the increase in biological mass) during the atmospheric residence time of a bacteria cell. While the growth of an individual bacteria cell cannot be monitored during its time in the atmosphere, there are several studies that support the hypothesis of growth, metabolic activity and possibly multiplication of cells in the atmosphere (Marr, 1991; Middelboe, 2000; Price and Sowers, 2004; Sattler et al., 2001; Vrede et al., 2002).

The referee is correct that to date, any conclusion on the extent to which such processes affect PBAP properties are speculative. Indeed, they are not comprehensively or at all explored yet in atmospheric models due to the lack of suitable data sets. This lack of knowledge is one of our main reasons to keep Figure 11 in the manuscript as we hope that it may initiate field, lab and model studies. Data from such studies will help to identify the most important processes that modify PBAP radiative properties and adaptive strategies of microorganisms in the atmosphere.

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~~The effect of biological particles and their ageing processes on aerosol radiative properties: Model sensitivity studies~~

Sensitivities to biological aerosol particle properties and ageing processes: Potential implications for aerosol-cloud interactions and optical properties

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Abstract. ~~aerosol particles (BAPs) such as bacteria, viruses, fungi and pollen, represent a small fraction of the total aerosol burden. However due to their unique properties, they have been suggested to be important in for radiative forcing by the aerosol direct and indirect effects. By means of process model studies, we compare the sensitivity of these radiative effects to various physicochemical BAP properties (e.g. number concentration, diameter, hygroscopicity, surface tension, contact angle between ice and particles). Exceeding previous sensitivity studies, we explore not only the variability of these properties among different BAP types, but also the extent to which chemical (e.g. nitration), physical (e.g. fragmentation) and biological (e.g. bacteria cell generation) ageing processes of BAPs can modify these properties. Our model results lead to a ranking of the various properties for the radiative effects: (i) Given that BAPs contribute ~0.1% to total cloud condensation nuclei (CCN) number concentration, their effect on total CCN is likely small. (ii) BAPs number fraction of large particles (diameter > ~0.5 μm) is much higher, resulting in a relatively more important effect on direct radiative forcing. (iii) In mixed-phase clouds at T > -10 °C, BAPs can contribute ~100% to ice nuclei (IN), which makes their role as IN the most important. Our study highlights the need of implementing ageing processes of different BAPs into models as BAP size, CCN and IN activity and optical properties may be sufficiently altered to affect BAP's residence time and survival in the atmosphere. In particular, we suggest the potential role of biological processes, that are currently not included in aerosol models due to the sparsity of comprehensive data, could affect physicochemical BAP properties.~~

Primary biological aerosol particles (PBAPs) such as bacteria, viruses, fungi, and pollen, represent a small fraction of the total aerosol burden. Based on process model studies, we identify trends in the relative importance of PBAP properties, e.g. number concentration, diameter, hygroscopicity, surface tension, contact angle, for their aerosol-cloud interactions and optical properties. While the number concentration of PBAPs likely does not affect total CCN concentrations globally, small changes in the hygroscopicity of submicron PBAPs might affect their CCN ability and thus their inclusion into clouds. Given that PBAPs are highly efficient atmospheric ice nuclei at $T > -10$ °C, we suggest that small changes in their sizes or surface properties due to chemical, physical or biological processing might translate into large impacts on ice initiation in clouds. Predicted differences in the direct interaction of PBAPs with radiation can be equally large between different species of the same PBAP type and among different PBAP types. Our study shows that not only variability of PBAP types, but also their physical, chemical, and biological ageing processes might alter their CCN and IN activities and optical properties to affect their aerosol-cloud interactions and optical properties. While these properties and processes likely affect radiative forcing only on small spatial and temporal scales, we highlight their potential importance for PBAP survival, dispersion and transport in the atmosphere.

1. Introduction

Although primary b~~B~~iological aerosol particles (PBAPs) contribute a small fraction (50 Tg yr^{-1}, with an upper limit of 1000 Tg yr^{-1}) to the total natural global aerosol emissions of ~2900-13000 Tg yr^{-1} (Stocker et al., 2013), they have attracted great interest in the atmospheric science and public health community as they might affect the climate and be responsible for spreading diseases (Asadi et al., 2020; Behzad et al., 2018; Khaled et al., 2020)~~.~~. They consist of bacteria, proteins, viruses, fungi, pollen and other biologically-derived materials with potentially infectious, allergenic, or toxic properties (Fröhlich-Nowoisky et al., 2016). ~~They have attracted great interest in the atmospheric science and public health community as they might affect the climate and be responsible for spreading diseases (Asadi et al., 2020; Behzad et al., 2018).~~

Their mass (Graham et al., 2003; Heald and Spracklen, 2009), number concentrations (Huffman et al., 2013; Matthias-Maser et al., 1999; ~~)(~~Forde et al., 2019a), and fractions (Jaenicke, 2005) can greatly vary depending on ~~the~~ location (Schumacher et al., 2013; Shen et al., 2019; Wei et al., 2016; Yu et al., 2016), time of day (Kang et al., 2012) and other conditions (Graham et al., 2003; Jiaxian et al., 2019; Wu et al., 2016; ~~)(~~Forde et al., 2019b). For example, in the Amazonian rainforest, PBAPs contribute ~20% to the mass of submicron organic aerosol (Schneider et al., 2011). In the semirural ~~an urban~~ area of Mainz in central Europe, the number fraction was 1~~5~~-50% for particles with diameter (D) > 0.4 μm (Jaenicke, 2005). Above the ocean, 1% of particles with 0.2 μm < D < 0.7 μm contain biological materials (Pósfai et al., 1998). Temporal variability of PBAPs was observed exhibiting peaks in the morning, during and after rain (Huffman et al., 2013; Zhang et al., 2019). To the total global PBAP emissions, bacteria contribute 0.4-1.8 Tg yr^{-1}, which is less than 25-31 Tg yr^{-1} by fungal spores (Heald and Spracklen, 2009; Hoose et al., 2010) and 47 Tg yr^{-1} by pollen (Burrows et al., 2009a, 2009b). Although the mass fraction of bacteria is small, their number concentration (~0.001-1 cm^{-3}) (Lighthart and Shaffer, 1995; Tong and Lighthart, 2000) is larger than that of fungal spores (~0.001-0.01 cm^{-3}) and pollen (~0.001 cm^{-3}) (Huffman et al., 2010). The concentration of viruses can reach up to ~0.1 cm^{-3} in indoor air (Prussin et al., 2015) and decreases to ~0.01 cm^{-3} outdoors (Després et al., 2012; Weesendorp et al., 2008). The comparably small size of viruses and bacteria ($D_{viruses}$ ~0.1 μm, $D_{bacteria}$ ~1 μm~~, D_{pollen} ~10 μm~~) enables relatively long residence times of several days in the atmosphere (Burrows et al., 2009a; Verreault et al., 2008).

In numerous recent review articles, it has been suggested that PBAPs can affect radiative forcing in multiple ways (***Figure 1***) (Coluzza et al., 2017; Després et al., 2012; Haddrell and Thomas, 2017; Hu et al., 2018; Šantl-Temkiv et al., 2020; Smets et al., 2016): PBAPs might directly interact with radiation by scattering or absorbing light (***Figure 1a***). While their aerosol direct effect is likely globally small due to low PBAP number concentration (Löndahl, 2014)~~(Löndahl et al., 2014)~~, it may be of greater interest locally and for specific wavelength ranges due to the large size of PBAPs (Myhre et al., 2013). The optical properties of

PBAPs (Arakawa et al., 2003; Hu et al., 2019; Thrush et al., 2010) resemble those of other organic particles as PBAPs are largely composed of proteins and other macromolecules. Accordingly, PBAPs' optical properties can be ascribed to specific organic functional entities such as amino groups or aromatic structures (Hill et al., 2015; Hu et al., 2019). At subsaturated relative humidity (RH) conditions, the hygroscopicity (κ_{PBAP}) determines their ability to take up water (Petters and Kreidenweis, 2007) and thus their equilibrium size, which might affect~~s~~ their direct radiative properties. Their hygroscopicity shows a large range ($0.03 \leq \kappa_{PBAP} \leq 0.25$), which is explained by variation of surface composition due to different types of PBAPs and/or ageing processes (Bauer et al., 2003; Haddrell and Thomas, 2017; Šantl-Temkiv et al., 2020; Sun and Ariya, 2006).

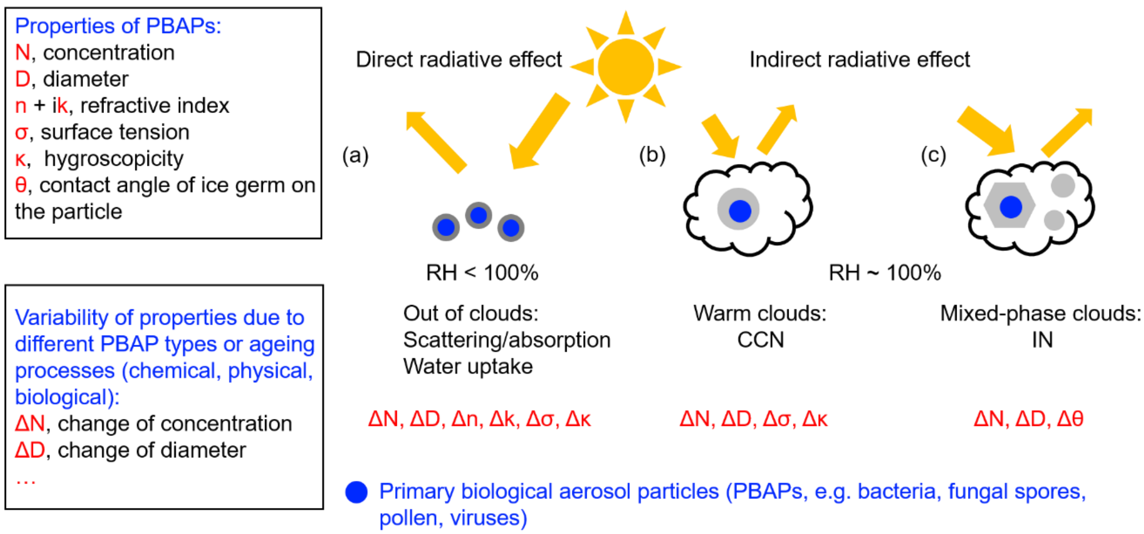


Figure 1. Schematic of the influence of PBAP properties and ageing processes on direct and indirect radiative effects. (a) The direct radiative forcing might be~~is~~ influenced by PBAP concentration (N_{PBAP}), diameter (D_{PBAP}), refractive index ($m_{PBAP} = n + ik$), surface tension (σ_{PBAP}), and hygroscopicity (κ_{PBAP}) ~~affect scattering/absorption of aerosol populations~~ at RH < 100%. (b) N_{PBAP}, D_{PBAP}, ~~surface tension of aqueous particles (~~σ_{PBAP}~~)~~, and ~~hygroscopicity (~~κ_{PBAP}~~)~~ might affect CCN activity and properties of warm clouds. (c) N_{PBAP}, D_{PBAP}, and contact angle of ice germ on the particle~~substrate~~ (θ_{PBAP}) might affect the evolution of mixed-phase clouds.

Convective and precipitating clouds lead to efficient particle redistribution by vertical transport and removal of particles by wet deposition. Therefore, cloud-related physicochemical properties need to be constrained to determine the distribution and residence time of PBAPs in the atmosphere. Since PBAPs often have supermicron sizes, they may act as 'giant CCN' and thus induce early precipitation (Barahona et al., 2010; DeLeon-Rodriguez et al., 2013; Feingold et al., 1999). Based on a global model study, it was concluded that CCN-relevant properties need to be refined in order to further probe their role in the climate system

(Konstantinidis, 2014). In particular pollen rupture leads to a huge increase in the number of subpollen particles (SPPs) (Bacsi et al., 2006; Suphioglu et al., 1992; Taylor et al., 2004; Wozniak et al., 2018). By assuming that one pollen grain releases up to 10^6 SPPs, regional model studies suggested that the resulting SPPs can significantly suppress seasonal precipitation (Wozniak et al., 2018). Several experimental studies have explored the CCN properties of PBAPs and determined their hygroscopicity (κ) (Ariya et al., 2009; Sun and Ariya, 2006). The role of biosurfactant production by bacteria and fungi has been also discussed in the context of their CCN activity since a lower surface tension (σ_{PBAP}) enhances water uptake (Renard et al., 2016). In addition, biosurfactant molecules that are produced by bacteria and fungi, while they reside on leaves or other surfaces, might attach to other particles, thus, increasing their CCN ability as well.

In addition to acting as CCN, some species of ~~plant pathogen~~ bacteria~~, and~~ fungi, and pollen can nucleate ice at ~~T > -10°C~~high temperatures (Hoose and Möhler, 2012; Morris et al., 2004, 2008; Pouzet et al., 2017; Diehl et al., 2001, 2002), which makes them unique in terms of ice nucleation to affect the evolution of mixed-phase clouds at these temperatures (***Figure 1c***). Above vegetated forests (Tobo et al., 2013) and near the surface of the Southern Ocean (Burrows et al., 2013), PBAPs have been shown to contribute significantly to the total abundance of IN.~~:~~ In a high altitude mountain region of the United States, ambient measurements suggest that 16 to ~~-~~76% of IN at -30 °C consist of primary biological material (~~Pratt et al., 2009~~ Prenni et al., 2009); a similar proportion (33%) was reported at -31 to -34 °C in the Amazon basin (~~Prenni et al., 2009~~Pratt et al., 2009).

The radiative impacts of PBAPs, influenced by the physicochemical properties (N_{PBAP}, D_{PBAP}, κ_{PBAP}, σ_{PBAP}, mP_{BAP}, θ_{PBAP}), summarized in ***Figure 1,*** can largely differ on spatial and temporal scales, leading to different conclusions regarding the climatic impacts of PBAPs (Burrows et al., 2009a, 2009b; Hoose et al., 2010; Junge and Swanson, 2008; Konstantinidis, 2014; Sahyoun et al., 2017; Sesartic et al., 2012). These properties are even more variable than represented in current models as PBAPs undergo chemical, physical and biological ageing processes (Coluzza et al., 2017; Deguillaume et al., 2008; Pöschl, 2005; Vaïtilingom et al., 2010).

- *Physical* transformations include agglomeration/fragmentation of cells (Coluzza et al., 2017; Lighthart, 1997; Zhang et al., 2019), coating with organic or inorganic components (Pöschl and Shiraiwa, 2015; Joly et al., 2015), or with solid ice or liquid water (Joly et al., 2013). These processes might alter various physicochemical properties listed in ***Figure 1***. For example, the break-up of pollen or fungi due to rupture can lead to higher number concentrations by several orders of magnitude (Suphioglu et al., 1992; Wozniak et al., 2018).
- *Chemical* transformations include oxidation (Jayaraman et al., 2008; Vaïtilingom et al., 2010), nitration (Franze et al., 2005), oligomerization (Tolocka et al., 2004), degradation of macromolecules (Estillore et

al., 2016), and changes of the protein conformations due to exposure to different pH (Kristinsson and Hultin, 2004). These processes lead to the modification of the protein structures and other macromolecules and thus affect PBAP optical properties (Myhre et al., 2013), CCN activity (Sun and Ariya, 2006), and IN ability (Attard et al., 2012; Kunert et al., 2019).

- *Biological* processes might be initiated by living microorganisms in PBAPs, unlike in other aerosol particles in the atmosphere (Amato et al., 2017; Delort et al., 2017; Joly et al., 2015). Such processes are generally driven by strategies to adapt to the harsh conditions in the atmosphere (e.g., rapid temperature and RH changes, thaw/freeze cycles, humidification/desiccation, UV exposure) (Hamilton and Lenton, 1998; Horneck et al., 1994; Joly et al., 2015; Setlow, 2007) or to limit their atmospheric residence time by initiating precipitation (Hernandez and Lindow, 2019). These processes include nutrient uptake by biodegradation (Khaled et al., 2020), bacteria cell generation that enhances particle size and surface area (Ervens and Amato, 2020), formation of biofilms (extracellular polymeric substances) which enables PBAPs to form aggregates (Monier and Lindow, 2003, 2005; Morris et al., 2008; Sheng et al., 2010), expression of ice-nucleating proteins (Joly et al., 2013; Kjelleberg and Hermansson, 1984), formation of biosurfactants that enhances water uptake (Hernandez and Lindow, 2019; Neu, 1996), desiccation that decreases size of PBAPs (Barnard et al., 2013), formation of pigments (Pšenčík et al., 2004; Fong et al., 2001) enhancing light absorption, fungal spore germination (Ayerst, 1969) or formation of bacteria endospores (Enguita et al., 2003) that increases N_{PBAP}, and metabolism of cellular components (membranes, proteins, saccharides, osmolytes, etc) (Fox and Howlett, 2008; Xie et al., 2010). To date, the uncertainties introduced by these PBAP ageing processes in the estimate of PBAP radiative effects, their atmospheric residence time and distribution can only be assessed qualitatively due to the lack of comprehensive data. However, it may be expected that some of ~~them~~ these ageing processes lead to similar differences in PBAP properties than differences between PBAP types.

In our study, we give a brief overview of the PBAP properties in ***Figure 1*** and summarize which *chemical*, *physical* and *biological* processes may alter these properties (***Section 2***). By means of process models (***Section 3***), we explore in a simplistic way the relative importance of these PBAP properties and ageing processes for the effects depicted in ***Figure 1*** (***Section 4***). Our model sensitivity studies are set up such that we identify trends and their relative importance to show the sensitivities to individual properties and ageing processes that impact PBAP properties in the atmosphere. The results of our sensitivity studies allow a ranking of the importance of the various PBAP properties and processes in terms of their ~~radiative impacts~~ aerosol-cloud interactions and optical properties (***Section 5***). Finally, we give some guidance on the need of future laboratory, field and model studies to more accurately describe potential radiative effects, distribution and residence time of PBAPs in the atmosphere.

2. Physicochemical properties and processes of PBAPs

Literature data on physicochemical parameters of PBAPs are summarized in ***Table 1***. It is not our goal to repeat exhaustive reviews on these individual properties; for this, we refer to previous overview articles (Bauer et al., 2003; Coluzza et al., 2017; Deguillaume et al., 2008; Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Hoose and Möhler, 2012; Huffman et al., 2020; Šantl-Temkiv et al., 2020). We rather aim at using characteristic orders of magnitude of these properties as input data to our process models (***Section 3***). Therefore, we only give a brief overview on the ranges and variability of these properties for different PBAP types and due to various ageing processes.

2.1 PBAP number size distribution parameters (N_{PBAP} and D_{PBAP})

The number concentration (N_{PBAP}) of most PBAP types is in the range of $0.001 \leq N_{PBAP} \leq 0.1$ cm^{-3} (***Table 1***). The number concentration of bacteria is higher than that of fungal spores and pollen although the mass concentration of bacteria is lower (Burrows et al., 2009a; Heald and Spracklen, 2009; Hoose et al., 2010). N_{PBAP} can vary by about three orders of magnitude among different ecosystems, locations, seasons, and time of the day (Huffman et al., 2010, 2020; Matthias-Maser et al., 2000a, 2000b; Schumacher et al., 2013). The PBAP diameter (D_{PBAP}) covers a broad range of 0.01 µm $\leq D_{PBAP} \leq$ 100 µm. This parameter usually refers to the mass equivalent diameter, which is the diameter of a sphere with the same mass as a non-spherical PBAP. The size depends on the type of PBAPs, and on changes due to biological and physical processing. Viruses are reported to be the smallest PBAPs (0.01 µm $\leq D_{viruses} \leq$ 0.3 µm) while pollen is the largest (5 µm $\leq D_{pollen} \leq$ 100 µm) (***Table 1***). Biological processing, such as cell generation, might increase the size of particles by producing secondary biological aerosol mass (Ervens and Amato, 2020; Sattler et al., 2001). Typical bacterial cell generation rates are in the range of 0.1 to 0.9 h^{-1} (Ervens and Amato, 2020). Efficient generation in the atmosphere is assumed to be largely restricted to the time of cell exposure to liquid water (i.e., in-cloud). With an average atmospheric residence time of ~1 week (Burrows et al., 2009b) and an average in-cloud time fraction of ~15% (Lelieveld and Crutzen, 1990), it can be estimated that the generation time scale of bacteria cells in the atmosphere is on the order of ~20 h. Thus, for example, $D_{bacteria}$ may increase from 1 µm to 2 µm after one week in the atmosphere assuming a generation rate of 0.3 h^{-1}. Other rates, such as the cell growth, are usually much smaller (Marr, 1991; Middelboe, 2000; Price and Sowers, 2004; Sattler et al., 2001; Vrede et al., 2002), and thus, contribute less efficiently to a change in D_{PBAP}. In addition, the formation of extracellular polymeric substances might lead to the formation of biofilms, which increase PBAP size by forming agglomerates (Monier and Lindow, 2003, 2005). Agglomerate formation might be also described as a physical process, when PBAPs (e.g. bacteria) attach to other particles (e.g. dust) (Després et al., 2012; Lighthart, 1997), which can result in particle sizes on the order of ~10 µm. At high RH and during precipitation or thunderstorms, pollen absorb water and one pollen grain can release ~10^3

SPPs due to osmotic pressure (Grote et al., 2001; Suphioglu et al., 1992).This process can result in fragments with diameter of 1-4 µm and number concentrations of N_{SPP} ~0.1 cm^{-3} during thunderstorms (Zhang et al., 2019). These concentrations correspond to ~1 to 25 ng m^{-3} (D_{SPP} < 2 µm) (Miguel et al., 2006). Laboratory chamber measurements have shown that SPPs from rupture of fresh birch pollen or grass pollen have diameters of in the range of 0.03 to 4.7 -µm (Taylor et al., 2002, 2004). Recent laboratory measurements suggest that also fungal spores can rupture, resulting in subfungi particles (SFPs) with D_{SFP} of 0.03 to 0.9 µm after exposure to high relative humidity (China et al., 2016). Ambient measurements suggest N_{SFP} of 150 to 455 cm^{-3} (10 nm < D_{SFP} < 100 nm) after rainfall; observed peaks in aerosol size distributions at 20 nm < D_{SFP} < 50 nm which frequently appeared 1.5 days after rain events were ascribed to such rupture events (Lawler et al., 2020).

2.2 Optical properties of PBAPs: Complex refractive index (m_{PBAP} = n + ik)

The scattering and absorption of particles are commonly described by the refractive index m_{PBAP} with real part (n_{PBAP}) and imaginary parts (k_{PBAP}) that depend on the chemical composition and wavelength of irradiation. Arakawa et al. (2003) reported $1.5 \leq n_{PBAP} \leq 1.56$ and $3 \cdot 10^{-5} \leq k_{PBAP} \leq 6 \cdot 10^{-4}$ for bacteria (*Erwinia herbicola*) in the wavelength range of 0.3-2.5 µm. Other groups found a broader range of n and k (***Table 1***) for different ~~types of~~ PBAPs and irradiation wavelengths (Hu et al., 2019; Thrush et al., 2010). The imaginary part can vary by three orders of magnitude for different PBAP types (Hu et al., 2019). Hill et al. (2015) showed that the refractive index of PBAPs can be estimated based on the chemical composition. They reported 1.59 + i0.045 for *Bacillus vegetative* cells at 0.266 µm. Also PBAP shape (e.g. core-shell structure, hexagonal grids, and barbs), as it has been demonstrated for pollen, influences the optical properties (Liu and Yin, 2016). Due to the similarity of the molecular structure of organic macromolecules (e.g. proteins) and secondary organic aerosols (SOA), it can be likely assumed that nitration might alter the PBAP refractive index similar to that of SOA. Experimental results show $1.528 \leq n \leq 1.576$ and $0 \leq k \leq 0.02$ for fresh SOA in the wavelength range of 0.3-0.56 µm; after nitration, the real part increases to $1.549 \leq n \leq 1.594$ and the imaginary part increases to $0.0002 \leq k \leq 0.04$ (Liu et al., 2015).

Table 1. Physicochemical properties of various types of PBAPs and their changes due to physical, chemical and biological ageing processes based on literature data.

PBAP types	**Physicochemical properties**					
	Concentration N~~BAP~~ (cm^{-3})	Diameter D~~BAP~~ (µm)	Complex refractive index m (λ) = n + ik ~~Hygroscopicity κ~~	~~Complex refractive index m_{BAP} (λ) = n + ik~~ Hygroscopicity κ	Surface tension σ (mN m^{-1})	~~IN active number fraction~~ Number fraction of PBAPs with IN	Contact angle θ~~BAP~~ (°)

						active molecules	
Bacteria	0.001-1 (1) ~~0.01-1.4 (2)~~	1 (17) 0.6-7 (18)	n: 1.5-1.56, k: $3\cdot10^{-5}$-$6\cdot10^{-4}$ (24). n: 1.5-1.56, k: 0-0.04 (25). n: 1.25-1.87, k: 0-0.5 (26)~~0.11-0.25 (11)~~	~~n: 1.5-1.56, k: $3\cdot10^{-5}$-$6\cdot10^{-4}$ (17).~~ ~~n: 1.5-1.56, k: 0-0.04 (18).~~ ~~n: 1.25-1.87, k: 0-0.45 (19)~~0.11-0.25 (27)	25, 30, 55, 72 (35~~20~~)	~0.1%, ~1%, ~10% (36~~21~~)	32-34 (39~~24~~);~~.~~ 4-20 (40~~25~~). 28, 33, 44 (41~~26~~)
Fungal spores	0.001-0.01 (2~~3~~)	3-5 (4); 1-30 (5);	n: 1.25-1.75, k: 0-0.32 (26)				30-33 (42~~27~~)
Subfungi particles (SFPs)	150-455 (3)	0.01-0.1 (3); 0.02-0.05 (3); 0.03-0.9 (19)					
Fern spores	10^{-5} (4)	1-30 (4)					
Pollen	0.001 (5)	5-100 (20~~9~~)	~~0.03-0.073 (12). 0.036-0.048 (13). 0.05-0.1 (14). 0.08-0.17 (15). 0.14-0.24 (16)~~n:1.3-1.75, k: 0.01-0.2 (26)	0.03-0.073 (28); 0.036-0.048 (29); 0.05-0.1 (30); 0.08-0.17 (31)		~100% (37,38~~22, 23~~)	14-30 (40~~25~~);~~.~~ 15, 16.3 (43~~28~~)
Subpollen particles (SPPs)~~Pollen fragments~~	0.1 (6)	1-4 (6); 0.03-4 (21); 0.12-4.67 (22)		0.14-0.24 (32); 0.12-0.13 (33); 0.1-0.2 (34)			
Viruses	0.01 (4)	0.01-0.3 (4) 0.04-0.2 (23~~10~~)					
Ambient PBAPs	0.1-1 (7); 1-8 (8)	> 0.4 (7,8)					
Ambient PBAPs	0.2-1.2 (9); 0.04-0.13 (10); 0.012-0.095 (11); 0.01-1.4 (12); 0.57-3.3 (13); 0.1-0.43 (14); 0.02-0.09 (15); 0.005-0.5 (16)	> 1 (9-16)					

		Ageing processes of PBAPs	
	Physical ageing	Chemical ageing	Biological ageing
Bacteria	Agglomeration: $\Delta D_{BAP} > 0$, $\Delta N_{BAP} < 0$ (18)	Nitration: Δn_{BAP} > 0~~~0.02~~, Δk_{BAP} >0 ~~~0.03~~ (44~~29~~);~~.~~ Nitration: $\Delta\theta_{BAP}$ ~1° (41~~26~~). pH changes:	Biosurfactant production: $\sigma_{BAP} < 0$ (35~~20~~);~~.~~ Biofilm formation: $\Delta D_{BAP} > 0$ (45~~30~~);~~.~~

		$\Delta\theta_{\cancel{BAP}} \sim 1.5°$ (41~~26~~).	Endospore formation: $\Delta N_{\cancel{BAP}} > 0$ (46~~31~~);~~.~~ Cell generation: $\Delta D_{\cancel{BAP}} > 0$ (47~~32~~);~~.~~ Desiccation: $\Delta D_{\cancel{BAP}} < 0$ (48~~33~~);~~.~~ Pigment formation ~~(34, 35)~~: $\Delta k > 0$ (49,50); IN protein expression: $\Delta\theta < 0$ (no data yet).~~.~~
Fungi	Rupture: $\Delta D < 0$, $\Delta N > 0$ (3,19)		Biosurfactant production: $\sigma_{\cancel{BAP}} < 0$ (35~~20~~);~~.~~ Germination: $\Delta N_{\cancel{BAP}} > 0$ (49~~34~~) ;~~.~~ Desiccation: $\Delta D_{BAP} < 0$ (48~~33~~).
Pollen	Rupture: $\Delta D_{\cancel{BAP}} < 0$, $\Delta N_{\cancel{BAP}} > 0$ (6,21,22)	Oxidation: $0.5 \le \Delta\theta_{\cancel{BAP}} \le 0.8°$ (43~~28~~)	

(1) Total bacteria, Tong and Lighthart et al., 1999; ~~(2) Under haze conditions in Beijing, Wei et al., 2016. (3) Elbert et al., 2007; (4) Després et al., 2012; (5) blooming times, Huffman et al. 2010; (6) thunderstorm times, Zhang et al., 2019; (7) Burrows et al., 2009a; (8) Lighthart 1997; (9) Pöhlker et al., 2013; (10) Verreault et al., 2008; (11) Lee et al., 2002; (12) Pope et al. 2010; (13) Tang et al., 2019; (14) Chen et al., 2019; (15) Griffiths et al., 2012 ; (16) pollen kit, Prisle et al., 2019; (17) Arakawa et al., 2003; (18) Thrush et al., 2010; (19) Hu et al. 2019; (20) Renard et al., 2016; (21) T ~-10 °C, immersion freezing, *Pseudomonas syringae* bacteria, *Pseudoxanthomonas* sp., *Xanthomonas* sp., Joly et al., 2013; (22) deposition freezing for pollen, Diehl et al., 2001; (23) immersion and contact freezing for pollen, Diehl et al., 2002; (24) Hoose and Möhler, 2012; (25) Chen et al., 2008; (26) immersion freezing for *Pseudomonas syringae*, and *Pseudomonas fluorescens*, Attard et al., 2012; (27) immersion freezing for fungi, Kunert et al., 2019; (28) deposition freezing of silver birch and grey alder pollen, Gute and Abbatt, 2018; (29) nitrated SOA to represent nitrated BAP, Liu et al., 2015; (30) Morris et al., 2008; (31) Enguita et al., 2003; (32) Ervens and Amato, 2020; (33) Barnard et al., 2013; (34) Pšenčík et al., 2004; (35) Fong et al., 2001.~~ (2) Elbert et al., 2007; (3) After rainfall, Lawler et al., 2020; (4) Després et al., 2012; (5) blooming times, Huffman et al. 2010; (6) thunderstorm times, Zhang et al., 2019; (7) Based on protein dyes, Lake Baikal, Russia, Jaenicke, 2005; (8) Based on protein dyes, Mainz, Germany, Jaenicke, 2005; (9) In the Amazon, Whitehead et al., 2016; (10) In the Amazon, Huffman et al., 2012; (11) Puy de Dôme, Gabey et al. 2013; (12) In megacity Beijing, China, Wei et al., 2016; (13) In Megacity Nanjing, China, Yu et al., 2016; (14) High altitude, Ziemba et al., 2016; (15) High altitude, Perring et al. 2015; (16) High concentration observed during and after rain, Huffman et al., 2013; (9) to (16) are based on autofluorescence of PBAPs; (17) Burrows et al., 2009a; (18) Lighthart 1997; (19) China et al., 2016; (20) Pöhlker et al., 2013; (21) Taylor et al., 2004; (22) Taylor et al., 2002; (23) Verreault et al., 2008; (24) Arakawa et al., 2003; (25) Thrush et al., 2010; (26) Hu et al. 2019; (27) Lee et al., 2002; (28) Pope et al. 2010; (29) Tang et al., 2019; (30) Chen et al., 2019; (31) Griffiths et al., 2012 ; (32) pollenkitt, Prisle et al., 2019; (33) Mikhailov et al., 2019; (34) Mikhailov et al., 2020; (35) Renard et al., 2016; (36) T ~-10 °C, immersion freezing, *Pseudomonas syringae* bacteria, *Pseudoxanthomonas* sp., *Xanthomonas* sp., Joly et al., 2013; (37) deposition freezing for pollen, Diehl et al., 2001; (38) immersion and contact freezing for pollen, Diehl et al., 2002; (39) Hoose and Möhler, 2012; (40) Chen et al., 2008; (41) immersion freezing for *Pseudomonas syringae*, and *Pseudomonas fluorescens*, Attard et al., 2012; (42) immersion freezing for fungi, Kunert et al., 2019; (43) deposition freezing of silver birch and grey alder pollen, Gute and Abbatt, 2018; (44) nitrated SOA (toluene as precursor) to represent nitrated BAP, Liu et al., 2015; (45) Morris et al., 2008; (46) Enguita et al., 2003; (47) Ervens and Amato, 2020; (48) Barnard et al., 2013; (49) Pšenčík et al., 2004; (50) Fong et al., 2001.

2.3 PBAP Properties relevant for CCN activation

2.3.1 Hygroscopicity (κ_{PBAP}) of PBAPs

The hygroscopicity determines the PBAP hygroscopic growth factor (gf, as the ratio of wet to dry particle diameter) at subsaturated RH conditions and their CCN activity; it is usually expressed as the hygroscopicity parameter κ (Petters and Kreidenweis, 2007). Lee et al. (2002) reported gf = 1.16 for *Bacillus subtilis* ~~bacteria~~ and gf = 1.34 for *Escherichia coli* ~~bacteria~~ at RH ~85%. Based on these growth factors, $\kappa_{bacteria} = 0.11$ and $\kappa_{bacteria} = 0.25$ for these bacteria can be calculated. The hygroscopicity of pollen is similar to that of bacteria: The κ value of intact pollen grains falls into the range of $0.03 \leq \kappa_{pollen} \leq 0.17$ (Chen et al., 2019; Pope, 2010; Tang et al., 2019), ~~in agreement with κ of pollen kitts on the surface of pollen ($0.14 \leq \kappa_{pollen} \leq 0.24$) (Prisle et al., 2019).~~ pollenkitts (which are parts of pollen surface) and SPPs (which are fragments after rupture) are slightly more hygroscopic ($0.14 \leq \kappa_{pollenkitt} \leq 0.24$, $0.1 \leq \kappa_{SPP} \leq 0.2$) (Mikhailov et al., 2019; Prisle et al., 2019; Mikhailov et al., 2020) than intact pollen grains, which can be explained by the nonuniform composition of pollen (Campos et al., 2008).

2.3.2 Surface tension (σ_{PBAP}) of PBAPs

In most model studies that explore CCN activation, it is assumed that particles have a surface tension close to that of water (σ_{water} = 72 mN m^{-1}). This assumption is likely justified under many conditions due to the strong dilution of internally mixed aerosol particles near droplet activation. There are numerous studies that postulate that surfactants in aerosol particles might influence the surface tension sufficiently to significantly change their CCN activity (Bzdek et al., 2020; Facchini et al., 1999; Lowe et al., 2019; Nozière et al., 2014). These surfactants are usually assumed to have natural sources such as the ocean surface (Gérard et al., 2019; Ovadnevaite et al., 2017). Another source of surfactants might be living microorganisms that produce biosurfactants which enhance surface hygroscopicity and decrease surface tension (Akbari et al., 2018). These biosurfactants might not only be associated with PBAPs themselves as they are deposited on surfaces (e.g. leaves) where they can be taken up by other particles. Renard et al. (2016) reported that 41% of tested strains actively produce surfactant with σ_{PBAP} < 55 mN m^{-1} and 7% of tested strains can produce extremely efficient biosurfactants with σ_{PBAP} < 30 mN m^{-1}. All of these tested strains were collected and isolated in cloud water samples. The most efficient biosurfactants (σ_{PBAP} < 45 mN m^{-1}) are ~~mostly~~ produced by *Pseudomonas* and *Xanthomonas* bacteria (78%) and *Udeniomyces* fungi (11%). For these biosurfactants, we fit the following linear approximation based on the experimental data:

$$\sigma_{PBAP} = 89.6 - 2.9 \cdot C_{biosurf} \qquad \text{if } 6 \text{ mg L}^{-1} \leq C_{biosurf} \leq 22 \text{ mg L}^{-1} \tag{1}$$

where σ_{PBAP} is PBAP surface tension in (mN m^{-1}) and $C_{biosurf}$ is the biosurfactant concentration in (mg L^{-1}). Higher and lower biosurfactant concentrations may be approximated with 25 mN m^{-1} and 72 mN m^{-1} for simplicity. ***Equation 1*** implies that the concentration of biosurfactant on the surface is the same as in the bulk. Recent studies suggest that the surface concentration of surfactants is higher than the bulk concentration (Bzdek et al., 2020; Lowe et al., 2019; Ruehl et al., 2016). Thus, a smaller amount of biosurfactants (‘critical micelle concentration’) than suggested by ***Equation 1*** might be sufficient to significantly decrease σ_{PBAP}. The biosurfactant concentration depends both on the dilution (amount of water) and on the mass fraction of biosurfactants in the particle. The mass fraction has not been determined for biosurfactants; however, other surfactants have been shown to contribute ~0.1% to the total particle mass (Gérard et al., 2019).

2.4 PBAP p~~P~~roperties relevant for ice nucleation

2.4.1 Number fraction of PBAPs with IN active macromolecules~~particle number fraction~~

In freezing experiments of pollen, it has been demonstrated that all particles freeze at sufficiently low temperatures, i.e. the ~~IN active~~ number fraction of PBAPs that have IN active molecules can be assumed as ~100%. Both condensation and immersion/contact freezing led to frozen fractions of 100% at T = ~~∼~~-18 °C (Diehl et al., 2001) and T = ~~∼~~-20 °C (Diehl et al., 2002), respectively. ~~However, for bacteria such as *Pseudomonas syringae*, the maximum frozen fraction only reaches values of 0.1-10% at T ∼ -10 °C (Joly et al., 2013). This might be explained by the fact that not all of the bacteria cells express the same proteins even if they belong to the same species and the same population. It was observed that bacteria express more IN proteins under stress conditions (Kjelleberg and Hermansson, 1984), as a strategy to reach nutrients after destroying the cells of plants by freezing. However, to date it is not fully understood why in lab experiments some of the bacteria cells show freezing behaviour while others from the same population do not and why individual cells show stochastic behaviour in repeated experiments (Lukas et al., 2020).~~ However, it has been shown that bacteria of the same species and within the same population often exhibit different ice nucleation behavior (Bowers et al., 2009; Failor et al., 2017; Fall and Fall, 1998; Lindow et al., 1978; Morris et al., 2004). -This behavior has been explained by various expression levels of IN-active macromolecules that are located at the cell surface. Under conditions such as phosphate starvation, the expression level might be higher, which is a strategy to reach nutrients after destroying the cells of plants by freezing (Fall and Fall, 1998). For example, only 0.1-10% of *Pseudomonas syringae* cells express IN active macromolecules (Joly et al., 2013). Bacteria from the same population without expression of such molecules did not freeze under the experimental conditions.

2.4.2 Contact angle between substrate and ice (θ_{PBAP})

In agreement with previous studies, we base our discussion on the contact angle as a fitting parameter in the classical nucleation theory (CNT) to parametrize the frozen fraction observed in experiments. ~~Chen et al. (2008) reported 4° ≤ $\theta_{bacteria}$ ≤ 20° and 14° ≤ θ_{pollen} ≤ 30°. Similarly, based on the measurements by Attard et al. (2012), we derived values of 28°, 33°, and 44° for different types of bacteria. θ values for fungi based on the measurements by Kunert et al. (2019) are similar (30°≤ θ_{fungi} ≤ 33°). Gute and Abbatt (2018) performed deposition freezing experiments of pollen; based on their experiments, we fitted θ_{pollen} = 15° for silver birch and θ_{pollen} = 16.3° for grey alder. Hoose and Möhler (2012) reported the ice nucleation active surface site (INAS) density of various bacteria at -5 °C ($10^{2.5}$-10^{10} m^{-2}). INAS implies that freezing occurs deterministically as opposed to stochastic freezing described by CNT. As the sensitivity of ice nucleation to time is generally small compared to other parameters (Ervens and Feingold, 2013), we fitted their data using CNT and obtained a range of 32° ≤ $\theta_{bacteria}$ ≤ 34°, consistent with other bacteria (Attard et al., 2012).~~ If not reported in the respective experimental studies, we assumed a freezing time of 10 seconds to derive θ from experimental data, in agreement with many experimental conditions (Attard et al., 2012; Gute and Abbatt, 2018; Kunert et al., 2019). All CNT model equations and parameters are identical to those as described by Ervens and Feingold (2012); Hoose and Möhler (2012) discussed different assumptions made for the various variables in the CNT in previous ice nucleation studies. Chen et al. (2008) reported $4° \leq \theta_{bacteria} \leq 20°$ and $14° \leq \theta_{pollen} \leq 30°$. Based on the measurements by Attard et al. (2012), we derived values of 28°, 33°, and 44° for different species of bacteria. θ values for fungi based on the measurements by Kunert et al. (2019) are similar ($30° \leq \theta_{fungi} \leq 33°$). Gute and Abbatt (2018) performed deposition freezing experiments of pollen; based on their experiments, we fitted $\theta_{pollen} = 15°$ for silver birch and $\theta_{pollen} = 16.3°$ for grey alder. Hoose and Möhler (2012) reported the ice nucleation active surface site (INAS) density of various bacteria at -5 °C ($10^{2.5}$-10^{10} m^{-2}). Using CNT, we fitted a contact angle to their data, resulting in the range of $32° \leq \theta_{bacteria} \leq 34°$.

Chemical processes (e.g. nitration) can change the molecular surface of PBAPs by e.g., adding nitro groups to tyrosine residues of proteins (Estillore et al., 2016), which can alter the IN activity. Attard et al. (2012) measured the cumulative fraction of IN among a population of bacteria before and after nitration for 16-18 h. The residence time of aerosol particles in the atmosphere is from hours to weeks, which means that the experimental nitration times might be a realistic time scale. Based on these data, we calculated that the contact angle increased by ~1° after nitration for some bacteria. In contrast, Kunert et al. (2019) reported that protein nitration does not influence the cumulative fraction of IN for 65 species of fungi investigated. In order to study the oxidation effect, Gute and Abbatt (2018) exposed pollen to OH radicals and measured the cumulative frozen fraction of pollen in terms of deposition freezing. We calculated that the contact angle increased by $\sim 0.5° \leq \Delta\theta_{pollen} \leq 0.8°$ after oxidation. While experimental conditions are often optimized so that a large fraction of particles become nitrated or oxidized, only a small fraction of ambient proteins (~0.1%) have been found to be nitrated (Franze et al., 2005). In addition, Attard et al., (2012) showed that a decrease of pH from 7.0 to 4.1, led to a decrease of the cumulative fraction of IN of *P. syringae* (32b-74) from 10^{-2} to 10^{-8} at T = -4 °C. This change can be described by an increase of θ from 28.7° to 30.3° ($\Delta\theta_{bacteria}$ ~1.6°). *P. syringae* (CC0242), Snomax®, and *P. fluorescens* exhibited similar increases of $\Delta\theta_{bacteria} \sim 1.5°$ for the same change in pH.

3. Model description

3.1 Box model: Scattering/absorption of wet particles at RH < 100% calculated by Mie theory

A box model was used to calculate total scattering/absorption based on Mie theory (Bohren, 1983) for a constant aerosol distribution at different RH. Water uptake by particles is calculated based on Köhler theory. Mie theory is applied to calculate total scattering and absorption of the wet aerosol population as a function of D, N, and m at different wavelengths (λ). The input aerosol size distribution is based on ambient measurements by an ultraviolet aerodynamic particle sizer (UV-APS) in central Europe (Zhang et al., 2019) that cannot detect particles with D < 0.5 µm. At $\lambda \geq 300$ nm, the particles with D > 3 µm interact with light by geometric scattering, rather than Mie scattering. Therefore, we only consider particles with diameters of 0.5 µm < D < 2.8 µm in 24 size classes to represent ambient aerosol particles relevant for our study with a concentration of $N_{other, S(opt)} = 1.4$ cm^{-3}. Thus, the simulations focus on PBAPs in this size range and exclude smaller (e.g. viruses, SFPs or SPPs) and larger (e.g. pollen grains) particles. We consider one additional PBAP size class in the model with specific parameters (N_{PBAP}, D_{PBAP}, m_{PBAP}, κ_{PBAP}, σ_{PBAP}).

Calculations are performed for RH of 10% and 90%, i.e. for different PBAP growth factors. In a series of sensitivity studies of optical simulations (S_{opt1} to S_{opt13}; ***Table 2***), ~~(S_{opt1} – S_{opt11}; ***Table 2***),~~ we explore the sensitivity of scattering and absorption to N_{PBAP}, D_{PBAP}, κ_{PBAP}, and m_{PBAP} (m_{PBAP} = n + ik). We not only compare model results for properties representing different PBAP types (e.g. $D_{bacteria}$ vs D_{fungal}), but also explore the ranges of property variation due to ageing processes of individual PBAP types e.g., the potential increase of bacteria diameter (ΔD) due to cell generation.

Table 2. Model sensitivity studies assume different physicochemical BAP parameters to investigate their effect on the optical properties (***Section 3.1***), CCN activation (***Section 3.2***) and ice nucleation (***Section 3.3***).

Scattering/Absorption: 0.5 µm < $D_{other, S(opt)}$ < 2.8 µm; $N_{other, S(opt)} = 1.4$ cm^{-3}; $\kappa_{other, S(opt)}$: 0.3. $\sigma_{PBAP} = \sigma_{other, S(opt)} = 72$ mN m^{-1} Composition of other particles: 90% ammonium sulfate + 10% soot					
Simulation	N_{PBAP} (cm^{-3})	D_{PBAP} (µm)	$n(\lambda)_{PBAP}$; $k(\lambda)_{PBAP}$	RH	κ_{PBAP}
S_{opt1}	0	-	-	10%	-
S_{opt2}	0.01	1	*E. herbicola:* 1.5-1.56; $3\cdot10^{-5}$-$6\cdot10^{-4}$		0.25
S_{opt3}	0.1				
S_{opt4}	1				
S_{opt5}	0.1	2			
S_{opt6}		3			
S_{opt7}	1	2			0.03
S_{opt8}					0.25
S_{opt9}				90%	0.03
S_{opt10}					0.25
S_{opt11}			*B subtilis*: 1.25-1.6; 0.001-0.1	10%	
S_{opt12}			Fresh PBAPs: 1.528-1.576; 0-0.02		
S_{opt13}			Nitrated PBAPs: 1.549-1.594; 0.0002-0.04		
Cloud condensation nuclei (CCN): 5 nm < $D_{other, S(CCN)}$ < 7.7 µm; $N_{other, S(CCN)} = 902$ cm^{-3}					
	D_{PBAP} (µm)		Hygroscopicity κ_{PBAP}	Surface tension σ_{PBAP} (mN m^{-1})	
S_{CCN1}	0.5		0.25	72	
S_{CCN2}			0.03		
S_{CCN3}	0.1		0.25		
S_{CCN4}			0.03		
S_{CCN5}	0.05		0.25		

S_{CCN6}		0.03		
S_{CCN7}	0.5	0.03		25
S_{CCN8}	0.1			
S_{CCN9}	0.05			
Ice nuclei (IN): 46nm < $D_{other, S(IN)}$ < 2.5 µm; $N_{other, S(IN)}$ = 100 cm^{-3} $N_{PBAP, IN}$ / N_{PBAP} = 10% $\theta_{other, S(IN)}$: 80°				
	N_{PBAP} (cm^{-3})	D_{PBAP} (µm)	Contact angle (θ_{PBAP}) of ice germ	Cloud base temperature (°C)
S_{IN1}	1	1	37°	-8
S_{IN2}	0.01			
S_{IN3}	0.001			
S_{IN4}	0.01	2		
S_{IN5}		5		
S_{IN6}		1	4°	-5.5
S_{IN7}			20°	-6
S_{IN8}			40°	-9
S_{IN9}			38°	-8

3.2 Adiabatic parcel model

3.2.1 CCN activation in warm clouds

An adiabatic parcel model was applied to simulate the formation of warm clouds (Ervens et al., 2005; Feingold and Heymsfield, 1992). The activation of an aerosol population to cloud droplets is described as a function of N, D, κ, and σ. The dry aerosol size distribution covers a size range of 5 nm < $D_{other, S(CCN)}$ < 7.7 µm with $N_{other, S(CCN)}$ = 902 cm^{-3}, as being typical for moderately polluted continental conditions.~~.~~ ~~-~~Similar to the studies on optical properties (***Section 3.1***), we assume that one aerosol size class is composed of biological material, for which we vary D_{PBAP}, κ_{PBAP}, and σ_{PBAP} to explore the role of differences in PBAP types and ageing processes on cloud droplet activation with CCN simulations (S_{CCN1} to S_{CCN9}, ***Table 2***). ~~(S_{CCN1}–S_{CCN9}, ***Table 2***).~~

3.2.2 Ice nucleation in mixed-phase clouds

The adiabatic parcel model as used for the CCN calculations was extended by the description of immersion freezing based on classical nucleation theory (Ervens et al., 2011). At each model time step (1 second), the frozen fraction of PBAPs is calculated; if 1% or more of the IN size class are predicted to freeze in a given time step, a new size class of ice particles is generated in the model, for which ice growth is described. We consider an aerosol size distribution with 46 nm < $D_{other, S(IN)}$ < 2.48 µm in nine size classes and $N_{other, S(IN)}$ = 100 cm^{-3}~~-~~, as found in Arctic mixed-phase clouds. The aerosol population includes ~~and~~ one additional PBAP size class, which is the only one that includes potentially freezing IN under the model conditions. ~~-~~Similar to the analysis by Ervens et al. (2011), we compare the evolution of the ice liquid water contents (IWC and

LWC) expressed in mass fractions [%] whereas 100% corresponds to the total water (ice + liquid + vapor) mixing ratio that is constant under the adiabatic model conditions. Input values of D_{PBAP}, N_{PBAP}, and θ_{PBAP} are varied in IN simulations (S_{IN1} to S_{IN9}) (***Table 2***).~~simulations S_{IN1} to S_{IN9} (***Table 2***).~~

4. Results and discussion

4.1 Sensitivity of optical properties at subsaturated conditions (RH < 100%) to PBAP properties

4.1.1 Influence of concentration (N_{PBAP}) and diameter (D_{PBAP}) on scattering and absorption

As explained in ***Section 3.1***, in the sensitivity studies of optical properties, we consider only particles with D in the same range as λ so that scattering and absorption can be calculated by Mie theory. ~~As ambient aerosol particles also include smaller and larger particles, our conclusions on BAP direct radiative effects should be regarded as the upper limit on total scattering and absorption.~~In ***Figure 2***, we compare the total scattering coefficient for a case without PBAPs (~~N_{BAP} = 0,~~ S_{opt1}) to that predicted for N_{PBAP} = 0.01 cm^{-3} (S_{opt2}), N_{PBAP} = 0.1 cm^{-3} (S_{opt3}) and N_{PBAP} = 1 cm^{-3} (S_{opt4}). At N_{PBAP} = 0.01 cm^{-3}, the effect on total scattering coefficient is negligible. At N_{PBAP} = 0.1 cm^{-3} the total scattering coefficient increases by 15% to 18% in the range of 0.3 μm ≤ λ ≤ 1.5 μm although the number fraction of PBAPs is only 6%. At a higher concentration (N_{PBAP} = 1 cm^{-3}), the total scattering coefficient changes by a factor of 0.5 to 2 depending on λ. Note that the atmospheric concentration of other particles ($N_{other,\,S(opt)}$) might be higher than used in the above model (1.4 cm^{-3}); therefore, the predicted increase of scattering coefficient is likely an overestimate. The absorption coefficient of the total aerosol population does not change (***Figure S1***).

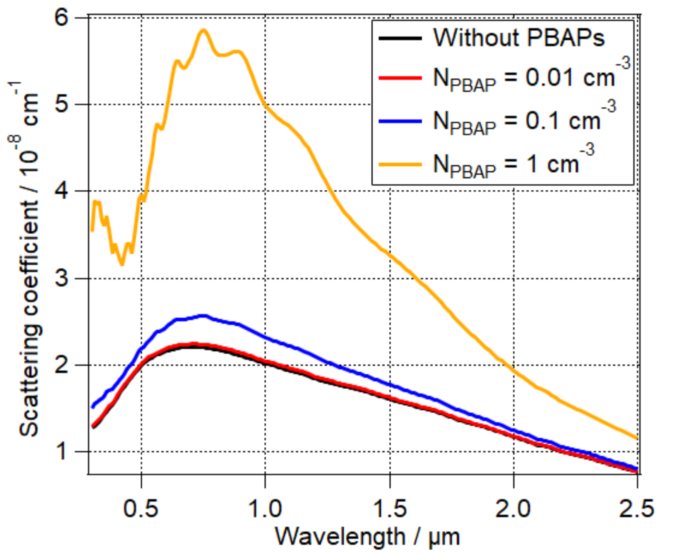


Figure 2. ~~Influence of BAP concentration on t~~Total scattering coefficient for different PBAP number concentrations. The detailed input parameters can be found in ***Table 2***. The black, red, blue, and brown lines correspond to S_{opt1}, S_{opt2}, S_{opt3}, and S_{opt4} in ***Table 2***, respectively.

D_{PBAP} also affects the scattering coefficient of the aerosol population significantly (***Figure 3***). D_{PBAP} = 1 µm (S_{opt3}) and D_{PBAP} = 2 µm (S_{opt5}) can be considered to represent different PBAP types such as bacteria and fungi, respectively, or an aged bacteria cell that has undergone processing by cell generation (Ervens and Amato, 2020). For these assumptions, the scattering coefficient increases depending on λ, with the largest changes of 73% to 100% at λ > l.5 µm when D_{PBAP} increases from 1 µm to 2 µm (S_{opt5}). Larger PBAPs (D_{PBAP} = 3 µm, S_{opt6}) such as SPPs and fungal spores ~~pollen fragments show~~ lead to an increase in the scattering coefficient by a factor of 1.4 to 4.7 depending on λ. The absorption coefficient of the aerosol population remains nearly the same (***Figure S2***).

The results in ***Figure 3*** clearly show that the size of PBAPs needs to be known in order to assess their optical properties. Even a relatively small variation in particle diameter from 1 to 2 µm due to different types or to cell diameter changes (ΔD_{PBAP}) might lead to change in scattering coefficient by 8-100 % depending on λ. Given that the diameter (D_{PBAP}) might vary by four orders of magnitude among different PBAP types, our analysis shows that different sizes for the various PBAP types need to be taken into account when their optical properties are evaluated.

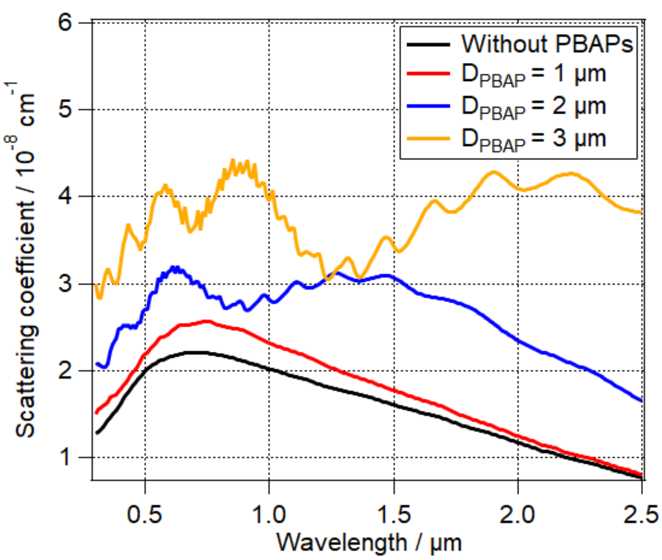
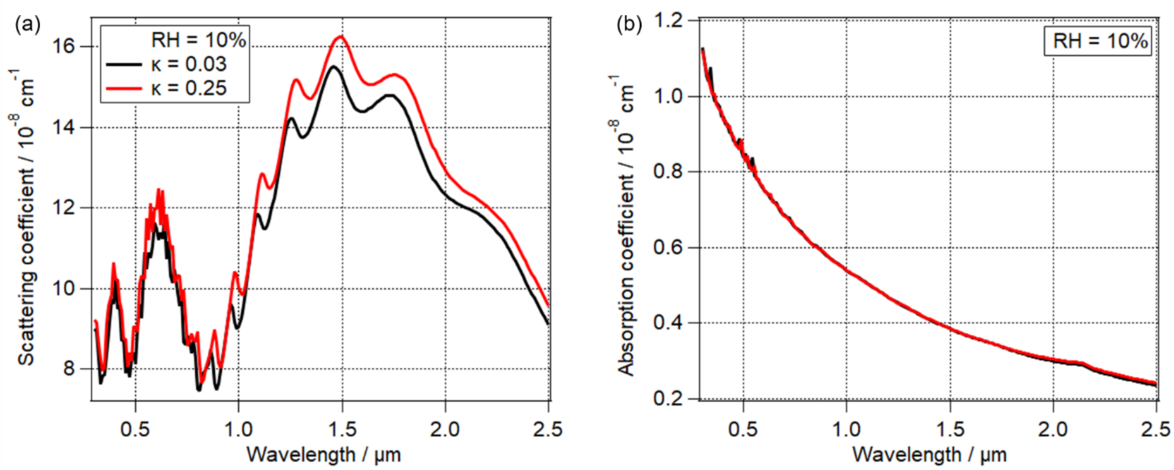


Figure 3. ~~Influence of BAP diameter on (a)~~ Total scattering coefficient for different PBAP diameters~~and (b) absorption coefficient of total particles~~. The detailed input parameters can be found in ***Table 2***. The black, red, blue, and brown lines correspond to S_{opt1}, S_{opt3}, S_{opt5}, and S_{opt6}, respectively.

In our model studies, we make the simplistic assumption of spherical PBAP particles. ~~Electron scanning~~ M~~m~~icroscopic imaging has shown that aerosol particles ~~BAP~~ are not spherical but exhibit a variety of different shapes (Valsan et al., 2015; Wittmaack et al., 2005; ~~)(~~O'Shea et al., 2019). The consequences of the assumptions of spherical versus non-spherical pollen on the derivation of optical properties at a wavelength of 0.65 µm have been recently discussed (Liu and Yin, 2016). The extinction efficiency (sum of scattering efficiency and absorption efficiency) can vary by a factor of one to three for small pollen with D < 4 µm. For larger pollen with D > 5 µm, the extinction efficiency varies by ~25% (Liu and Yin, 2016). ~~While we do not explore sensitivities of BAP geometry, it may be postulated that under atmospheric conditions, i.e. when BAP are wet, they are more spherical than under the experimental dry conditions, and thus effects due to non-sphericity may be reduced.~~ Non-sphericity of particles might translate into the same changes as caused by different particles sizes, which might induce uncertainties including optical depth and surface albedo (Kahnert et al., 2007). These uncertainties on scattering and absorption caused by non-spherical shape might be of comparable magnitude to that caused by the complex refractive index (Yi et al., 2011).

4.1.2 Influence of hygroscopicity (κ_{PBAP}) and surface tension (σ_{PBAP}) on scattering and absorption

As discussed in ***Section 2.3***, the growth factor (gf_{PBAP}) might vary depending on PBAP hygroscopicity (κ_{PBAP}) and surface tension (σ_{PBAP}). ***Figure 4*** shows the influence of κ on scattering and absorption at RH of 10% (S_{opt7}, S_{opt8}) and 90% (S_{opt9}, S_{opt10}). At RH = 10% (S_{opt7}, S_{opt8}), the influence of PBAPs on scattering coefficient of total particles is small (< 19%) and the influence on absorption coefficient is negligible. At high RH = 90%, the water content of particles is significantly higher when $\kappa = 0.25$ as compared to $\kappa = 0.03$. Assuming $\kappa = 0.25$ (S_{opt10}) instead of $\kappa = 0.03$ (S_{opt9}), leads to an increase of the scattering coefficient by 17 to -90% at RH = 90%. Also the absorption coefficient increases by ~40% at $\lambda > 2$ µm. This trend can be explained as the imaginary part of water is higher by three orders of magnitude at λ ~2 µm compared to that at λ ~1 µm -(Kou et al., 1993). It can be concluded that the importance of κ_{PBAP} increases at higher RH, as under these conditions PBAP hygroscopic growth is most efficient.



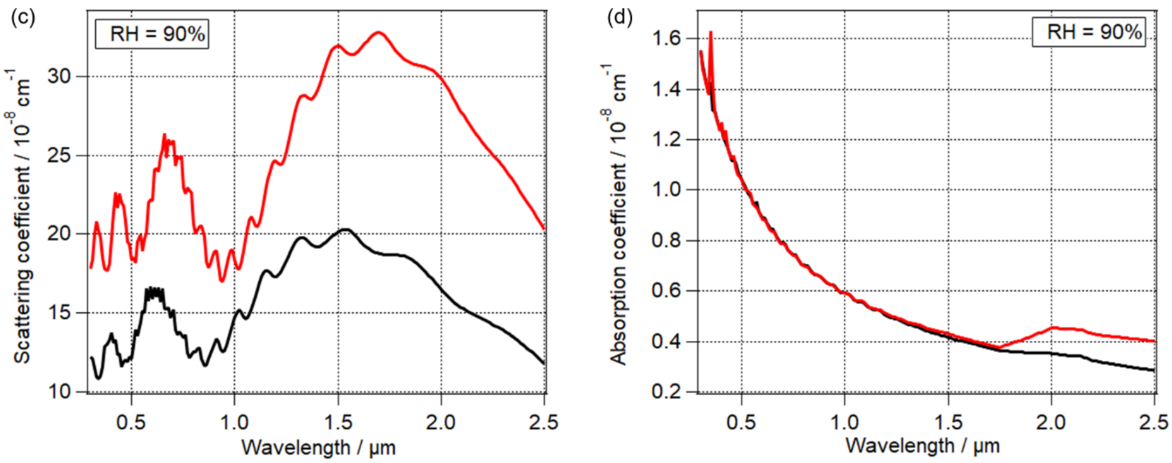


Figure 4. The effect of PBAP hygroscopicity (κ) on (a) scattering coefficient, (b) absorption coefficient of total particles at RH = 10% (S_{opt7}, S_{opt8}), ~~and~~ (c) scattering coefficient, and (d) absorption coefficient of total particles at RH = 90% (S_{opt9}, S_{opt10}). The black lines indicate κ = 0.03 and the red lines indicate κ = 0.25 for all panels.

In addition to hygroscopicity (κ_{PBAP}), we explore the importance of biosurfactants which decrease surface tension of particles (σ_{PBAP}). A lower surface tension leads to a reduced particle curvature which, in turn, enhances the water uptake. Numerically, this is expressed in the Köhler equation:

$$s = \exp\left(\frac{A(\sigma)}{D_{wet}} - \frac{B(\kappa)}{D_{wet}^3}\right) \tag{2}$$

where *s* is the equilibrium water vapor saturation ratio, D_{wet} the wet particle diameter, the first term in the parentheses is the Kelvin (curvature) term which is a function of surface tension (σ_{PBAP}) following ***Equation 3*** and the second term is the Raoult (solute) term which can be parameterized by κ_{PBAP} (Rose et al., 2008) following ***Equation 4***:

$$Kelvin\ term = \frac{A(\sigma)}{D_{wet}} = \frac{4\sigma_{sol}M_{\omega}}{\rho_{\omega}RTD_{wet}} \tag{3}$$

$$Raoult\ term = \frac{B(\kappa)}{D_{wet}^3} = -\ln\frac{D_{wet}^3 - D_s^3}{D_{wet}^3 - D_s^3(1-\kappa)} \tag{4}$$

where σ_{sol} is surface tension of solution droplet (72 mN m^{-1}); M_{ω} is molar mass of water (18 g mol^{-1}); ρ_{ω} is density of water (1 g cm^{-3}); R is the universal gas constant ($8.31 \cdot 10^7$ g cm^2 s^{-2} K^{-1} mol^{-1}); T is the absolute temperature (K); D_{wet} is droplet diameter (cm); and D_s is the diameter of the dry particle (cm).

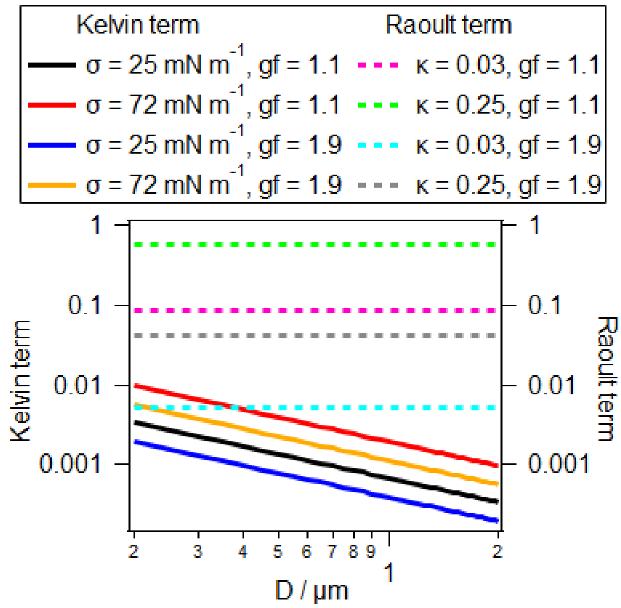


Figure 5. Kelvin term as a function of surface tension (σ_{PBAP}) for the σ range as found for PBAPs (left axis; solid lines). Raoult term as a function of hygroscopicity (κ_{PBAP}) for the range of κ as found for PBAPs (right axis; dashed lines).

The comparison of the two dimensionless terms shows that in most of the cases, the Raoult term exceeds the Kelvin term by at least one order of magnitude. Only for very small PBAPs, i.e. representative for viruses, SPPs or SFPs ~~or bacteria fragments~~ (***Section 2.1***), the curvature term significantly influences *s* (***Figure 5***). Based on this analysis, we can conclude that (bio)surfactants likely do not have a significant impact on the hygroscopic growth of PBAPs. A coating with surfactants might slow down the kinetics of the water uptake by particles (Davidovits et al., 2006). However, since the growth time scales of particles at RH < 100% are usually relatively long, the impact of surfactants on the time scale to reach equilibrium sizes is likely small, leading to a small importance of the effect of surfactant on water uptake and the corresponding optical properties.

4.1.3 Influence of complex refractive index ($m_{PBAP} = n + ik$) on scattering and absorption

The complex refractive index of PBAPs can be explained by their building blocks of various functional groups (Hill et al., 2015). ~~Hu et al. (2019) have measured the complex refractive indices of 12 types of BAP including bacteria, pollen, and spores.~~ Here the complex refractive indices of PBAPs are based on the measurements of *Erwinia herbicola* by Arakawa et al. (2003) and twelve other PBAPs by Hu et al. (2019); the complex refractive indices of 'other particles' in the model are the averaged values based on the volume fractions of ammonium sulfate, soot, and water (***Table 2***). ~~we use *Bacillus subtilis* bacteria and *Lactobacillus acidophilus* bacteria, *Aspergillus oryzae* fungal spores, and lotus pollen as representative BAP types (***Table***~~

~~*1*) to show how the refractive index of different BAP might affect scattering and absorption coefficients of total particles. The scattering coefficient can vary by a factor of two and the absorption coefficient by a factor of five, depending on the wavelength with the largest effects at λ > 2 μm (***Figure 6***).~~The calculated scattering and absorption coefficients of the total particle population are shown in ***Figure 6***. Scattering coefficients for different PBAPs vary by a factor of up to four and the absorption coefficients by a factor of up to six.

The difference of optical properties between bacteria species or fungi species can be larger than that between these two types of PBAPs. Therefore, detailed information on PBAP species is important in order to estimate their direct interaction with radiation (**Section 4.1.4**).

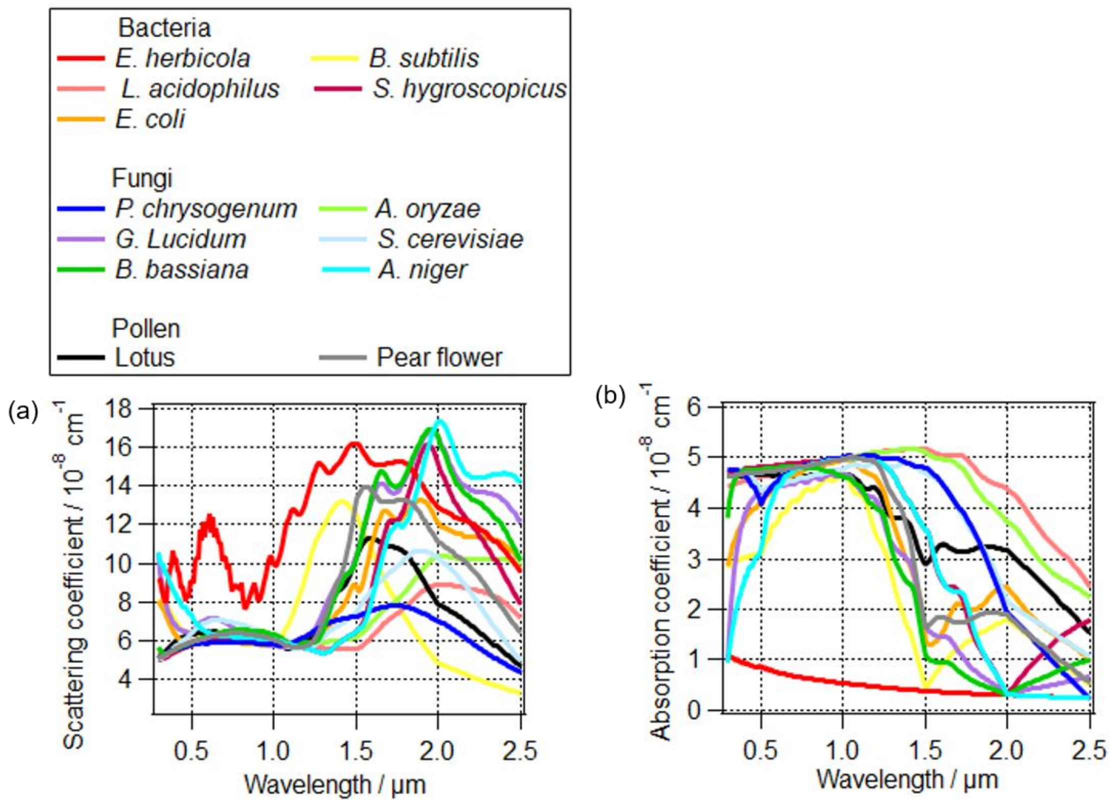


Figure 6. The influence of different ~~types of~~ PBAPs on (a) the scattering coefficient and (b) absorption coefficient of total particles. ~~The black, red, blue, and brown lines correspond to S_{opt11}, S_{opt12}, S_{opt13}, and S_{opt14}, respectively. All of other parameters are assumed to be equal (i.e. D_{BAP}, N_{BAP}, κ_{BAP} and RH).~~ The refractive indices are based on the measurements by Arakawa et al. (2003) and Hu et al. (2019).

In addition to the variability in refractive index due to PBAP types, chemical processing of the ~~macro~~molecules at the PBAP surface might modify the refractive index. It has been shown that nitration of SOA, i.e. the addition of a nitro group, leads to the formation of brown carbon (Moise et al., 2015). Qualitatively, it has been demonstrated that proteins can be nitrated, similar to SOA compounds (Shiraiwa et al., 2012). Due to the lack of data on the change of complex refractive index (Δm) for nitrated proteins in PBAPs, we assume PBAPs have a similar change in the refractive index to that of ~~as in~~ SOA ($S_{opt12\cancel{13}}$ and $S_{opt13\cancel{14}}$). ~~The scattering coefficient can change by up to 20% and the absorption coefficient by a factor of three at λ = 0.42 µm (***Figure 7***). Thus, the variability in scattering/absorption properties of BAP due to Δm caused by nitration is likely smaller than due to Δm caused by different BAP types.~~ After nitration, the scattering coefficient decreases by ~20% in the range of 300 nm < λ < 450 nm and is nearly constant in the range of 460 nm < λ < 560 nm (**Figure 7a**). The scattering coefficient depends non-linearly on the real and the imaginary parts. The absorption coefficient of nitrated PBAPs is higher by 14% to 160% in the range of 300 nm < λ < 540 nm (**Figure 7b**) and is nearly constant in the range of 550 nm < λ < 560 nm. The largest difference (~160%) for absorption coefficient is observed at 440 nm and the smallest difference (~6%) is observed at 560 nm, which can be attributed to the wavelength-dependent change of the imaginary part (Δk) (Liu et al., 2015). The assumptions on Δm made for the simulations shown in ***Figure 7*** are likely an overestimate of the chemical processing of PBAP constituents since (1) experimental conditions are often optimized so that a large fraction of particles is nitrated (Liu et al., 2015), as opposed to ~0.1% of nitrated proteins observed in the atmosphere (Franze et al., 2005), (2) we assume nitration to occur over the whole residence time of particles in the atmosphere while proteins can be nitrated only under conditions of sufficiently high NO_x levels (Shiraiwa et al., 2012), and (3) a rather high concentration of N_{PBAP} = 1 cm^{-3} is considered.

While generally, light-absorbing organics ('brown carbon') might contribute to the aerosol semi-direct effect (Brown et al., 2018; Hansen et al., 1997), i.e. the impact of aerosol heating on clouds, it seems unlikely that PBAPs have a significant contribution to it. Given the supermicron sizes of most PBAPs, their concentration decreases strongly as a function of altitude (Ziemba et al., 2016) and thus their concentration near cloud tops is likely negligible.

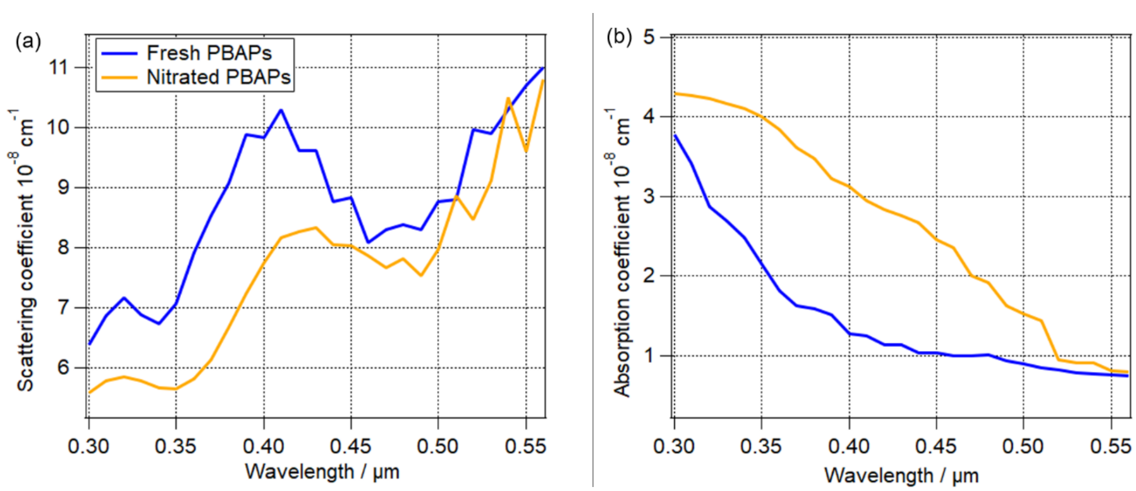


Figure 7. The influence of ~~protein~~ nitration on (a) the scattering coefficient and (b) absorption coefficient of total particles. The ~~black~~ blue and brown lines~~red correspond to~~ indicate fresh PBAPs (S_{opt12})~~15~~ and nitrated PBAPs (S_{opt13})~~16~~, respectively.

4.1.4 Estimate of change of radiative forcing introduced by PBAPs

The direct radiative effect of particles can be expressed in terms of the single scattering albedo (SSA, i.e. the ratio of scattering coefficient to extinction coefficient) and radiative forcing efficiency (RFE, i.e. radiative forcing per unit optical depth) (Dinar et al., 2007; Randles et al., 2004). ~~In order to give an estimate of the local radiative forcing due to BAPs, we applied the same approach as Dinar et al. (2007). The radiative forcing efficiency (~~The RFE ~~, i.e. radiative forcing per unit optical depth)~~ at 390 nm and 532 nm can be calculated as (Dinar et al., 2007):

$$RFE = S_{con}D_{len}(1-A_{cld})T_{atm}^2(1-R_{sfc})^2\left[2R_{sfc}\frac{1-\omega}{(1-R_{sfc})^2}-\beta\omega\right] \tag{5}$$

where S_{con} is the solar constant (1370 W m^{-2}); D_{len} is the fractional day length (0.5); A_{cld} is the fractional cloud cover (0.6); T_{atm} is the solar atmospheric transmittance (0.76), and R_{sfc} is surface albedo (0.15); ω is the single scattering albedo (SSA), which is the ratio of scattering coefficient to extinction coefficient; β is average upscatter fraction, which can be calculated as:

$$\beta = 0.082 + 1.85\mathrm{b} - 2.97b^2 \tag{6}$$

$$b = \frac{1-g^2}{2g}\left[\frac{1}{\sqrt{1+g^2}} - \frac{1}{1+g}\right] \tag{7}$$

where b is the ratio of backscattering to scattering coefficient, g is the asymmetry factor which is assumed as 0.65 as an average of ambient measurements (~0.59-0.72 (Andrews et al., 2006)). The calculated RFE are listed in ***Table 3*** for some of the simulations (input parameters are listed in ***Table 2***). ~~model results of the simulations listed in~~ ~~***Table 2***~~. The first row is the reference with internally mixed ammonium sulfate/soot particles ~~only~~ while PBAPs are absent. As expected, when λ increases from 390 nm to 532 nm, SSA increases due to less efficient absorption in the visible wavelength range (Kirchstetter et al., 2004).

~~With a typical concentration of N_{BAP} = 0.01 cm^{-3} (S_{opt2}), SSA increases both at λ = 390 nm and at λ = 532 nm, which means that BAP have a net cooling effect of ΔRFE = -0.22 W m^{-2} at λ = 390 nm and ΔRFE = -0.15 W m^{-2} at λ = 532 nm, respectively.~~

Table 3. Radiative forcing efficiency (RFE) at 390 nm and 532 nm calculated based on ***Equations 5, 6, and 7*** (Dinar et al., 2007). Some typical conditions are shown here to demonstrate the influence of various ~~properties of~~ PBAP properties such as concentration, size, and complex refractive index.

Simulation	SSA	RFE ($W m^{-2}$)	ΔRFE ($W m^{-2}$)	SSA	RFE ($W m^{-2}$)	ΔRFE ($W m^{-2}$)
	390 nm (ultraviolet)			532 nm (visible)		
S_{opt1} (without PBAPs, reference)	0.643	-0.5	-	0.728	-6.84	-
S_{opt2} (N = 0.01 cm^{-3}, D = 1 µm, $m_{E.\ herbicola}$)	0.646	-0.72	-0.22	0.73	-6.99	-0.15
S_{opt3} (N = 0.1 cm^{-3}, D = 1 µm, $m_{E.\ herbicola}$)	0.668	-2.36	-1.86	0.747	-8.26	-1.42
S_{opt5} (N = 0.1 cm^{-3}, D = 2 µm, $m_{E.\ herbicola}$)	0.738	-7.59	-7.09	0.791	-11.54	-4.68
S_{opt8} (N = 1 cm^{-3}, D = 2 µm, $m_{E.\ herbicola}$)	0.917	-20.94	-20.44	0.927	-21.68	-14.84
S_{opt11} (N = 1 cm^{-3}, D = 2 µm, $m_{B\ subtilis}$)	0.539	7.26	7.76	0.56	5.7	12.54
S_{opt12} (N = 1 cm^{-3}, D = 2 µm, $m_{Fresh\ PBAP}$)	0.868	-17.29	-16.79	0.927	-21.69	-14.85
S_{opt13} (N = 1 cm^{-3}, D = 2 µm, $m_{Nitrated\ PBAP}$)	0.692	-4.15	-3.65	0.909	-20.34	-13.5

The RFE values in ***Table 3*** only represent radiative forcing of a small range of particle sizes and a constant composition and number concentration of other particles; however, the differences (ΔRFE) allow evaluating the relative importance of the various PBAP parameters (N_{PBAP}, D_{PBAP}, m_{PBAP}) in terms of their direct interaction with radiation ~~radiative forcing~~. ~~A decrease in RFE implies less absorption, and thus more cooling of atmosphere (Dinar et al., 2007). ΔN_{BAP} (S_{opt3}) and ΔD_{BAP} (S_{opt4}) have a significant influence on ΔRFE. In addition, ΔRFE at λ = 390 nm is higher than that at λ = 532 nm, implying the increasing importance of BAP in the UV range.~~ A negative ΔRFE implies more scattering and a positive ΔRFE implies more absorption due to the presence of PBAPs.

With a typical concentration of $N_{Erwinia\ herbicola}$ = 0.01 cm^{-3} (S_{opt2}), the SSA increases and the RFE is more negative by 44% and 2% at λ = 390 nm and λ = 532 nm, respectively, as compared to the reference case (S_{opt1} without PBAPs). With a higher number concentration of $N_{Erwinia\ herbicola}$ = 0.1 cm^{-3} (S_{opt3}), the RFE becomes more negative by 228% and 18% at 390 nm and 532 nm, respectively, as compared to the low number concentration $N_{Erwinia\ herbicola}$ = 0.01 cm^{-3} (S_{opt2}). When the diameter increases to D = 2 µm (S_{opt5}), the RFE is more negative by 221% and 40% at 390 nm and 532 nm, respectively, as compared to the D = 1 µm (S_{opt3}). The above results suggest that (1) both the concentration and the size of PBAPs can enhance the RFE significantly and (2) PBAPs affects the optical properties more at the UV wavelength of 390 nm than at the visible wavelength of 532 nm.

All PBAPs for which refractive indices are listed in **Table 1**, show a wavelength dependence on scattering and absorption. The imaginary part (k) varies by three orders of magnitude between different PBAPs (***Table 1***), which makes both the sign and the absolute value of the direct radiative effects of PBAPs uncertain. For example, both *Erwinia herbicola* and *Bacillus subtilis* have been found in the atmosphere (Després et al., 2012). *E. herbicola* is expected to induce more scattering (S_{opt8}) whereas *B. subtilis* is expected to induce more absorption (S_{opt11}).

~~When fungal spores are considered instead of bacteria (i.e., m = n +ik is changed), SSA decreases and RFE even changes from a negative to a positive value (S_{opt13}), resulting in predicted RFE of 7.56 W m^{-2} at λ = 390 nm and 5.69 W m^{-2} at λ = 532 nm, respectively. This might be explained by the strong light absorption (very high k) of *Aspergillus oryzae* fungal spores. Generally, the imaginary part k can vary by three orders of magnitude between different types of BAP (**Table 1**), which makes both the sign and the absolute value of the radiative effects of BAP uncertain.~~ Due to the lack of data of nitrated PBAPs, we used the refractive index of nitrated SOA and fresh SOA (Liu et al., 2015) to represent nitrated PBAPs and fresh PBAPs. Compared to the fresh PBAPs ($S_{opt12\cancel{15}}$), the ~~cooling effect of~~ nitrated PBAPs ($S_{opt13\cancel{16}}$)~~-~~ cause less change of RFE~~decreases~~, which can be explained by the increase of k for nitrated PBAPs due to the formation of brown carbon.

~~These sensitivity studies demonstrate the significant effect of BAP in direct radiative forcing of supermicron particles. The properties of BAP (e.g. N_{BAP}, D_{BAP}, m_{BAP}) can vary depending on species of BAP and ageing processes. The Largest ΔRFE are caused by Δm_{BAP}.~~ Note that in the above simulations relatively high concentrations of PBPAs were assumed and should only be used to compare the relative importance of PBAP size and complex refractive index for their optical properties. The properties of PBAPs can vary depending on species of PBAPs and ageing processes. Given that the number concentration of PBAPs is generally small, the direct radiative effect of PBAPs is likely restricted to small spatial scales.

4.2 Sensitivity of CCN activity to PBAP properties

4.2.1 Influence of PBAP ~~concentration (N_{BAP}) and~~ diameter (D_{PBAP}) on CCN activation

~~N_{BAP} is low compared to the total CCN concentration (Chow et al., 2015; Sun and Ariya, 2006). The upper limit N_{BAP} is on the order of ~1 cm^{-3} (**Table 1**) while the number concentration of CCN is usually in the range of 10s to 1000s cm^{-3} (Ervens et al., 2010). The highest N_{BAP} was found under haze conditions together with very high total particle concentrations: During haze days in Beijing, N_{BAP} can reach up to ~1.4 cm^{-3} (Wei et al., 2016) when N_{CCN} ~10^3 cm^{-3} (Gunthe et al., 2011). Thus, the ratio of N_{BAP} / N_{total} or N_{BAP} / N_{CCN} is likely small, i.e. in a range of 0.01-0.14%, independent of location. While such a marginal increase in the number concentration of cloud droplets does not lead to an observable change in cloud properties, the properties related to the CCN activation of BAP should be considered being more important for biological reasons, i.e. for BAP to be surrounded by water and the significant modification of the atmospheric residence time of BAP that is consequently changed by the transport and precipitation in clouds.~~

The critical saturation s_c can be used as a measure to estimate whether a particle will be activated into a cloud droplet (Rose et al., 2008):

$$s_c = \exp\left(\sqrt{\frac{4A^3}{27\kappa {D_s}^3}}\right) \qquad (8\cancel{7})$$

where A can be found in ***Equation 5***, κ is hygroscopicity, and D_s (cm) is mass equivalent diameter of dry solute particle. Applying this equation, one finds that for particles with D_{PBAP} of 0.01 to 10 µm, the critical supersaturations ($S_c = (s_c\text{-}1) \cdot 100\%$) are in a broad range of 0.0007%-24% (assuming κ = 0.03; σ = 72mN m^{-1}). For large PBAPs with D_{PBAP} > 0.5 µm, the critical supersaturations S_c is smaller than 0.062%. Typical environmental supersaturations (S_{env}) in stratocumulus and convective cumulus clouds are in the range of ~0.1-0.5% and ~0.5-1%, respectively (Pruppacher and Klett, 1997). Comparison to $S_{c,PBAP}$ shows that most PBAPs (D_{PBAP} > 0.5 µm) are likely activated to droplets as their S_c are significantly smaller than S_{env} in clouds.

4.2.2 Influence of the hygroscopicity (κ_{BAP}) and surface tension (σ_{BAP}) on CCN activation

Figure 8a shows the range of critical supersaturation (S_c) for the κ values shown in ***Table 2*** for the small~~est~~ PBAPs with D_{PBAP} = 500 nm, 100 nm, and 50 nm. For D_{PBAP} = 500 nm, S_c is 0.02% (κ_{PBAP} = 0.25, S_{CCN1}) or 0.06% (κ_{PBAP} = 0.03, S_{CCN2}), which are both below typical environmental supersaturation (S_{env}) in clouds. Only for smaller PBAPs such as ~~bacterial fragments or~~ viruses, SPPs or SFPs with D_{PBAP} = 100 nm (S_{CCN3}, S_{CCN4}), S_c changes from 0.24% (S_{CCN3}) to 0.69% (S_{CCN4}) when κ_{PBAP} increases from 0.03 to 0.25~~$\Delta\kappa_{BAP}$ = 0.22~~. For even smaller D_{PBAP} (50 nm), S_c increases from 0.68% (S_{CCN5}) to 1.97% (S_{CCN6}) when κ_{PBAP} increases from 0.03 to 0.25~~$\Delta\kappa_{BAP}$ = 0.22~~. Thus, only for fairly small PBAPs such as viruses, SPPs and SFPs (D ≤ 100 nm), the hygroscopicity κ_{PBAP} may impact their CCN activation. Steiner et al. (2015) reported critical supersaturations (S_c) of 0.81 (± 0.07)% for 50-nm SPPs and 0.26 (± 0.03)% for 100 nm SPPs. These values are similar to the values discussed above (0.68% to 1.79% for 50-nm particles, 0.24% to 0.69% for 100-nm particles) and are also in agreement with values based on the hygroscopicity (0.1 ≤ κ_{SPP} ≤ 0.2) reported by Mikhailov et al. (2019, 2020).

Overlaid on the vertical lines for S_c in ***Figure 8a*** are S_{env} in the cloud as calculated in our parcel model for different updraft velocities (w = 10 cm s^{-1}, 100 cm s^{-1}, and 300 cm s^{-1}). The sensitivity of CCN properties to updraft velocity and S_{env} has been discussed in numerous previous studies, e.g., Ervens et al. (2005). ***Figure 8a*** corroborates the conclusions from these previous studies that the variation of the κ over wide ranges only introduces a small change in the CCN activity and in cloud properties (e.g., drop number concentration, LWC) and that particle composition is most important in clouds with low updraft velocities.

Similar to S_c ranges due to different κ_{PBAP} values, we compare in ***Figure 8b*** predicted S_c ranges due to different values of σ_{PBAP} for high biosurfactant concentrations (when mass fraction of surfactants to total particle mass > 0.1%, σ_{BAP} = 25 mN m^{-1}) to those predicted for very low surfactant concentrations (σ_{BAP} = 72mN m^{-1}). For PBAPs with D_{PBAP} = 500 nm, S_c changes from 0.01% (S_{CCN7}, σ_{PBAP} = 25 mN m^{-1}) to 0.06% (S_{CCN2}, σ_{PBAP} = 72 mN m^{-1}). As discussed before, these large PBAPs will be likely all activated in clouds and the small difference in S_c introduced by change of surface tension ($\Delta\sigma$) does not cause a difference in their CCN ability. For smaller PBAPs, such as ~~bacterial fragments or~~ viruses, SFPs and SPPs with D_{PBAP} = 100 nm, S_c changes from 0.14% (S_{CCN8}) to 0.69% (S_{CCN4}) when $\Delta\sigma_{PBAP}$ = 47 mN m^{-1}. When D_{PBAP} further decreases to 50 nm, S_c changes from 0.4% (S_{CCN9}) to 1.97% (S_{CCN6}) when $\Delta\sigma_{BAP}$ = 47 mN m$^{-1.}$ Therefore, the effect of biosurfactant needs to be considered for small PBAPs in terms of CCN activity if a sufficiently large mass fraction of strongly surface-active biosurfactant is present. Note that the assumption of σ_{PBAP} = 25 mN m^{-1} in ***Figure 8b*** likely represents an overestimate as most biosurfactants exhibit a range of 30 mN m^{-1} < σ_{BAP} < 55 mN m^{-1} (Renard et al., 2016). In addition, the biosurfactant concentration, and thus the surface tension according to ***Equation 1***, depends on the mass fraction of biosurfactants in the PBAPs, the growth factor and on diameter of PBAPs. If the mass fraction is very low, σ_{PBAP} = 72 mN m^{-1}; when the mass fraction of biosurfactants approaches ~0.1%, σ_{BAP} might be as low as 25 mN m^{-1}. Typical surfactant mass concentrations are on the order of ~0.1% (Gérard et al., 2019); mass fractions for specific biosurfactants have not been determined yet. Such low mass fraction implies that only a few (< 10 to ~~–~~100) surfactant molecules (with a molecular weight M ~1000 g mol^{-1}) are present on submicron particles and/or that only a fraction of particles is completely covered by surfactants and thus exhibits a reduced surface tension. While biosurfactants might be also taken up by other particles while they reside on surfaces (soil, vegetation) where PBAPs are~~were~~ active, our conclusions also hold for such particles. Our sensitivity studies show ~~once more that under dynamic conditions in clouds buffering reduces the feedbacks of particle composition on supersaturation (Ervens et al., 2005; Stevens and Feingold, 2009).~~relatively lower sensitivity of cloud properties to particle composition than that predicted based on equilibrium conditions, in agreement with previous sensitivity studies (Ervens et al., 2005). ~~-~~Therefore, previous estimates of surfactant effects on cloud properties that are based on a simplified assumption of equilibrium conditions in clouds (Facchini et al., 1999), led to an overestimate of the role of surfactants on CCN.

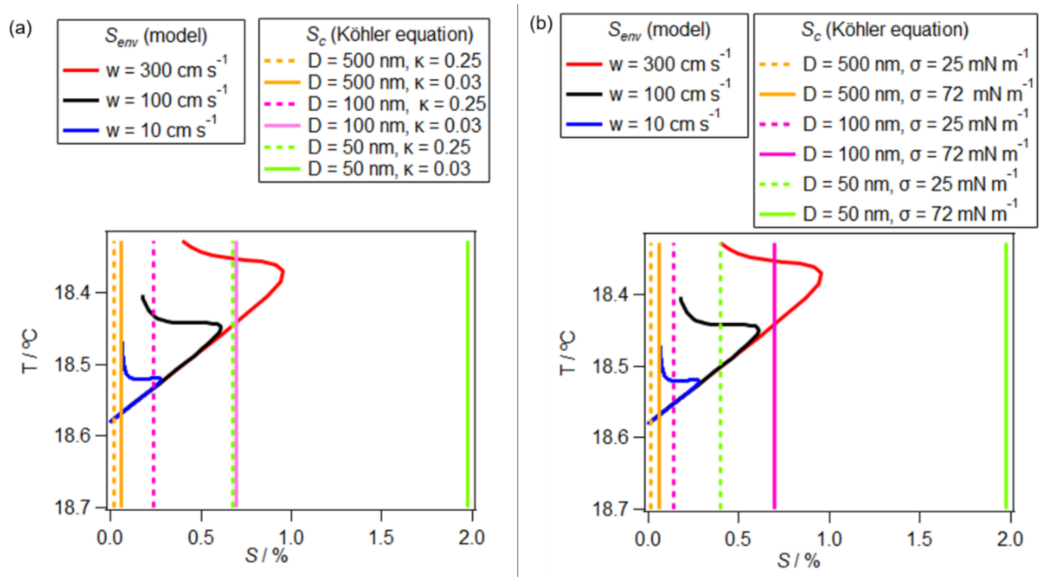


Figure 8. ~~Comparison of S_{env} to S_c of BAP with (a) different κ or (b) different σ. Model details can be found in ***Table 2***.~~ Comparison of the environmental supersaturation within the cloud (S_{env}) as predicted by the parcel model for different updraft velocities (w) to the critical supersaturation (S_c) of PBAPs based on Köhler theory. Results are shown as a function of (a) hygroscopicity parameters κ_{PBAP} and (b) surface tension σ_{PBAP}. Input parameters to the parcel model are listed in ***Table 2***.

We conclude that the mass concentration of biosurfactants needs to be quantified in order to better explore the biosurfactant effect on CCN activation of small particles. Given that the surface concentration of surfactants is likely higher than the bulk concentration (Bzdek et al., 2020; Lowe et al., 2019; Ruehl et al., 2016) as assumed here, even a smaller mass fraction of biosurfactants than calculated by ***Equation 1*** might be sufficient to decrease the surface tension of small aqueous PBAPs and the corresponding critical supersaturation. However, also for the concept of surface partitioning of biosurfactants, rather than for a bulk concentration, our conclusions hold true on the limited impact of surface tension suppression on CCN activation of supermicron PBAPs.

4.3 Sensitivity of mixed-phase cloud evolution to PBAP properties

4.3.1 Influence of PBAP concentration (N_{PBAP}) and diameter (D_{PBAP}) on ice nucleation

N_{PBAP} is on the same order of magnitude as that of total IN in some regions and at high temperatures ~~temperatures T > ~-10°C~~ (Pratt et al., 2009; Prenni et al., 2009), which makes PBAPs play an important role in mixed-phase clouds. Especially~~,~~ at the ~~these~~ relatively high temperatures of T > -10 °C, some bacteria and fungi have much higher nucleation site density than other aerosol particles (Atkinson et al., 2013; Hoose and Möhler, 2012~~a~~; Maters et al., 2019)~~can nucleate ice while other particles cannot~~, and therefore $N_{PBAP,\,IN}$

/ N_{IN} is ~100%. ***Figure 9a*** shows the change of percentage contribution of ice water content (% IWC, solid lines) and liquid water content (% LWC, dashed lines) to total adiabatic water content in a mixed-phase cloud (S_{IN1}, S_{IN2}, S_{IN3}). We define the onset of the Bergeron-Findeisen process as the temperature, at which the liquid water content fraction starts to efficiently decrease. With N_{PBAP} = 1 cm^{-3}, a~~A~~bove an IWC contribution of ~3%, ice particles start growing at the expense of liquid water (Bergeron-Findeisen-Process) (S_{IN1}). At lower N_{PBAP} = ~~∼~~0.01 cm^{-3} (S_{IN2}), the onset of the Bergeron-Findeisen-Process starts slightly later. With N_{PBAP} = ~~∼~~0.001 cm^{-3} (S_{IN3}), both IWC and LWC are predicted to increase simultaneously throughout the whole cloud, i.e. the Bergeron-Findeisen-Process is not initiated and cloud glaciation does not take place.

In ***Figure 9b***, we compare model results for simulations S_{IN4} and S_{IN5} in order to explore the effect of D_{PBAP}. With larger PBAP size such as D_{PBAP} = 2 µm (S_{IN4}) or D_{PBAP} = 5 µm (S_{IN5}), ice formation starts earlier in the cloud, but the onset of the Bergeron-Findeisen process occurs at approximately the same temperature as for smaller D_{PBAP} because of the feedbacks of IWC and LWC on the supersaturation in the cloud and vice versa. For SPPs and SFPs with D ≤ 100 nm, immersion freezing may be limited by the droplet formation on these particles (***Figure S3***). As ice formation is less efficient on non-activated particles ('condensation freezing'), the onset temperatures of freezing is significantly lower. As supermicron particles likely act as CCN under most conditions, this limitation might be smaller for large PBAPs. ~~In agreement with previous sensitivity studies (Ervens et al., 2011; Ervens and Feingold, 2013), these results confirm that the influence of D on the IN activity is relatively small (*Figure 9*). Based on these trends, it can be also concluded that processes that change the BAP size (e.g. ΔD~~~~$_{BAP}$~~ ~~by cell generation) are not critical to be included in models to represent the variability of IN property effect on mixed-phase clouds.~~

It should be noted that our adiabatic parcel model framework cannot fully represent the complexity of all processes occuring in mixed-phase clouds, such as complete glaciation followed by precipitation and demise of the cloud. However, we rather demonstrate the relative changes in percentage contribution of ice water content (%IWC, solid lines) and liquid water content (%LWC, dashed lines) to total adiabatic water content near the onset of ice nucleation. Thus, we apply our model in a similar way as in previous parcel model studies that explored the onset of the Bergeron-Findeisen process to various aspects of ice nucleation (Diehl et al., 2006; Eidhammer et al., 2009; Ervens et al., 2011; Khvorostyanov and Curry, 2005; Korolev, 2007; Korolev and Isaac, 2003).

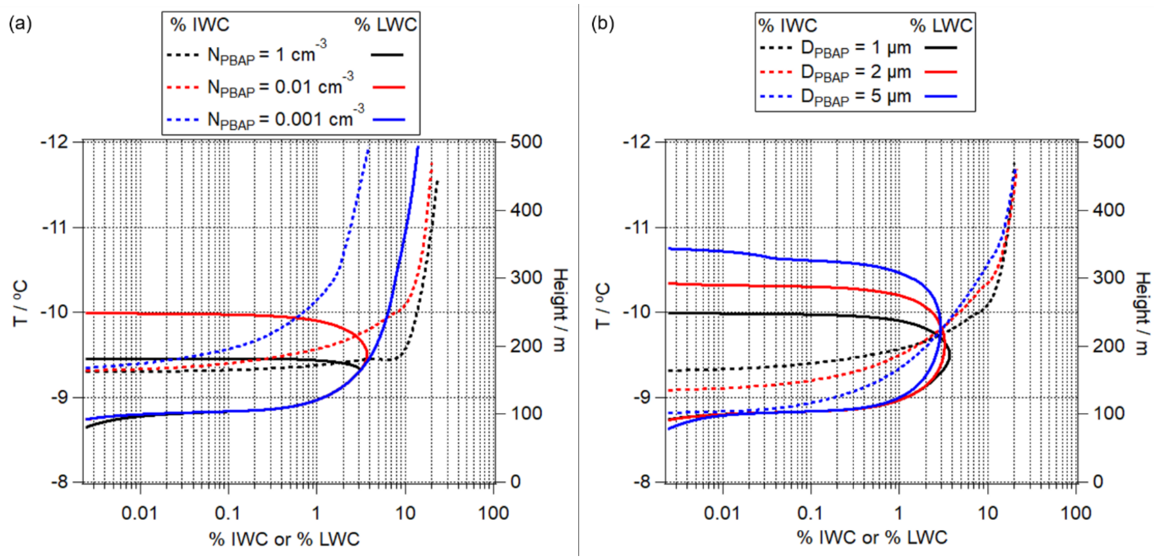
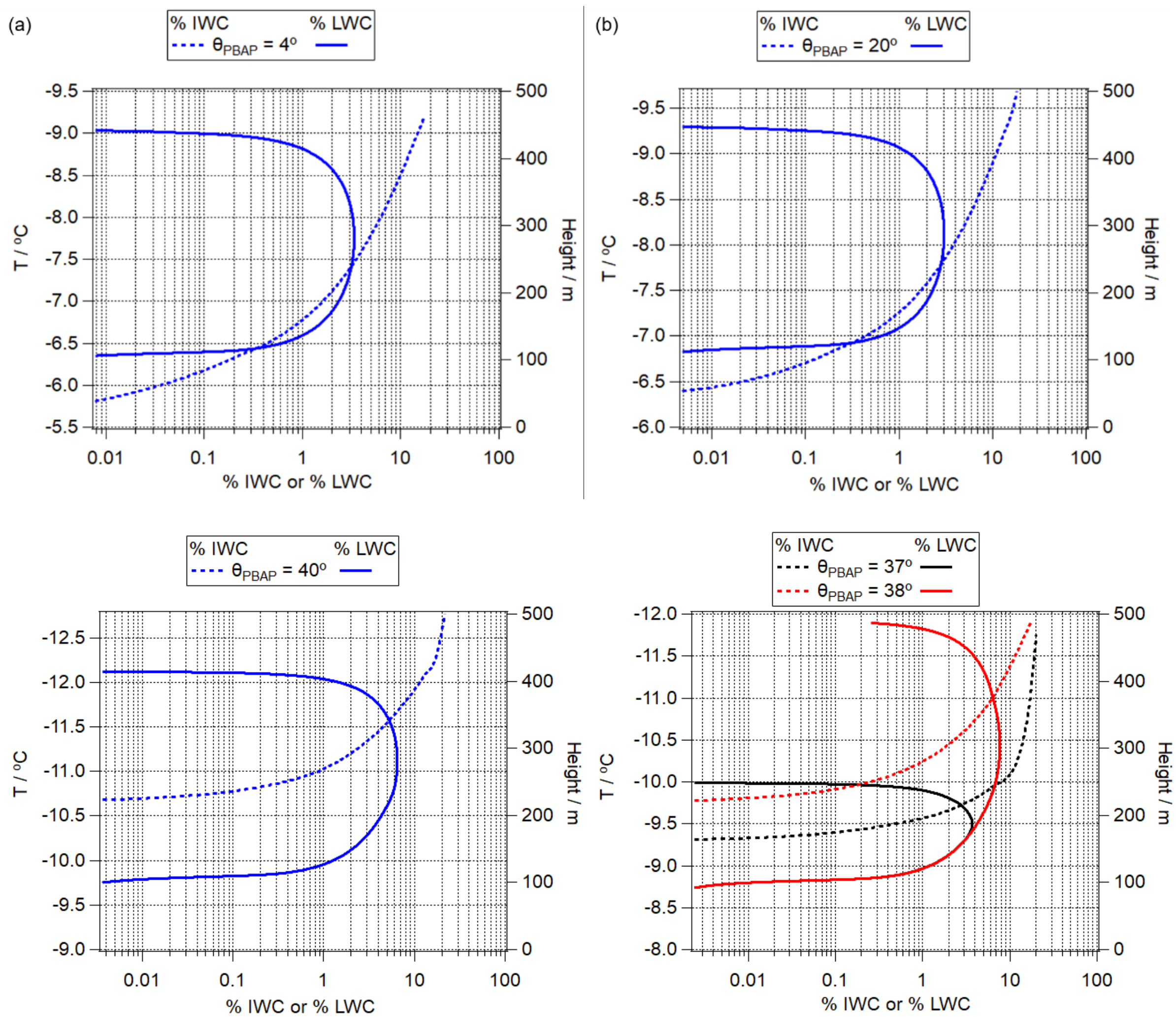


Figure 9. Percentage contribution of i~~I~~ce water content (%IWC, dashed lines) and liquid water content (%LWC, solid lines) to the total adiabatic water content as a function of (a) N_{PBAP} and (b) D_{PBAP}. Details on the simulations can be found in ***Table 2***.

4.3.2 Influence of the contact angle (θ_{BAP}) on ice nucleation

~~Different types of~~ PBAPs exhibit a wide range of contact angles of 4° < θ_{PBAP} < 44° (***Table 1***). ~~As shown in~~ ***Figure 10*** compares the predicted relative contributions of %IWC and %LWC to the total adiabatic water content. The comparison of Figures 10a and 10b shows that the onset temperatures of the LWC decrease are at ~ -7.7 °C (θ_{PBAP} = 4°) and ~ -8.3 °C (θ_{PBAP} = 20°), respectively, i.e. resulting in a difference of ~~,~~ΔT ~0.6 °C. This difference is predicted to be larger (ΔT ~3.3 °C) for PBAPs with θ_{PBAP} = 40°.~~ different BAP types that have θ_{BAP} of 4° or 20°, respectively, lead to a difference in temperature, at which the Bergeron-Findeisen process occurs, by ΔT ~0.6 °C. For BAP with even higher θ_{BAP} (40°), the Bergeron-Findeisen process occurs even at a lower temperature (ΔT ~3.3°C).~~



~~**Figure 10.** Ice water content (IWC, solid lines) and liquid water content (LWC, dashed lines) as a function of θ_{BAP}. Even when the contact angle increases by 1°, the initiation of Bergeron-Findeisen process might be influenced significantly.~~

Figure 10. Percentage contribution of ice water content (%IWC, dashed lines) and liquid water content (%LWC, solid lines) total adiabatic water content for θ_{PBAP} of (a) 4°; (b) 20°; (c) 40° and (d) 37° and 38°. The curves in the first three panels exhibit similar shapes for different temperature ranges, i.e. the Bergeron-Findeisen process starts at different temperatures. The last panel shows that even when the contact angle increases by 1°, the temperature, at which the LWC fraction starts decreasing, differs significantly.

As discussed in ***Section 2***, chemical (e.g., nitration, oxidation, adjustments due to pH) or physical processing of IN surfaces might lead to $\Delta\theta_{PBAP}$ ~1°. In ***Figure 10d***, we show %IWC and %LWC by comparing S_{IN2} and S_{IN9}. ~~IWC and LWC evolution by comparing S_{IN2} and S_{IN9}. It is clear~~ The results show that even such a small change of 1° in θ can cause a significant difference in the predicted IWC and LWC evolutions. The temperature, at which the %LWC starts decreasing ~~Bergeron-Findeisen process occurs~~ differs by ΔT ~1.3 °C. ~~These results suggest that a small change of contact angle due to different types of BAP or due to processing (Δθ) might affect the Bergeron-Findeisen process significantly. We only exemplarily explore Δθ for nitration based on the experiments by Attard et al. (2012).~~ Such a change in θ may be induced by pH changes; for example, it was found that Δθ is ~1.5° for bacteria such as *Pseudomonas syringae* when the cells were exposed to solutions of pH 7.0 and 4.1 at temperatures of T > -10 °C. Denaturation of IN protein's agglomerates (polymers) occurs at pH below 4.5 (Schmid et al., 1997; Turner et al., 1990), suggesting that changes in IN activities due to pH might be reversible at least above this pH value.

Similar differences in θ could be also caused due to other processes, such as the oxidation of pollen that lead to Δθ ~1.5° at T ~ -39 °C (Gute and Abbatt, 2018). However, at this much lower temperature, the sensitivity of the frozen fraction to Δθ decreases (Ervens and Feingold, 2013). Overall, ~~it can be concluded that chemical processing of bacteria or other BAP that freeze~~ our model results suggest that a small change in the contact angle due to different types of PBAPs or due to ageing processes might have a large impact on ice nucleation in clouds. ~~at relatively high temperatures in the atmospheric for extended periods of time might sufficiently alter their surface to induce a significant change in their IN ability.~~These differences might translate into feedbacks on other subgrid and dynamical processes in the cloud that amplify or reduce the efficiency of glaciation. However, such processes cannot be further explored in the adiabatic parcel model framework.

5. Conclusions

~~Based on our model sensitivity studies, we can rank the importance of the various parameters and processes of BAPs shown in ***Figure 1*** in terms of their radiative effects: The increasing importance and sensitivity are summarized in ***Figure 11***.~~

~~As the number concentration of BAPs only contribute ~0.1% to the total CCN concentration, even under conditions of high N_{BAP}, their role in CCN activation in warm clouds is negligible as they do not lead to any significant change in cloud properties. Since BAPs have usually supermicron sizes, they will act as CCN and even small changes in their chemical composition do not affect their CCN activity. The CCN activation of smaller BAPs such as bacteria fragments or viruses might be influenced by their hygroscopicity (κ_{BAP}) and surface tension (σ_{BAP}). κ_{BAP} might be modified by chemical (e.g., nitration, oxidation), physical (e.g., condensation of gases), and biological processes (e.g., formation of metabolic products, biosurfactants). Biosurfactants decrease the surface tension of BAPs (σ_{BAP}) and possibly even of other particles (σ_{other}) to increase their CCN ability. Even though the CCN activation of BAPs might be of very limited importance for cloud properties, it is more important due to biological aspects as it is a survival strategy of~~

~~microorganisms to improve their environmental conditions by water uptake, drop formation, and spreading on hydrophobic surfaces to enhance their survival time in the biosphere and atmosphere.~~

~~BAPs contribute ~1% to large particles with D > 0.5 µm, which makes them relatively important for the aerosol direct effect. BAPs have a direct cooling effect for most properties explored here whereas they could also change to a direct warming effect for certain species of BAPs. The most sensitive BAP property is the complex refractive index (m = n + ik), especially the imaginary part (k) which varies by three orders of magnitude among different types of BAPs. Assuming, for example, that all BAPs have the optical properties of *Aspergillus oryzae* fungal spores, the predicted direct aerosol effect changes from a cooling to a warming effect. Our RFE estimates clearly represent an overestimate as we only consider a small particle size range and concentration. Thus, the identified relative changes in RFE due to different BAP types and properties should be considered more representative than the absolute numbers.~~

~~The complex refractive index m_{BAP} can be modified due to chemical or biological processing. For example, nitration could lead to an enhancement of the imaginary part (absorptive properties), but the difference of scattering and absorption coefficient induced by nitration is much smaller compared to the differences caused by the refractive indices of different BAP types. Biological processing such as pigment formation (Pšenčík et al., 2004; Fong et al., 2001) might also lead to Δm_{BAP} to a significant extent, but we cannot quantify the role of this process in our model framework due to the lack of corresponding data. The second ranked parameter is ΔD_{BAP}, which also differs among different types of BAP or might change for one BAP type due cell generation or desiccation in the atmosphere. Obviously, the total number of BAP (NBAP) is of importance for all effects discussed here. However, as it has been shown that at many location N_{BAP} / N_{total} is approximately constant, the relative role of BAP likely does not change due to differences in absolute BAP concentration. Hygroscopicity κ might have an effect under high RH conditions. The effect of surface tension σ on direct radiative property is negligible.~~

~~The most important role of BAP is to act as IN because $N_{BAP, IN}$ / N_{IN} can reach up to ~100% at T > -10 °C. Given the high sensitivity of BAPs that initiate freezing, it is clear that not only the total N_{BAP} but also the fraction that can freeze needs to be constrained. While this fraction is usually ~100% for pollen, it can be as small as 0.01%-10% for bacteria. As identified in previous sensitivity studies, the surface composition properties, often expressed in terms of a contact angle θ_{BAP}, shows the highest importance to IN activity and therefore to the evolution of mixed-phase clouds (Bergeron-Findeisen process). The variability of θ_{BAP} between different types of BAP (4° < θ_{BAP} < 44°) determines the onset temperature of freezing and the temperature interval in which the Bergeron-Findeisen process may occur. Even a small change of $\Delta\theta_{BAP}$ ~1° as caused by chemical processing on BAP surfaces or pH change might affect the onset of the Bergeron-~~

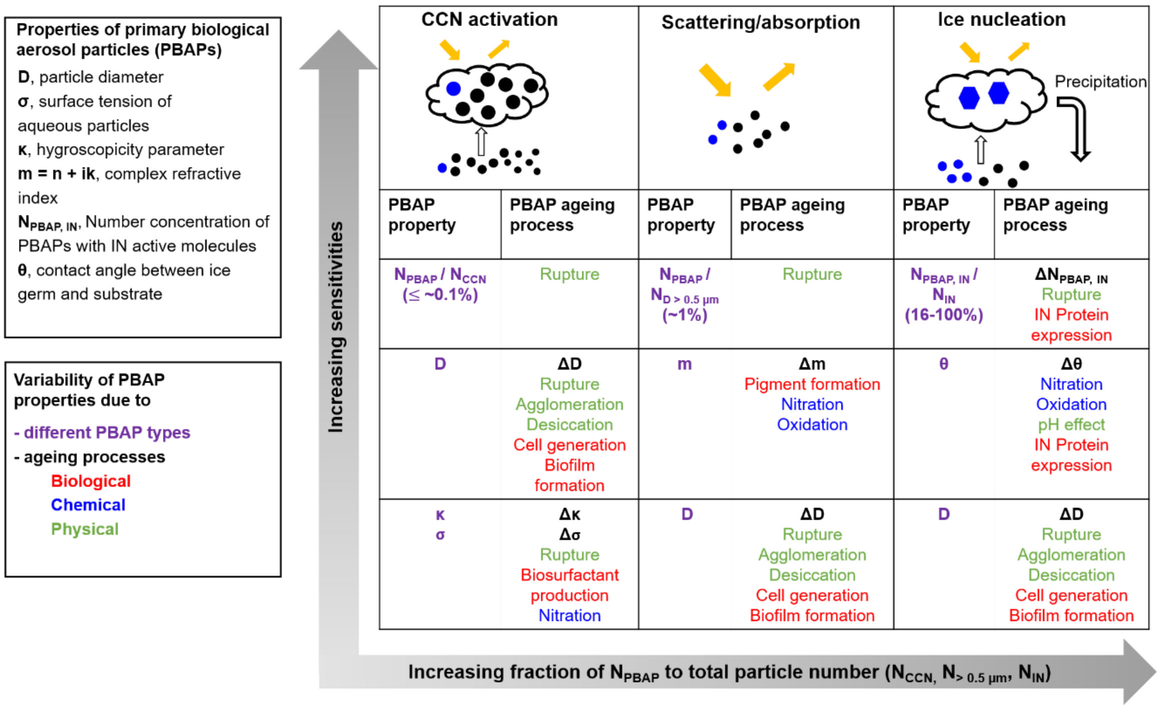
~~Findeisen process significantly. Thus, not only various BAP types should be parameterized with different θ_{BAP} in models but also $\Delta\theta_{BAP}$ due to modification by chemical and possibly biological processes.~~

~~The trends discussed above are summarized in ***Figure 11*** and show the relative importance of BAPs in the atmosphere, increasing from their roles in CCN activation, to the aerosol direct effect and to mixed-phase cloud evolution. The arrows on the left and on the bottom point to the most sensitive and most important parameters, respectively, which are placed in the upper right corner of the table.~~

~~Our study highlights the possible importance of BAP processing as not only chemical and physical processes but also biological ageing processes can modify the chemical composition and physical properties of BAPs. While the former two process types commonly occur on/in many other ambient particles as well (e.g. Δm due to nitration of SOA, or ΔD due to condensation of low volatility material), biological processing is unique to BAPs and currently not comprehensively included and explored in atmospheric models. For example, we suggest that cell generation or the expression of specific proteins might significantly affect BAP's IN ability. While the role of biosurfactant production ($\Delta\sigma_{BAP}$) is limited in modulating warm cloud properties and the aerosol direct effect, the biological aspects of this process might be of much larger importance: Enhanced water uptake by BAPs may extend lifetime of the microorganisms by improving their living conditions, i.e. reduce stress due to harsh ambient conditions (e.g. high ionic strength, low pH, desiccation). In addition, their inclusion in clouds as IN or CCN will lead to a more efficient transport and distribution across the atmosphere.~~

~~In addition to the few biological processes discussed in our study, additional biological processes (e.g., pigment formation, carotenoid accumulation, formation of metabolic products, biofilm formation) are included in ***Figure 11*** to give a more complete picture of ageing processes of BAP that may affect their radiative properties. Several of our results repeat findings from previous sensitivity studies of aerosol properties on the direct and indirect radiative effects. However, our study should be considered as guidance to future field, lab and model studies to further characterize the role of biological particles in the atmosphere as their emissions, budgets and processing are currently poorly constrained (Khaled et al., 2020) compared to more abundant aerosol types, despite their unique characteristics of living organisms that may affect not only climate but also public health.~~

Based on our model sensitivity studies, we can rank the relative importance of the PBAP properties and processes in ***Figure 1*** for their aerosol-cloud interactions and optical properties. Given the limitations of our process models~~,~~ in terms of scales, dimensions and parameter spaces, our results should be considered as qualitative, rather than quantitative estimates; the focus of our study is the comparison of relative changes due to various physicochemical parameters. Several findings of our model sensitivity results repeat those that have been drawn previously for other atmospheric particle types (Hoose and Möhler, 2012; McFiggans et al., 2005; Moise et al., 2015). However, in addition, unlike other atmospheric particles, PBAPs may constitute living microorganisms; thus, their properties may not only be modified by chemical and physical processes (marked in green and blue, respectively, in ***Figure 11***), but also by biological processes (marked in red in ***Figure 11***). To date, the extent to which these biological processes affect PBAP properties in the atmosphere is not known due to the lack of suitable data sets for atmospheric models. Our sensitivity studies, in combination with ***Figure 11***, give a first idea on which biological processes could modify relevant PBAP properties.



~~**Figure 11**. Schematic of the importance of BAP in the climate system and the sensitivity of radiative effect to BAP properties. The bottom arrow shows the increasing importance of BAP in CCN, scattering/absorption, and IN. The left arrow indicates the increasing sensitivity to BAP properties, which depend on the type of BAP and ageing processes.~~

Figure 11. Schematic of PBAP types and ageing processes that affect their aerosol-cloud interactions and optical properties. The bottom arrow shows the increasing fraction of N_{PBAP} to total particles (N_{CCN}, $N_{>5\mu m}$, and N_{IN}, respectively). The left arrow indicates the increasing sensitivity to PBAP properties as predicted based on our process model studies. The various properties might be modified by physical (green), chemical (blue) and biological (red) ageing processes.

(1) For any climate-related effects, the number concentration of PBAPs (N_{PBAP}) is the most important parameter. The PBAP number concentrations assumed in our estimates are based on measurements near the ground (Huffman et al., 2012; Jaenicke, 2005; Tong and Lighthart, 2000; Whitehead et al., 2016), which

typically decrease with altitude (Gabey et al., 2013; Perring et al., 2015; Ziemba et al., 2016). Thus, processes that affect N_{PBAP} in the atmosphere need to be well constrained; these processes include not only direct emissions but also particle fragmentation (rupture) or possibly new cell generation (multiplication). The number fraction of PBAPs to total CCN is relatively small ($\leq$ ~0.1%). For example, in the Amazon, it is on the order of 0.01 to 0.1% based on the reported ranges of PBAP number concentrations ($0.2 < N_{PBAP} < 1.2$ cm^{-3} (Whitehead et al., 2016); $0.04 < N_{PBAP} < 0.13$ cm^{-3} (Huffman et al., 2012)) and CCN concentration (N_{CCN} ~260 cm^{-3}, at 1% supersaturation (Roberts et al., 2001)). A similar ratio of N_{PBAP}/N_{CCN} (~0.01 to 0.1%) can be derived based on measurements in the megacity Beijing with $N_{PBAP} \leq 1.4$ cm^{-3} (Wei et al., 2016) during haze days and $N_{CCN} \leq 9.9 \cdot 10^3$ cm^{-3} (at 0.86% supersaturation) (Gunthe et al., 2011). Thus, a small change in N_{PBAP} likely does not significantly affect cloud droplet number concentration. Only in rare events, e.g. when pollen grains rupture with high efficiency, N_{pollen} might considerably affect N_{CCN} (Wozniak et al., 2018). However, droplet formation on PBAPs increases microorganisms' survival rate and decreases their atmospheric residence time due to precipitation, so the knowledge of their CCN-relevant properties is of biological relevance.

PBAPs contribute ~1% to large particles with D > 0.5 µm (Zhang et al., 2019), which makes them relatively important for scattering/absorption at a limited range of wavelengths. Only in the presence of high N_{PBAP}, it is expected that they have (local) impacts on the direct aerosol effect.

The number concentration of PBAPs that nucleate ice at T > -10°C is on the order of 10^{-5}-10^{-3} cm^{-3} (Murray et al., 2012). PBAPs comprise the predominant fraction of atmospheric particles that efficiently nucleate ice at these temperatures, i.e. N_{PBAP}/N_{IN} ~100% at T > -10°C (Hoose and Möhler, 2012). This fraction decreases at temperatures at which more abundant particles (such as dust) are also efficient ice nuclei: For example, at -30 °C, PBAPs contribute 16%-76% (Prenni et al., 2009) or 33% (Pratt et al., 2009) to total IN in mixed-phase clouds. Lab measurements have shown that up to 100% of pollen grains have IN nucleating macromolecules on their surface, whereas only 0.01-10% of bacteria express the proteins or other macromolecules that initiate ice nucleation (Failor et al., 2017; Joly et al., 2013; Pummer et al., 2015).

(2) The size of PBAPs influences the effects in ***Figure 11*** to different extents: While it is likely the most important parameter to determine their ability to act as CCN compared to hygroscopicity and surface tension, its role for PBAPs' optical properties is smaller than that of the refractive index. Also PBAP size plays a less important role than surface properties in the efficiency of ice nucleation. While several biological processes may increase the size of PBAP (e.g. agglomeration, cell generation), these changes are likely not important for the CCN activity of supermicron PBAPs since they will be activated under most conditions and thus an increase in their size does not affect their CCN behavior. However, modifications in the size, hygroscopicity (κ_{PBAP}), and surface tension (σ_{PBAP}) of smaller PBAPs, such as viruses, SPPs and SPFs, can

influence their CCN activation. κ_{PBAP} might be modified by physical (e.g., release of inner molecules due to rupture of pollen and fungal spores, condensation of gases), biological (e.g., formation of biosurfactants or other metabolic products), and chemical (e.g., nitration, oxidation) processes. Thus, processes that modify hygroscopic or surface tension properties of these smaller PBAPs might significantly change their ability to take up water vapor and form cloud droplets.

(3) The optical properties of PBAP are mostly determined by their complex refractive index (m = n + ik), especially by the imaginary part (k) which varies by three orders of magnitude among PBAPs. Under conditions when PBAPs significantly affect Mie scattering, small variabilities in the refractive index due to PBAP types or ageing processes might enhance (or diminish) their direct interaction with radiation (scattering/absorption). Modification processes include pigment formation as a defense mechanism of bacteria to oxidative stress (Fong et al., 2001; Noctor et al., 2015; Pšenčík et al., 2004; Wirgot et al., 2017) and nitration/oxidation of surface molecules (He et al., 2018; Liu et al., 2015; Nakayama et al., 2018). Additional biological processes such as biofilm formation are also included in ***Figure 11*** although experimental data are lacking to estimate their impact on PBAP optical properties.

(4) The ice nucleation activity of aerosol particles is often parameterized with a single contact angle (θ) between the particle surface and ice. ***Table 1*** shows that θ significantly differs among different PBAP types. In addition, our model sensitivity studies suggest that even a small change ($\Delta\theta_{PBAP}$ ~1°) as caused by chemical processing of surfaces, pH change of the surrounding aqueous phase, or biological processes such as protein expression level might significantly affect this activity. At temperatures at which PBAPs are the predominant IN (T > -10 °C), such a small change might translate into large changes in the onset temperature of freezing and cloud glaciation can be affected. Thus, in order to comprehensively account for ice nucleation of PBAPs, not only various PBAP types, but also $\Delta\theta_{PBAP}$ due to modification by chemical and possibly biological processes should be considered in models.

Exceeding numerous recent review articles that highlight the importance of PBAPs in general (Coluzza et al., 2017; Després et al., 2012; Fröhlich-Nowoisky et al., 2016; Haddrell and Thomas, 2017; Šantl-Temkiv et al., 2020; Smets et al., 2016), ***Figure 11*** gives more specific guidance on future measurements of the most sensitive PBAP properties in terms of their interaction with radiation and with water vapor. The detailed knowledge of PBAP properties might be of limited importance for global radiative forcing estimates, but is also relevant to properly describe PBAP transport, dispersion and lifetime in the atmosphere, which eventually affects biodiversity (Morris et al., 2014) and public health (Fröhlich-Nowoisky et al., 2016). While previous studies only focused on the physical and chemical properties, we highlight the uniqueness of PBAPs undergoing biological processes to adapt to the harsh atmospheric conditions; such processes might affect the adaption of PBAPs to atmospheric conditions which impacts their survival, transport and dispersion in the atmosphere.

Code and data availability: Details on the model codes and further model results can be obtained from the corresponding author upon request.

Author contributions: MZ and BE designed the model framework. AK, PA, AD contributed by fruitful discussions and commented on the manuscript.

Competing interests: The authors declare that they do not have any competing interests.

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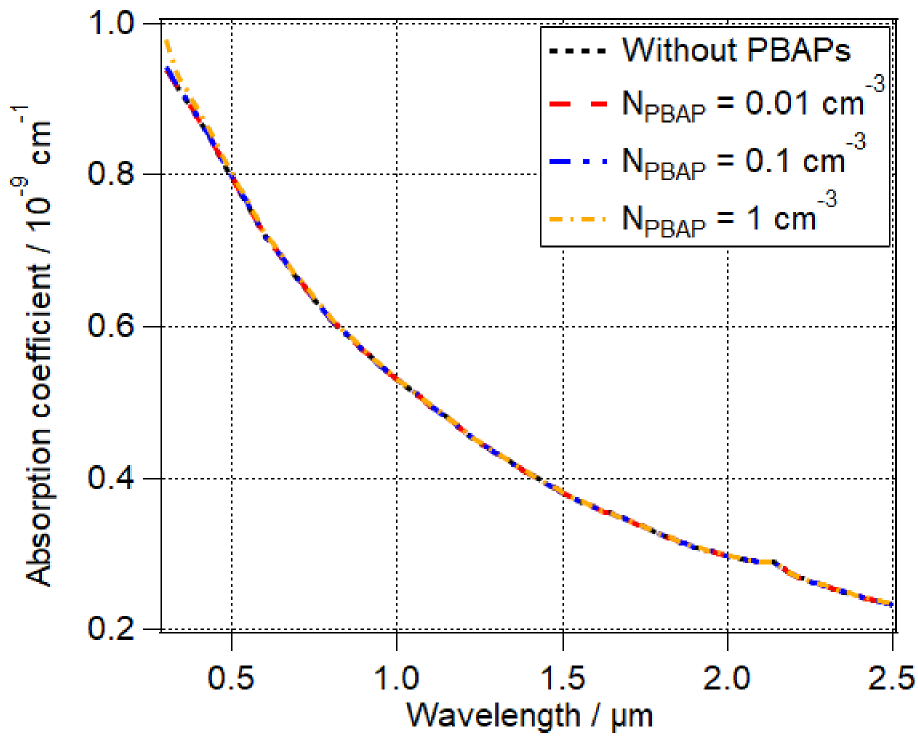

Figure S1. Total absorption coefficient for different PBAP number concentrations. The detailed input parameters can be found in ***Table 2***. The black, red, blue, and brown lines correspond to S_{opt1}, S_{opt2}, S_{opt3}, and S_{opt4} in ***Table 2***, respectively. No obvious change was predicted for the absorption coefficient.

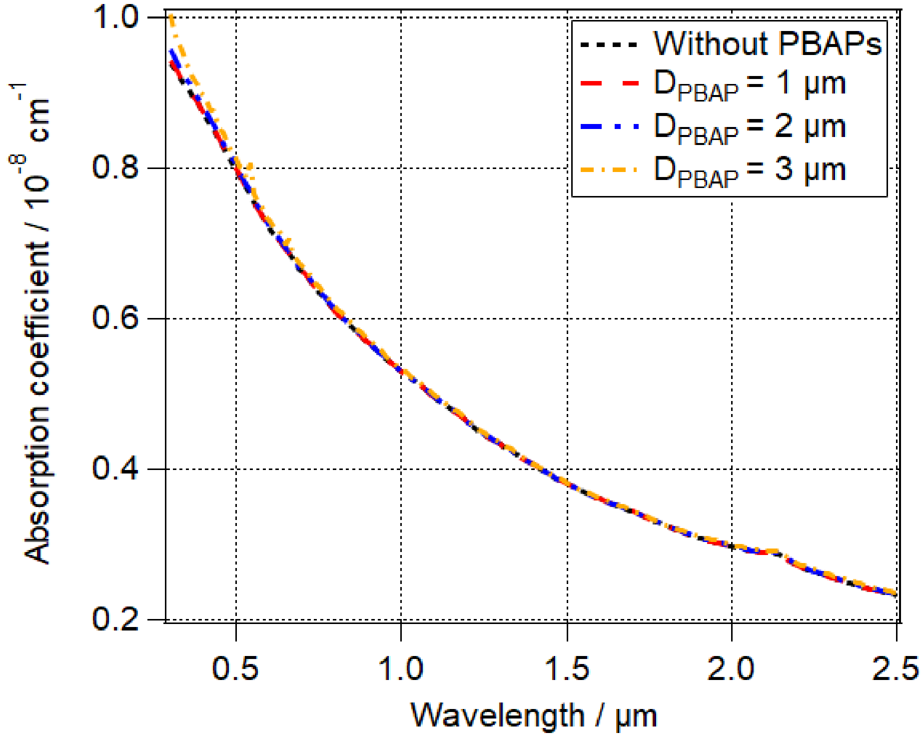

Figure S2. Total absorption coefficient for different PBAP diameters. The detailed input parameters can be found in ***Table 2***. The black line, red line, blue line, and brown line correspond to S_{opt1}, S_{opt3}, S_{opt5}, and S_{opt6}, respectively. No obvious change was predicted when the diameter of PBAPs increased.

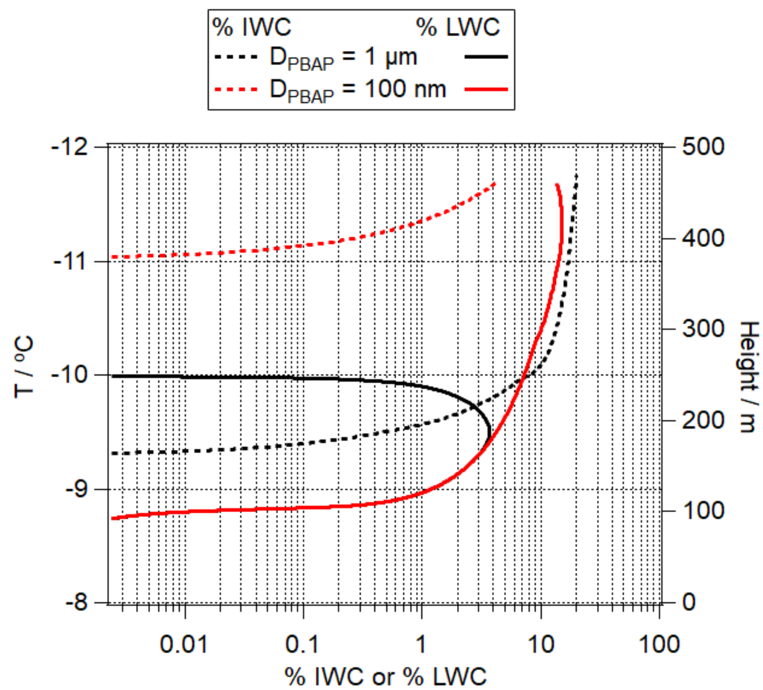

Figure S3. Percentage contribution of ice water content (%IWC, dashed lines) and liquid water content (%LWC, solid lines) to total adiabatic water content as a function of D_{PBAP}.