

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 seem 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)