

Dear reviewer,

thank you also for carefully reading and checking part 2 of the two MOSAiC manuscripts. The comments were very helpful and we considered most of them.

Both reviews forced us to dig deeper into the MOSAiC data and into the literature. At the end, we re-wrote both manuscripts and changed many parts significantly, motivated by the constructive suggestions of both reviewers.

All essential changes in the main text body are **marked in bold**

First of all, two statement of the editor:

Here I would like to point to a study that might be worth mentioning in the revised version of your manuscript. It also describes lidar measurements and simulations of cirrus clouds, but on the background of volcanic ash particles:

Rolf, C., Krämer, M., Schiller, C., Hildebrandt, M., and Riese, M.: Lidar observation and model simulation of a volcanic-ash-induced cirrus cloud during the Eyjafjallajökull eruption, Atmos. Chem. Phys., 12, 10281–10294, <https://doi.org/10.5194/acp-12-10281-2012>, 2012.

We consider the paper of Rolf et al. (2012) in the introduction of part 1, not in part 2. We found it more appropriate to mention it right in the beginning of the entire work.

In the revised manuscript, I would also like to ask you to shorten the abstract to ~250, as recommended for Part 1 of the study.

We shortened the introduction to about 286 words. We hope that is ok!

Content

The study describes the associated model simulations to polar lidar observations of cirrus clouds and aerosols in the time frame between October 2019 and March 2020 during the famous MOSAiC expedition described in the first part of this paper series. The idealized simulations focus on the impact of aged wildfire aerosol on ice nucleation by testing the sensitivity of different synoptic updrafts, temperatures, the impact of sedimentation, and the impact of very idealized gravity waves. It was found that only wildfire aerosol as heterogeneous INP can explain the cirrus observation and it is stated that heterogeneous freezing was the dominant freezing pathway in the observed cirrus cases.

Overall impression and rating

I am a reviewer of both manuscript parts and find the first part really excellent. The second part is also important and valuable for the scientific community, but I find the linking of the idealized simulations with the observations and especially with the respective synoptic situation not well done. I think the way of using idealized simulations is good in principle, but what is missing here is to show which dynamic situation fits best to the observations. As is also emphasized in the paper, the dynamics (updrafts, gravity waves, air mass history) plays a decisive role in addition to the influence of the wildfire INPs. In particular, trajectory calculations for the individual cases would be helpful to better classify the possible updrafts and to better estimate the temporal development along the air mass history (see comment below). This would clearly help to better support the main message of whether heterogeneous or homogeneous freezing is the dominant nucleation pathway.

We should already start here with our response! Sorry, but we do not agree! The observations clearly suggest that randomly occurring short-term updraft events (resulting from the interference of omnipresent gravity waves) dominated the vertical movements and were mainly responsible for

cirrus ice nucleation in an environment (upper troposphere) with an unlimited INP reservoir. This can be clearly concluded from all the lidar, radar, and radiosonde observations during the MOSAiC winter half year of 2019-2020. The probability for ice nucleation is highest at the top of the cirrus layers. The respective air parcels, in which heterogeneous ice nucleation occurred, were continuously in contact with the smoke polluted environment. The air parcels were never isolated and thus the INP reservoir in all these air parcels were never empty. The INP reservoir was continuously refilled from above, i.e., from the lower stratosphere. Homogeneous freezing needs absolutely smoke-INP-free conditions. These conditions were almost never given in the uppermost parts of cirrus during the winter 2019-2020 in the central Arctic .

Randomly distributed short-term updraft events dominated and initiated ice nucleation. In this case, air mass history analyses do not help (to our opinion). Randomly occurring updrafts are not covered by atmospheric modelling and backward trajectory calculations. Therefore, we introduce the observations of Podglajen et al. (2016) (superpressure balloon observations in the Antarctic stratosphere) to get an idea about the randomly occurring updraft events (amplitudes, vertical velocities, durations, etc.).

To corroborate the omnipresence of wildfire smoke in the upper troposphere during the MOSAiC winter half year, we present a new figure (Fig. 4 in Sect. 3). This Fig.4 shows that there was always smoke in the top region of the cirrus layers during a long lasting cirrus formation period of six days (21-26 January 2020). And Fig. 8 in part 1 showed that there was no decrease of the smoke particle concentration during all these days as the smoke particle profiles observed on 21 and 27 January 2020 indicated. Our best example of the omnipresent smoke in which cirrus started to develop was shown in Fig.14 in Ansmann et al. (2023) and covered a five day period from 25-29 February 2020.

We simply cannot ignore all these observations. Nevertheless, we follow the suggestion of the reviewer and state several times that homogenous freezing events are probably widely suppressed, but they may have occurred.

In general, the manuscript is well written and structured, the illustrations are also excellent, but I am still missing a few points in the second part of this paper series that need to be addressed before the manuscript can be published. For this reasons, I recommend publication in ACP after addressing my comments and some manuscript revisions.

Main comments/questions:

- Section 3: The section of the manuscript in which the simulations are described (especially pages 12-17) is well explained and easy to read. However, a large part of it is already explained and described in detail in some other studies on cirrus simulations, so that the added value for the scientific community is rather limited. Examples include the studies by Kärcher et al 2019, Krämer et al. 2016 or Spichtinger and Cziczo (2010). It would be good either to shorten it a bit or at least cite some of the studies showing the same effects were cited in the text.

Thank you for these hints. We agree! As a direct response, we removed Figure 6 (showing sedimentation effects in detail). We looked more carefully into the mentioned papers (Kärcher et al 2019, Krämer et al. 2016, Spichtinger and Cziczo, 2010). We shortened the text, we added the papers Spichtinger and Gierens (2009), Spichtinger and Cziczo (2010), and Krämer et al. (2016) to the references and we discuss in more detail the sedimentation aspects (Sect. 3.2, page 10). However, we did not change our 'primitive' or simple sedimentation approach because the sedimentation impact on our simulation results is quite low. By switching on and off our simple sedimentation correction routine we cover the maximum range of a possible sedimentation-related effects. Compared to the results in Spichtinger and Cziczo (2010), our sedimentation correction is

too large. With other words, the sedimentation impact is lower if we follow the approach of Spichtinger and Cziczó (2010). However, in simulations of short-term updrafts at the low Arctic temperatures, sedimentation effects were found to be generally low, even by using our simple approach.

- Figure 5/6, e.g. line 82: Test of lower updraft velocities also in combination with gravity waves. In Kärcher and Lohmann 2002 the synoptic updraft range span over 0.01m/s to 0.1m/s which was also tested in Krämer et al, 2016 in comparison to research aircraft data. The low updraft can also produce low ICNC values in case of homogeneous ice nucleation. Also the combination of low updraft with gravity waves can produce low ICNC in the range of  $<10^{-1}$  as shown by Kärcher et al 2019. It would be good to also include this updraft range of 0.01 m/s in your study as it seems to be important for final answer about the dominant nucleation pathway.

We agree. We now include simulation scenarios with large scale lofting events in the revised version of part 2 (Figure 6 in the revised version). We show simulations with large-scale lofting and updraft velocities of 1 and 3 cm/s in Sect. 4.1 (page 15). We include also scenarios with 10 and 20 cm/s updraft speed in this figure to better discuss the dependence of ice nucleation on updraft speed. However we do not simulate super positions (scenarios of large scaling lofting, combined with short-term lofting on top).

Regarding low values ICNC, produced during homogeneous freezing events, we now state in the conclusion section, that information about ICNC (alone) cannot be used to identify the ice nucleation mode. Both, heterogeneous ice nucleation as well as homogeneous freezing can produce high as well as low numbers of ICNC.

We should add in this context that we improved almost all figures so that a better discussion of the roles of heterogeneous and homogeneous ice nucleation is possible now. We show and compare now scenarios with heterogeneous ice nucleation (in smoke polluted air) and scenarios with homogeneous ice nucleation (in smoke-free air) and discuss at which conditions homogeneous ice nucleation can set in.

- I find the assumption of any climatological updrafts a bit too simple to answer such an important question as the dominance of nucleation mechanism. You have clearly explained the role of updraft and gravity waves. In order to show which meteorological conditions were present during the cirrus observations, it is essential to look at the air mass history. Why not simply use trajectories calculated from meteorological fields, e.g. ECMWF ERA5, to estimate the large-scale updraft. ERA5 already includes some gravity waves, so you would only need to estimate the smallest scale gravity waves in your simulations in addition. Another advantage is also that one could see how long the cloud has potentially already existed before your observation and how many nucleation cycles may have already occurred. As you describe, both have a significant influence on how many INP have already been consumed and sedimented and whether homogeneous freezing might also play a role. I therefore suggest that you make similar trajectory calculations for the cirrus cases, as already shown in Part 1, and determine the updraft of the airmasses during and especially before reaching the observation site. This could be used to create a PDF plot, which could then support the hypothesis described with your assumed updrafts. I guess this would support and better substantiate your statement.

We explained already in the beginning of this reply letter that the observation never indicated that the smoke INP reservoir was empty in the cirrus top region, where ice nucleation usually starts. Nevertheless, we state several times that it remains a realistic option that there were air parcels (or air masses) in which the INP reservoir was empty so that homogeneous freezing could set in. As mentioned in the beginning, we do not think that air mass history analyses are helpful and would

improve the discussion. We checked the trajectories and time periods with high vertical motions showed always vertical velocities smaller than 5 cm/s and they were all related to large-scale lofting.

Guided by many papers (especially the ones of Kärcher et al.) we never concentrated on large-scale lofting (frontal lofting, orographic lofting, etc.) when we performed the simulations for this paper. The message was always that short-term updrafts are most important, and then, trajectory analysis and air mass history analysis (covering large scale lofting events, but not randomly occurring short-term updraft events) are to our opinion not very helpful to describe the actual updraft conditions.

Furthermore, the observed virga (occurrence frequency, temporal width of the virga in height-time displays, structures, ICNCs) clearly corroborate the hypothesis that short-term updraft events dominated and provided favorable conditions for ice nucleation.

Last point here, motivated by the comments of the reviewers and since all hints pointed to the direction that short-term updrafts played an important role, we included the study of Podglajen et al. (2016) in our discussion. We discuss the observation of Podglajen et al. (2016) mainly in part 1. Podglajen et al. (2016) quantified wave-induced fluctuations of temperature, vertical displacement of air parcels, and vertical velocity in the lower stratosphere over polar regions by using measurements with superpressure balloons. The observations allowed the whole gravity wave spectrum (up- and downdraft events) to be described and provided unprecedented information on both the intrinsic frequency spectrum and the probability distribution function of wave fluctuations. These up and downdraft events showed a wide spectrum of amplitudes and updraft velocities. They are randomly distributed in space and occur randomly in time. The observations of Podglajen et al. (2016) are used to interpret our observation findings and guided us in the simulation studies and the development of the simulation strategy.

- You state in your text that the simulations showed that the INP reservoir was never completely used. But the cloud typically exists over longer time periods than just the 2500s used in your idealized simulations and also can have multiple life cycles during the air mass transport i.e. multiple uplifts, nucleation, sedimentation. And even before your observations there might already be multiple cloud occurrences over hours to days within this air mass which just passed your observation site at a specific time. Why shouldn't all the INP have already been used up in that time frame? And how do new INP get into the cloud then? This comment is closely linked to my previous comment about the air mass history and should also be shown and discussed in the text.

We discussed all this already above. The air parcels in the cirrus top region were not isolated. They were in permanent contact with the polluted environment. It seems to be unrealistic to assume that any of the air parcel was free of any smoke INP so that homogeneous freezing could set in.

The MOSAiC observation suggest that the smoke reservoir was continuously refilled from above. At least we saw almost constant smoke levels in the upper troposphere over many months, and higher smoke values in the lower stratosphere and lower smoke values in the middle troposphere (probably indicating the removal INPs by the virga of falling ice crystals). Examples of smoke profiles are shown in part 1.

Nevertheless, we include discussions regarding multiple uplifts and the possibility that homogeneous freezing conditions were given. However, the observations provide the opposite impression, that favorable homogenous freezing conditions were practically never given because of the omnipresent smoke.

- Line 168-169: Are the radiosonde data measured outside or inside of cirrus ? This would of course has significant influence on the initial values for the simulation.

The sondes and lidar observations were always in perfect harmony. When the lidar did not detect any cirrus feature before, e.g., 24 UTC, then the sonde, launched at 23 UTC, did not measure any ice supersaturation. When the lidar detected a cirrus field, the respective sondes showed ice supersaturation indicating that they ascended through the cirrus field (detected with lidar). The full cirrus lidar backscatter profiles were usually in line with the full RH profile measured with radiosonde, from virga base height to cirrus top. Such examples are shown in part 1 (for the 22 January case study).

- line 257-260 and Figure 3: Extrapolation especially on a logarithmic scale can lead to extremely large errors and deviations. As the ice crystal number concentration (ICNC) is so important for the simulation results to compare with, you should at least show maybe based from an example of another measurement campaign, that such an extrapolation is approximately valid and reasonable.

We changed the discussion in part 1 and part 2, because we do not apply any extrapolation approach. We do not correct aggregation effects at all. We only provide a discussion about a potential aggregation effect (in part 1, Sect. 3.4) based on papers of Kienast-Sjoegren (2013), Field and Heymsfield (2003), Wolf et al. (2018) and Mitchell et al. (2018). Kienast-Sjoegren et al. (2013) demonstrate that aggregation effects are rather small in cases with cirrus temperatures of -60 to -75°C. Wolf et al (2018) provide observational support for this. Our observations are usually in line with this finding. However, we mention the two further papers of Field and Heymsfield (2003) and Mitchell et al., (2018). These studies suggest that the aggregation effects may lead to large changes in ICNC (by a factor of 2-5) during falling. However, Field and Heymsfield (2003) show mid latitude cirrus cases with ICNC of about 200 L<sup>-1</sup> at cirrus top.

- line 267: "and would probably widely prevent the occurrence of high ice saturation ratios of 1.3-1.4." This is true, if not all INPs are already consumed. Otherwise, it is of course possible that the supersaturation will continue to increase until homogeneous nucleation sets in at some point.

We provided our opinion regarding an empty INP reservoir already several times in this reply letter. We can only offer and accumulate our arguments and these arguments are (given in part 1, and repeated in part 2 and are in line with the simulations): 1) the upper troposphere was always polluted and thus never clean, 2) the INP reservoir was permanently refilled from above, 3) radiosonde observations of the saturation ratio ice point to inefficient INPs such as smoke particles. To construct a scenario so that homogeneous freezing dominates at the end simply ignores all facts that were measured with lidar, radar, and radiosondes.

- line 391-392: "Figure 9 provides an overview of the smoke impact on ice formation for the main range of MOSAiC cirrus top temperatures from 199-213 K". Why are the cloud top temperatures lower than showed in Figure 4 of part 1? There you could see temperatures ranging from 197-225 K.

We change a bit the strategy here and just provide two different scenarios (213 K and 199 K). To our opinion, it is not necessary to simulate the entire range of possible cirrus top temperatures. To keep the discussions short (and to show only the essential simulations) we consider two cirrus top temperatures: 213 K (cirrus top temperature) representing the November-December 2019 cirrus clouds and 199 K representing the January-February 2020 cirrus clouds. Simulations with these two temperatures are sufficient to show the impact of different temperatures (and thus different amounts of water vapor).

- Line 395: Difference between ICNC values in the virga (range of 0.1-20 L<sup>-1</sup>) and cloud top (4-300 L<sup>-1</sup>, line 259) obtained by your extrapolation method. High values are also partly visible in your cases which are shown in part 1 with ICNC in the upper part of the ICNC observation in the range of 50-100

L-1. Where do they come from? Are they coming from multiple nucleation events and are just too small to sediment?

As mentioned, we do not apply any extrapolation to estimate ICNC in the ice nucleation zone at cirrus top. Multiple nucleation events may lead to large ICNC numbers. But even single, but strong updrafts (with relatively large amplitude) can lead to large ICNC values. To our opinion the ICNC value range reflects to a large extent simply the range of amplitudes of the short term updrafts. This is shown and discussed in Sect. 4.4 in part 2.

- Figure 9: What would be the impact on the starting time of your idealized gravity wave? You always start with the ascending part of the wave together with the start of the simulations. I would assume, if you start with the descending part of the wave you could create even higher cooling rates / updrafts at the time of nucleation and maybe possible even high enough to trigger homogeneous nucleation to occur. I guess the phase shift is similar sensitive as the different wavelengths of the wave and should also be tested in this study.

First of all, we start now all simulations with the same super saturation ratio of 1.2 to make all the simulations better comparable. If we start with the descent phase of an air parcel first then we will have an ice saturation ratio of about 0.9 at the minimum height. Then the ascent phase (two amplitudes long) begins. At the end, we should have the same results as presented in part 2. If the saturation ratio is 1.2 at the beginning of the two-amplitude lofting then we may reach the saturation ratio of homogeneous freezing. Sure! But homogeneous freezing will not occur as long as INPs are available, and smoke-free conditions were not given.

However, we leave the door open, as suggested by the reviewer, we mention several times that homogeneous freezing may have occurred occasionally. We do not exclude this option. We state that even our point of view, based on all these solid and consistent observations and facts, remain hypotheses.

Technical comments/suggestions:

- line 39: " level, in (c) the ", I guess you meant "level, and (c) the"

Yes, improved!

- line 97: Skip one "is".

Done!

At the end, a few further comments to the revised version and some new aspects are added.

We changed the result section considerably compared to the result section in the original version! These changes were motivated by the many reviewer comments. We now start with large-scale lofting (Sect. 4.1) and then we move forward to gravity-wave-induced updraft events (Sect. 4.2).

The dominance of these short-term events on ice nucleation is even visible in our MOSAiC virga observations. We emphasize that now in part 1 a, and mention that again in part 2: The virga structures, intensity, and occurrence frequency contain information about updraft frequency, duration, and related ice nucleation intensity. And the virga occurrence frequency and structures clearly point to the dominance of single, short-term updraft events providing the conditions for heterogeneous (and homogeneous) ice formation.

Another new point is that we include the observations of Podglajen et al. (2016) in the discussion. These superpressure balloon observations help to explain our findings much better and provides answers to the question: Why were the ICNCs frequently so low?

According to Podglajen et al. (2016) updrafts with shallow amplitude, leading to low ICNCs occur much more frequently in the upper troposphere than updrafts with large amplitudes. The simulations confirm that during shallow updraft events the nucleated number of ice crystals is typically low. This is shown in Fig. 10.

Some remarks to the simulation figure:

old Fig. 5, now new Fig. 6: new figure considers large scale lofting simulations.

old Fig. 6, now removed.

old Figs. 7-9, also new Figs. 7-9, these figures, dealing with gravity wave simulations, are now also improved. Always homogeneous ice nucleation is simulated for better comparison.

old Fig. 10, now still Fig. 10, but improved.

old Fig. 11, now removed.

old Fig. 12, now Fig. 11, and improved (or better: changed a bit)



Dear reviewer,

thank you for accepting to review the manuscript, to read it carefully, and to provide us with numerous good, constructive, and friendly comments. We were happy to get reviews from two experienced scientists! We tried to do our best to integrate most of the comments and suggestions.

The essential changes in the main text body are indicated in bold.

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 has 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?

We apologize that we bothered the reviewer with this not well written section. We improved the description of the simulation model, so that we are at least happy with section 2 now. We followed the good idea to introduce subsections. We did not consider all the mentioned recommendations. We hope that this is ok.



Now, we provide a short overview of the essential simulation steps as given in Figure 1, in the beginning of section 2, before we continue with details in the new subsections 2.1, 2.2, and 2.3. To avoid confusion about the ice crystal size distribution (monodisperse or not), we state right in the beginning that ice nucleation is a time dependent process and all nucleated particles start immediately to grow after birth so that a broad size distribution builds up with time (also written in Sect.2.2, page 9).

We provide much more information about dimensions and units now. We removed 'm' and 'M' and introduced 'Q' for mass (or better mass concentration), because all computations are related to m-3.

We better explain the index 'l' to avoid confusion.

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 .....

As stated above, we rewrote the introduction part of Section 2 (model description) and introduced subsections. However, we did not change the equations.

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.

We introduced these sections and we improved the contents of the subsections as recommended.

- 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.

We improved the resolution of the model, and are now able to run the simulations with 0.01 s resolution. We found that we need a resolution of 0.1 s in simulations with 10 and 20 cm/s vertical velocity to avoid underestimations (ice crystal number concentration) of the order of 20%. And 0.5 s and 1 s is ok for slow ascent rates of 3 cm/s and 1 cm/s, respectively. The used resolutions are given in the manuscript, e.g., in the figure caption of Fig.6.

- 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).

We went through the text and now provide much more dimensions and units than before. We hope, journals will leave the freedom to use a mix of cgs and SI....

- 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.

We improved that! This was a wrong statement in the text, in the model it was fortunately ok.

- 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.

We introduced a simple (trivial) method to consider sedimentation effects. It is a first step! We just wanted to get an idea how important it is to consider (or ignore) sedimentation effects. In future, we should implement the method of Spichtinger and Cziczo (2010), also applied by other groups (Krämer et al., 2016). However, for these cold cirrus cases with 199 K top temperatures, sedimentation effects have not a big impact. This is already a bit different when simulating cirrus with top temperatures of 213 K cirrus. But in the case of gravity wave simulations (short term updraft phase) the sedimentation effect was at all small. So, we did not change our approach. We improved the description in the new subsection, Sect. 2.3, mention the appropriate sedimentation schemes, provide references, and state, that our 'primitive' method covers the maximum possible impact of sedimentation, and when we switch that off, we have the minimum impact. We removed the figure (Fig.6 in the submitted version) where we explicitly showed the impact of sedimentation. However, we compared our results with findings in the papers of Spichtinger and Cziczo (2010) and Krämer et al. (2016) and found that we see similar oscillating structures as presented in these papers. That convinced us that our simulation approach is not so bad, and acceptable in the case of simulations of cold Arctic cirrus clouds.

Again, to avoid confusion, the size distribution of ice crystal is not monodisperse. Only the ice-nucleating particles have the same size, as in many other papers. However, the ice nucleation is a time-dependent process and together with ice growth a broad spectrum automatically builds up with time. In the sedimentation subroutine, we omit all sizes that exceeded a given threshold diameter (of 20 micrometer).

Final point here: We mention at the end of Sect. 2.3 (page 10) that we compared our simulations with results of Spichtinger and Cziczo (2010) (in their figure 4) and found that our simulations are roughly in agreement with the time series of humidity and ice crystal number concentration (ICNC) in their figure 4 if we consider a sedimentation threshold radius of 30-35 micrometer instead of 10 micrometer in our sedimentation impact computations.

## 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 m s<sup>-1</sup>. 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 m s<sup>-1</sup>. 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 K and for the pressure range (just one value  $p = 218$  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.

### Point by point answers:

Updrafts are typically related to large-scale lofting (with 1-5 cm/s updraft velocity) and short-term lofting events as a result of gravity wave activity, windshear turbulence, and orographic impact causing vertical velocities of mostly 10-30 cm/s. We now show simulations of large scale lofting events in Figure 6. We add simulations with 10 and 20cm/s just for comparison and to better discuss the impact of vertical velocity on ICNC.

The observed ice virga occurrence frequencies and structure clearly show that short-term updraft events dominate and ice nucleation takes mainly place during the short-term updraft events. We provide the link to real-world updraft properties now via observations by Podglajen et al. (2016). Podglajen et al. (2016) quantified wave-induced fluctuations of temperature, vertical displacement of air parcels, and vertical velocity in the lower stratosphere over polar regions by using measurements with superpressure balloons. The observations allowed the whole gravity wave spectrum (up- and downdraft events) to be described and provided unprecedented information on both the intrinsic frequency spectrum and the probability distribution function of wave fluctuations. These up and downdrafts events showed a wide spectrum of amplitudes and updraft velocities. An important finding of Podglajen et al. (2016) is that updraft events with an amplitude of, e.g., 100 m occur by an order of magnitude more frequently than updraft events with an amplitude of 200 m. We can conclude that the most frequently occurring updraft events, that contribute to ice productions, are those with low amplitudes. However, they lead to low amounts of ice crystals. The ice nucleation events caused by frequently occurring shallow updrafts thus dominate the cirrus characteristics including the ICNC levels. All this is now discussed in part 1 (Sect. 3.5 in part 1) and briefly also given in part 2.

Concerning simulations with cirrus to temperatures of 199-218 K: Here, we even reduced the simulations in the revised version. We show simulations with 199 K and 212 hPa (representing well cirrus top temperature and pressure conditions of the MOSAiC January and February 2020 cirrus cases) and simulations with 213 K and 265 hPa (representing well the MOSAiC November and December 2019 cirrus cases). In a paper with focus on the impact of wildfires smoke on ice formation in the upper troposphere during the MOSAiC expeditions this is sufficient.

### 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.

We disagree! ... because we are convinced that a few key simulations as given in part 2 are sufficient to demonstrate that the observed smoke pollution levels (particle surface area concentration of  $10 \mu\text{m}^2 \text{cm}^{-3}$ ) are able to dominate ice nucleation and to suppress homogeneous freezing and to confirm in this way the hypotheses (based on all the observations) discussed in part 1.

And we asked ourselves: What is wrong with our simulations, findings, and conclusions when based on an excellent set of meteorological and environment input parameters? We have realistic (measured) temperatures, realistic (measured) humidity values, realistic (measured) smoke particle properties, and realistic updraft scenarios! Furthermore, if we look at all these simulation papers, most of them also focus on a few case studies because these few case studies contain the key messages.

But ok, we agree! If we want to 'quantify' the overall impact of wildfire smoke on Arctic cirrus formation we probably need a more sophisticated approach (e.g., Monte Carlo approach considering the full range of values of the atmospheric and environmental variables).

Nevertheless, we conducted a large number of simulations (manually!) and varied all parameters in their typical ranges. However, at the end, we wanted to provide the key messages only

Our 'strong statements' are just hypotheses and we are convinced that the reader will realize that we argue always in a hypothetical way. And even if we would have Monte-Carlo-like results, what would that help? Real proofs are not available. Hypotheses remain hypotheses.

Our hypotheses are reasonable, we think. They are, to our opinion, reasonable conclusions drawn from all the good observations and supported by realistic simulations. To our opinion the results are very robust. We are not aware of any other (remote sensing) approach that brought together such dense smoke aerosol and cirrus data sets (optical and microphysical properties plus height time displays providing clear impressions about virga evolution and entire cirrus life cycle). These data are clearly sufficient to draw conclusions on the potential capability of the detected smoke aerosol on cirrus formation.

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  $M_w, m_{ol}$  etc., this adds to the confusion.

We change it. Now we have 'Q' and 'q', no 'm' and 'M' anymore..

## 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.

Section 4.1 page 16 (extracted from the text in tex) ...

In our study, the onset ice saturation ratio  $S_{(i,on)}$  is defined by the ice saturation ratio  $S_{(i)}$  for which the ice crystal number concentration,  $n_{(i,hom)}$  or  $n_{(i,het)}$ , nucleated within  $\Delta t = 1\text{ s}$ , exceeds (for the first time) the value of  $0.001\text{ L}^{-1}$  in an ascending air parcel.

The onset ice saturation ratios are  $S_{(i,on)}=1.556$  and  $1.474$  for homogeneous and heterogeneous ice nucleation at  $199\text{ K}$ , respectively and, for  $213\text{ K}$ ,  $S_{(i,on)} = 1.515$  (homogeneous freezing) and  $1.349$  (heterogeneous ice nucleation).