Review of

Impact of wildfire smoke on Arctic cirrus formation, part 2: simulation of MOSAiC 2019-2020 cases

by Ansmann et al.

Summary and general comment:
In this study the possible impact of wildfire smoke on the formation of ice crystals in Arctic clouds is investigated. For this purpose a parcel model is developed including two different classes of ice, as discriminated by different formation mechanisms (homogeneous freezing of solution droplets, heterogeneous nucleation). The model is used for several sensitivity studies in order to determine the impact of heterogeneous nucleation on these clouds. As a major result, it is stated that smoke particles act as ice nucleation particles and can suppress homogeneous freezing of solution droplets for conditions as measured during MOSAiC.

In general this is an interesting contribution, and it fits quite well into the scope of ACP. However, there are several major issues, which should be resolved before the manuscript can be considered for publication. Therefore, I would recommend major revisions for the manuscript. In the following I will explain my concerns in details.

Major issues

1. Model description

   - Overall formulation of the model:
     The model description is in a very bad shape. Generally, a usual parcel model is built using relevant processes as nucleation, diffusional growth, and sedimentation, as driven by a prescribed vertical motion. One would expect to see a set of ordinary differential equations for the variables ice number/mass concentration, water vapor mixing ratio, temperature and pressure, respectively. These equations are numerically solved for the time evolution. This kind of approach is described in former studies (e.g. Kärcher et al., 2022). In the manuscript a crude mixture of formulation is used. Some forcing terms and components are represented in the usual way (e.g. growth equation), but others are formulated in the discretized way, i.e. including already the time step (as the nucleation terms in eqs. 1 and 2). In addition, many assumptions for the model description are not stated clearly, but are merely included in an implicit way. From the formulation of the growth term and the treatment of sedimentation, one could conclude that a monodisperse size distribution of ice crystals is assumed. The variables as treated in the model are also not explicitly mentioned. From reading through the manuscript, one have the impression that the variables are as stated above, but this is never stated clearly. On the other hand, details of the heterogeneous nucleation are explicitly explained. The formulation of the growth equation (13) also puzzles me. In the text, layers with index l are mentioned, and this index also shows up in the equation. However, at the beginning of the manuscript, it is (more or less) explicitly stated that the authors investigate nucleation in a parcel model. Is it a parcel model or a column model with layers?

     I suggest to carefully rewrite the whole model description in a consistent way. The description would highly benefit from a stronger structure, i.e. subsections for the different processes and their formulation in terms of rate equations. For instance, the model description might start with equations for the temporal evolution of the environmental conditions, such as

$$\frac{\mathrm{d}T}{\mathrm{d}t} = -\frac{g}{c_p}w + \frac{L}{c_p}\frac{\mathrm{d}q_v}{\mathrm{d}t}\Big|_{\text{growth}} \tag{1}$$

Processes of nucleation, growth and sedimentation should be treated in separate subsections, with a summary of all equations (even in an abstract formulation) at the end of the model description section.

- Numerical issues:
The time evolution is written in a discrete way using indices i for the time level and a time step; in fact, the authors describe an explicit Euler step for the integration of the differential equations. This method is probably appropriate for a parcel model. However, the time step of one second seems to be quite large, so convergence of the scheme might be an issue. Since the nucleation rate is a very steep function in saturation ratio S_i, changing the time step to smaller values (see discussion in Spichtinger & Gierens, 2009) should be considered, otherwise overshoots or even unphysical behavior might result. Especially for the GW setups with high vertical updrafts, this should be taken into account.
- Units of the terms:
For some terms, the units are not included; for instance, the formulation of the mean free path lacks any units (line 150); the same is true for the conductivity of air (equation 12); there are some other examples in the manuscript, I will not list all of them. In cloud (micro) physics, often the cgs system (centimeter, gram, second) is used; however, it would be more appropriate to formulate the terms in SI units (maybe this is also a requirement of the journal).
- Wrong variable in the growth equation:
In line 132 the growth term is reformulated for the radius of a single crystal, thus the variable n_i must be omitted.
- Formulation of sedimentation:
In contrast to the formulation of other processes, for sedimentation a kind of discrete approach is used; particles larger than a threshold size are instantaneously removed from the parcel. This approach is in contrast to usual ways of parcel modeling, where advection of particles in a 1D column is assumed with a continuous terminal velocity (depending on mass) and solving some approximation of the (linear) transport equation. In this continuous approach, even for small ice crystals sedimentation has an impact on the system (see, e.g., Spichtinger & Cziczo, 2010). It is not clear to me if the authors have investigated the real differences between these different approaches; the effect of sedimentation might be small for cold temperatures, but for using the discrete approach some more information about possible differences must be provided. Also the implicit use of a monodisperse size distribution vs. broad size distributions (as e.g. lognormal distributions) might play a role. I suggest to test these different approaches, at least in an idealized way in order to provide robust statements about the meaningful use of the discrete sedimentation scheme. It might be that this discrete approach is a meaningful and efficient approximation, but at the current state of the manuscript this is not clear.

2. Environmental conditions
The whole study is concentrated on aerosol properties, these details are very clearly stated and explained. However, for the use of the environmental conditions, i.e. the initial conditions for the simulations, the authors remain very vague. For instance, it is completely unclear, why they use mean vertical velocities in the range between 0.1 and $1\,\mathrm{m\,s^{-1}}$. Actually, these

values are quite high for the middle and upper troposphere, synoptic scenarios as air motions along warm fronts lead to values in the order of $0.01 - 0.05\,\mathrm{m\,s^{-1}}$. It might be, that these high values were observed during the measurement periods, but this must be justified somewhere. The same holds for the temperature range $199 - 218\,\mathrm{K}$ and for the pressure range (just one value $p = 218\,\mathrm{hPa}$). For the gravity wave activity, the values for amplitudes and periods are also not well justified, just some hints are given, that similar values were measured in earlier campaigns. Overall, the authors have to make the link to the observations and measured conditions clearer and have to justify the chosen conditions in a more systematic way, e.g. deriving ranges of values from additional measurements. Maybe typical conditions could be derived from meteorological analyses, which are certainly available for this measurement period.

3. Variability and sensitivity
   As stated in the major point above, there is not a large variety in the environmental conditions, i.e. initial conditions for the simulations. However, the authors conclude from a very small sample of initial conditions and settings of the model several strong statements about the impact of smoke as heterogeneous IN. Since the parameter space is quite large and the chosen variability of initial conditions and model settings does not allow to probe the parameter space in a meaningful way, it is not clear at all how robust these results are. I suggest to investigate the impact of the variability of the environmental conditions (see above) as well as of the model settings on the simulated clouds in sensitivity studies. The authors should extend their initial settings (environmental conditions and model settings as nucleation parameters, e.g. the value of n_{250}) in order to obtain statistically relevant and robust results. Since neither the environmental conditions nor the nucleation parameters can be determined in a narrow range, this kind of ensemble or Monte Carlo approach of modeling might lead to robust insight of the impact of smoke particles. Since the model is probably quite cheap in terms of computational resources, a large number of simulations can be obtained quite easily.

Minor issues:

1. Variables
   For (bulk) cloud models, often the mass concentrations of water particles and water vapor are denoted by the letter q; I at least associate the letter m (or M) with a mass rather than a mass concentration, so it would therefore be worth considering a change in the variables' names. Since the molar masses are denoted by $M_{w,\mathrm{mol}}$ etc., this adds to the confusion.

2. Investigations of different nucleation pathways
   At several places in the manuscript, the author mention threshold or onset values for the saturation ratio in order to switch on the nucleation. However, the formulation of the nucleation rates (although often written in a threshold way, see, e.g., Spichtinger et al. 2023) lead to a start of nucleation for values well below the "threshold". This kind of onset value rather depends on the definition of a nucleation event in terms of a minimum value of freshly produced ice crystals. Maybe, the authors can clarify their definition of nucleation events and thus the definition of these onset values for the whole analysis.