Response to the comments about the submitted paper: Modelling mixed-phase clouds with large-eddy model UCLALES-SALSA, Ref. ACP-2019-1182.

Dear Editor, dear Reviewers, we would like to thank the Editorial Board for considering our paper for publication in ACP and the reviewers for their constructive comments. We have addressed all of them and modified the paper accordingly. Our detailed answers follow. Text from the original manuscript that has been removed in the revised manuscript is marked in red. New text in the revised manuscript is marked in blue.

Answers to Reviewer 1

General comment R1.1 This paper offers a description of the microphysical updates regarding freezing processes in the LES model UCLALES-SALSA. A cloud case observed during ISDAC, that has been used for LES intercomparisons in the past, is also simulated here. This demonstrates the general agreement of the model with other LESs that are widely used for the study of mixed-phase clouds. A comparison of the newly implemented prognostic treatment of ice nucleation to a more simplified method is also presented. This paper will be useful to future users of UCLALES-SALSA, as it will serve as reference for the model's ice nucleation scheme. The few scientific findings are also interesting, specifically the role of INP treatment in cloud glaciation time and the impact of entrained INPs on ice formation throughout the cloud layer. For these reasons I recommend the paper for publication. However, I have a few suggestions that aim to (1) improve the documentation of the implemented freezing processes and (2) expand the scientific analysis and thus increase the scientific impact.
Answer to R1.1 We thank the referee for these comments and do our best to improve the manuscript according to the suggestions.

Major comment R1.2 Since this paper will likely serve as a documentation of the freezing processes in future studies conducted with this model, I recommend to provide a description of all processes in the Appendix, not just the immersion mode.
Answer to R1.2 We have now provided a detailed description for homogeneous and deposition freezing processes in the Appendices.

Major comment R1.3 The prognostic simulation is conducted with assumed aerosol concentrations to reconstruct an IWP similar to the ICE4 experiment. However I recommend to use aerosol measurements from ISDAC in an additional simulation (e.g. as in Savre and Ekman 2015) and compare the results to the observations. If the prognostic scheme results in good agreement with reality or not is a critical piece of information for the cloud modelling community. Moreover, you can conduct a few more sensitivity simulations and activate other freezing processes as well, and show how these experiments compare with microphysical measurements.
Answer to R1.3 There are some aerosol measurements covering number size distributions and bulk chemical composition but information on ice nucleation activity of different compounds is missing. When this information is missing, predicting ice number concentration is uncertain. For this reason, in the prognostic simulation (Sect. 3.3), we selected dust as a common INP type and a reasonable mixing state, and adjusted the contact angle to yield a similar IWP to that in the ICE4 experiment in the beginning of the simulation. A similar approach was used by Savre and Ekman (2015). As described in the manuscript, this approach was chosen so that our simulated IWP could be compared with the corresponding ICE4 simulations and because then the ice number concentration is close to the tipping point where cloud either stabilises or glaciates. It also turned out that this initially adjusted IWP eventually lead to number concentration values and cloud persistence seen in measurements.

Although it is not mentioned in the manuscript, we made various quick sensitivity simulations and tested different freezing mechanisms. These sensitivity simulations examined, for example, INP mixing state and contact angle. For the temperatures in the ISDAC case, both immersion and contact freezing can produce ice and the relevant mechanism depends on the mixing state of the INP. Test simulations showed that the outcome depends mostly on the resulting ice number concentration rather than the actual mechanism. For this reason, we showed just one prognostic case based on immersion freezing, which is the dominating mechanism for mixed-phase clouds. In answer to the question R1.5 we further clarify how our aerosol conditions match with observations.

Changes in the manuscript:

Remark about freezing processes in Sect. 2:

In our simulations (Sect. 3.3), only immersion freezing is active. This is done to keep the intercomparison simple enough and also due to high temperatureswhen immersion freezing is the dominant freezing mechanism. A more detailed description of the treatment of immersion freezing is given in Appendix A, when homogeneous freezing is not possible, and mixing state of the INP leading to aqueous droplets, when deposition and contact freezing are not feasible.

3rd chapter of Sect. 3.3 rewritten as:

To achieve the target IWP, we set INPadjusted accordingly the freezing properties of aerosols that can act as an INP. The total number concentration and freezing rate of INP appropriately. First, size distribution of the aerosol remain the same as in the fixed ice number simulations (Sect. 3.1 and 3.2), thus they are the thus same as in Ovchinnikov et al. (2014) . In the absence of more detailed aerosol observations, INP number concentration and mixing state, and contact angle were considered as adjustable parameters impacting ice nucleation ability. Here, contact angle represents the angle between the ice embryo and the ice nucleus in an aqueous medium.

First, in order to set the INP number concentrationis was by incorporating 2b bins that contained fractions x of dust and , we incorporated b bins (For bin description, see Sect. 2 and Fig. 1). Proportion $x = 0.015$ of the total aerosol number concentration was partitioned in b bins as INPs. Proportion $(1-x)$ of ammonium bisulphate, where $x = 0.015$. The number concentration of dust containing particles is 238.5e3 and 9.75e3 remained in a bins. Resulting number concentrations of INPs in accumulation and coarse modes were 238.5×10^3 and $9.75 \times 10^3 kg^{-1}$, respectively. Here, the

Second, the INP mixing state was adjusted so that the particles in the b bins were set to have an insoluble dust core(50 , 50% of the dry mass) acts as the INP. The total number concentration of the aerosol, and thus also the droplet number concentration in cloud, is the same as in fixed ice number simulations (Sect. 3.1 and 3.2). Second, and ammonium bisulphate for the other half. Here, dust acts as the INP.

Third, the freezing rate was adjust adjusted by setting the cosine of the contact angle of dust to $m_{is} = 0.57$ (Eq. A3 in Appendix A). Contact angle represent the angle between the ice embryo and the ice nucleus (IN) in an aqueous medium. These two parameters (

It should be noted that the target IWP could have been reached using different combinations of INP mixing state, x and m_{is} but these simulations showed that the results depend mostly on the resulting ice number concentration rather than the applied parametrisation. These characteristics of aerosol are uniform throughout the whole simulation domain.

Minor comment R1.4 Line 40: Do you mean that a high aerosol load is associated with higher occurrence of mixed- phase clouds or with more liquid in the mixed-phase clouds? Please clarify.
Answer to R1.4 We clarified in the manuscript that a high aerosol load is associated with higher occurrence of mixed-phase clouds.

Minor comment R1.5 Lines 246-249: I don't see any point in comparing with observations since you simulated random aerosol conditions and not the observed.
Answer to R1.5 We have sharpened our statement how the initial aerosol number concentration and size distribution is the same as in Ovchinnikov et al (2014). Ovchinnikov et al (2014) cites that these parameters provide the best fit to the measured distributions below the liquid cloud layer (Earle et al., 2011). However, in our prognostic simulation, we altered the number of aerosols that contain an ice nucleating core and the contact angle between the ice embryo and the ice nucleus. This latter quality of the aerosol condition, i.e. freezing rate efficiency, is not available from observations.

Changes in the manuscript:

3rd chapter of Sect. 3.3 rewritten as already given in the answer to the question R1.3.

The paragraph that the referee mentioned:

Figures 7a and 7b illustrate ~~, how~~ that super-cooled liquid droplets are dominant in the upper layers of the mixed-phase cloud compared to ice crystals. Here the total ice number concentration stabilises at approximately $0.44 L^{-1}$, whereas it is obvious from Sect. 3.2 that a much higher concentration is needed to completely glaciate the cloud. ~~Similarly~~Correspondingly, the cloud droplet number concentration ~~is~~ stabilises at approximately 175 ~~. Although our original goal was not to compare directly against observation~~cm^{-3}. Remarkably, these values are in ~~good~~ line with aircraft observations (Flight F31) of this ISDAC case. The observed ice and cloud droplet number concentration are $0.35 L^{-1}$ and 185 cm^{-3}, respectively (McFarquhar et al., 2011; Savre and Ekman, 2015). ~~Concentration~~ Ice number concentration is also approximately two orders of magnitude less than the number concentration of efficient ~~IN~~ INPs above the cloud layer. From that we can estimate that the concentration of ~~IN~~ INPs entraining from above the cloud should be in the order of 0.1 to 1.0 cm^{-3} to glaciate the cloud.

Minor comment R1.6 Both INP and IN terms are used. I suggest to use the same term throughout the text for consistency (I think 'INP' has become more popular in the past few years)
Answer to R1.6 Manuscript was corrected to use only INP terms as suggested.

Answers to Reviewer 2

General comment R2.1 This study adds a heterogenous ice nucleation parameterization to the UCLALES- SALSA model. The model is tested with fixed ice crystal number concentration by using a case from the ISDAC campaign that was the focus of an intercomparison study. This paper is well written, and the figures clearly illustrate the main points.

As to the results of the study, allowing prognostic INP will reduce the number of ice crystals because of precipitation, causing there to be more sustained cloud liquid, but how is this a new result? Many studies have already shown this (Fridlind et al. 2012; Solomon et al. 2015; Solomon et al. 2018).

Also, the variability in the control studies differ significantly from the ISDAC intercomparison, which needs to be explained.

Also, it needs to be explained how aerosol concentration above cloud top were chosen and what role the prognostic CCN is playing in the simulations.

This model will be a very useful tool for studying mixed-phase cloud processes, but I think this study is better suited for a technical report than a scientific publication.
Answer to R2.1 We thank the referee for these comments and agree that the part describing the ice microphysics of UCLALES-SALSA for the first time is technical in nature. However, we argue that the manuscript also contains new scientific findings, such as the impact of entrained INPs on ice formation throughout the cloud layer, which Referee 1 also highlighted. We have added citations to the articles the referee mentioned and now state that our findings regarding prognostic INP and cloud resilience are in line with previous modelling studies. Here below, we answer in detail to the remarks raised in this general question.

We cite the articles the referee mentioned and state the study is also in line with previous modelling studies regarding prognostic INP and cloud resilience.

Changes in the manuscript (Sect. 3.3):

In the beginning of the prognostic ice run, domain mean of dust containing aerosols is approximately $27L^{-1}$. After 32 hours of simulation the same mean value is about $13L^{-1}$. Here, the loss of INPs limits the ice number concentration. The mixed-phase cloud persists because the ice number concentration can change. This is so-called self-adjustment of INPs which better reproduces observed evolution of mixed-phase clouds since usually they are more resilient in observations than in models(Andronache, 2017; Morrison et al., 2011a). This is also in line with previous modelling studies, where prognostic INP will reduce the number of ice crystals because of precipitation, thus allowing cloud liquid to sustain (Fridlind et al., 2012; Solomon et al., 2015, 2018).

Changes in Conclusions:

In the second part, we constructed a case where ice formation is modelled using a ~~heterogenous~~ heterogeneous ice nucleation scheme and a prognostic ice nucleating particle population containing mineral dust. This so-called prognostic ice simulation was designed so that it matched with the previous fixed ice number concentration simulation where the cloud was close to the tipping point. When the simulation with fixed ice concentration showed a complete glaciation after about 12 hours, the prognostic ice simulation reached an equilibrium state which lasted up to end of the 32 hour simulation. With this the prognostic simulation showed the importance of the self-adjustment of ice nucleation active particles. This is in good agreement with previous modelling studies (Fridlind et al., 2012; Solomon et al., 2015, 2018) and a observational study where resilient mixed-phase clouds are seen together with relatively high ice nuclei concentrations (Filioglou et al., 2019).

We reply to the comment about the variability in the control studies in the answer to question R2.3.

We specify in the manuscript that the initial aerosol concentration and size distribution is uniform throughout the domain thus the concentration above cloud top is not any different than elsewhere. Answer to the comment about role of prognostic CCN is given in the answers to questions R2.2 and R2.4.

Major comment R2.2 Need to include basic detailed about the model in Section 2 even though they may be available in other papers. All details needed to understand the simulations need to be included in this section (CCN activation, etc).
Answer to R2.2 We rewrote the Model description (Sect. 2) particularly regarding radiative cooling, CCN activation and ice microphysics. A more detailed description of freezing processes is also given in the Appendices.

Changes in the Sect. 2 in the revised manuscript:

First paragraph of Sect. 2.

The UCLALES-SALSA (Tonttila et al. 2017) model consists of two components: first, the widely used large eddy simulator UCLALES~~that handles the atmospheric dynamics including turbulence~~ (Stevens et al., 1999, 2005), and second the aerosol bin microphysics model SALSA (Sectional Aerosol module for Large-Scale Applications) (Kokkola et al., 2008; Tonttila et al., 2017; Kokkola et al., 2018). UCLALES handles e.g. surface fluxes, transportation of microphysical prognostic variables and atmospheric dynamics including turbulence. The previous version of UCLALES-SALSA incorporated interactions between aerosols, clouds and drizzle (Tonttila et al. 2017). Now we have extended the model with a description for ice crystals. In this study, we focus on how ice crystals and ice nucleating particles (INP) interact with clouds while tracking sectional aerosol size distribution.

5th paragraph of Sect. 2. and onwards rewritten:

In UCLALES-SALSA, recently implemented processes involving ice crystals are droplet freezing, deposition of water vapour, sublimation, melting when $T > 0°C$, coagulation between different sized hydrometeors, sedimentation, and interactions with radiation (see also Fig. 1). Most of these processes are included in ~~the same~~ a similar way as in the previously published version of UCLALES-SALSA (Tonttila et al. 2017). ~~For instance, interaction with radiation is~~

implemented with the same four-stream radiative transfer solver (Fu and Liou, 1993) as before but extended to include ice crystals. Condensation of water vapour to ice crystals is based on the analytical predictor of condensation (APC) scheme by Jacobson (2005) and implemented following Tonttila et al. (2017) (Eqs. 7 and 8). For solids, the condensation does not require Kelvin or Roult terms. Furthermore, UCLALES-SALSA was upgraded with minor bug fixes and improvements. For example, hygroscopicity is now calculated with κ-Köhler (Petters et al., 2006; Petters and Kreidenweis, 20 instead of previously used ZSR method (Stokes and Robinson, 1966).

Regarding the scope of this study, we describe droplet freezing in higher detail. There are five mechanisms for droplet freezing and they are all currently implemented in UCLALES-SALSA.

- Immersion freezing is possible for aqueous droplets that have an insoluble core, which in UCLALES-SALSA is either dust (DU) or black carbon (BC). The rate of heterogeneous germ formation in a supercooled droplet of water or solution is calculated mostly following Khvorostyanov and Curry (2000) and additional parameters are from Jeffery and Austin (1997), Khvorostyanov and K. Sassen (1998), Khvorostyanov and Curry (2004) and Li et al. (2013). See also App. A.

- Homogeneous freezing is possible for any aqueous droplet with or without insoluble particles. This is applied to the model according to Khvorostyanov and K. Sassen (1998). See also App. B.

- Deposition freezing is possible for dry insoluble aerosol at sub-saturated conditions ($RH <$ 100%). This is implemented following Khvorostyanov and Curry (2000) and additional parameters from Hoose et al. (2010). See also App. C.

- Contact freezing is implemented in UCLALES-SALSA following Hoose et al. (2010) so that first the coagulation code is used to calculate collision rates between dry particles and liquid droplets and then immersion freezing code gives the freezing probability.

- Condensation freezing is implemented as a part of immersion freezing, because these droplets can freeze during the modelled condensational growth.

In our simulations (Sect. 3.3), only immersion freezing is active. This is done to keep the intercomparison simple enough and also due to high temperatureswhen immersion freezing is the dominant freezing mechanism. A more detailed description of the treatment of immersion freezing is given in Appendix A, when homogeneous freezing is not possible, and mixing state of the INP leading to aqueous droplets, when deposition and contact freezing are not feasible.

Deposition of water, i.e. diffusion limited condensation or evaporation of water vapour, is defined for aerosol when relative humidity (RH) is over 98% and always for other hydrometeors. This is based on the analytical predictor of condensation (APC) scheme by Jacobson (2005) and implemented following Tonttila et al. (2017) (Eqs. 7 and 8). According to this definition, the particles compete for the available water vapour. For solids, the condensation equation does not require Kelvin or Raoult terms.

Activation of aerosols to cloud droplets happens when RH is over 100% and aerosol wet diameter exceeds the critical limit corresponding to the resolved supersaturation. At this time, a certain proportion of activated aerosols (i.e. cloud condensation nuclei, CCN) are moved to cloud droplet bins.

Sedimentation is defined as before in Tonttila et al. (2017) and now extended for ice particles. For simulations in this study, a fall rate of ice particles is set as in Ovchinnikov et al. (2014).

Coagulation is implemented with the same way as before and now including also ice particles. Coagulation is affected by diffusion, especially aerosols, and by sedimentation, especially large particles. In a collision, bigger particles absorb smaller particles.

Interaction with radiation is implemented either with the same four-stream radiative transfer solver (Fu and Liou, 1993) as in Tonttila et al. (2017) with extension to include ice particles or parametrised as in Ovchinnikov et al. (2014). We used the latter method in our simulation. In the parametrised radiation, the net upward long-wave radiative flux is computed as a function of liquid water mixing ratio profile. The effect of interaction with radiation can be seen in simulations how radiative cooling weakens after liquid water path decreases below a specific value.

Furthermore, UCLALES-SALSA was upgraded with minor bug fixes and improvements. For example, hygroscopicity is now calculated with κ-Köhler (Petters et al., 2006; Petters and Kreidenweis, 2007) instead of previously used ZSR method (Stokes and Robinson, 1966).

Major comment R2.3 This model is clearly more sensitive to ice formation than all the models included in the ISDAC intercomparison. It is important to understand why to understand the sensitivity studies with the new ice nucleation parametrisation.
Answer to R2.3 One contributing reason is that dry particle size is tracked in UCLALES-SALSA instead of ice crystal size and this lower size resolution seems to have an effect on ice crystal sedimentation. The other reason is related to the model dependent technical implementations such as the advection flux limiter method. Overall, the initial profiles in the presented case study are such that even a small decrease in LWP leads to decreased radiative cooling and turbulence, and this will prevent mixing of moisture from low altitude to cloud base. Thus, the model with default setup is more sensitive to ice formation close to the tipping point where technical details have largest impact.

Changes in the related paragraph in the revised manuscript:

Compared to the model results in Ovchinnikov et al. (2014), IWP in UCLALES-SALSA declines faster after the peak IWP has been reached in ICE4. One reason for this is that dry particle size is tracked in UCLALES-SALSA and this seem to have an important effect on ice crystal sedimentation. The other reason is related to the model dependent technical details~~such as the advection flux limiter method~~. In Ovchinnikov et al. (2014) it was also stated that when the ice number concentration gets higher the differences between models are more caused by discrepancies in microphysics than cloud dynamics. This underlines the sensitivity and significance of microphysics.

Major comment R2.4 How is droplet number concentration specified in the ISDAC ICE4 simulation? Is this prognostic? If so, it would be insightful to see the droplet number concentration in Figure 4. Is this why the results are so different than the intercomparison?
Answer to R2.4 We have now added the droplet number concentration time series to Fig. 4. Droplet number concentration is prognostic in all fixed ice and prognostic ice simulations. Droplet number concentration decreases when ice number concentration is increasing but that is not the driving force behind complete removal of liquid phase, as explained below in the revised manuscript text. Additional explanation to this question is given also in the answer to the question R2.3.

Changes in the related paragraph in the revised manuscript:

Figure 4a shows that in the prognostic ice simulation LWP starts to increase after 4.5 hours of simulation. This is caused by a decrease of ice number concentration (Fig. 4c) to such a low level at which leaves which allows more water vapour for condensation to liquid droplets. The same figure also depicts how the ice number concentration is set to a target value (simulation ICE4) and how the concentration is stable until the cloud dissipates. Figure 4d depicts how droplet number concentration lowers especially right after spinup period when ice number concentration is increasing. However, changes in droplet number concentration is not the driving force behind complete removal of liquid phase. Figure 4e illustrates how the whole cloud with prognostic droplet freezing descends and how the ICE4 is affected by entrainment both below and above the cloud,cloud gets thinner and dissipates. In all simulations, droplet number concentration is specified as prognostic variable.

Major comment R2.5 It is not clear how the artificial movement of aerosols between bins for numerical stability is affecting the results (lines 240-243).
Answer to R2.5 The movement of aerosols between bins is a feature of the model that is needed for stability. This explains the numerical artefact seen in Fig. 6, but has no effect on the results. This is now stated in the manuscript.

Related part of the paragraph in the revised manuscript:

The increase in the total number of particles in bin 1 is a numerical artefact caused by the bin adjustment routine, which can move particles from one bin to another in order to keep the dry size within the predefined bin limits. When a large fraction proportion of particles in bin 2 are activated as cloud droplets, some of the remaining are moved to the smaller bin to avoid numerical problems. However, this numerical artefact does not affect the results.

Major comment R2.6 Please explain why droplet freezing occurs throughout the cloud while for the same case Savre and Ekman (2015) found droplet freezing at cloud top. A more detailed discussion of how aerosols and droplet and ice crystal activation are represented in the two models is needed to understand why simulations in the two studies differ.
Answer to R2.6 Upon closer inspection, we found that the comparison to Savre and Ekman (2015) was somewhat flawed and hence the comparison was removed from the manuscript. They used a different cloud case and this could explain the differences in droplet freezing profiles.

What happens within the cloud layer in our model is that, when supersaturation and cloud activation are explicitly modelled as in UCLALES-SALSA, unactivated particles can penetrate through the cloud layer in a down-draft with low supersaturation and later come back to the cloud with up-drafts and activate due to the higher supersaturation. Then freezing happens in the up-drafts throughout the cloud. If activation is not modelled with this level of details (any model,not just Savre and Ekman, 2015), activation and freezing might happen too early or late and in a wrong part of the cloud

Changes in the manuscript:

Figure 7c further illustrates an interesting behaviour of ice particle formation. In the beginning of the simulation ice particles are formed throughout the cloud, but later the most intensive formation takes place at the top of cloud where fresh IN particles INPs are entrained into the cloud layer. However, the maximum supersaturation in these entraining downdrafts is so low, that only the largest particles are able to form cloud droplets and consequently freeze. The smaller ones penetrate through the cloud layer as interstitial aerosol particles (i.e. unactivated particle), and are able to form cloud droplets (i.e. activate) and ice particles at the cloud base when they are recirculated back to the cloud with higher supersaturation. This can be well seen at the end of simulation as there is two peaks in vertical profile of freezing rate. Such phenomena can be only simulated with explicit calculation of in-cloud supersaturation and representation of aerosol size distribution and chemical composition like is done in UCLALES-SALSA. ~~Compared to Savre and Ekman (2015), where most intensive freezing is at the cloud top, in UCLALES-SALSA droplet freezing occurs throughout the cloud~~If activation is not modelled with this level of details, activation and freezing might happen too early or late and in a wrong part of the cloud. Overall, Figs. 6 and 7c nicely demonstrate how the relative ~~fractions~~ proportions of particles in different hydrometeors are size dependent and how sectional description for aerosols is required to be able to simulate such processes in LES models.

Major comment R2.7 Lines 275-277: More details of the simulations are needed to understand whether this is a correct statement.
Answer to R2.7 This comment has been addressed in answer to question R2.3

Minor comment R2.8 Line 198: ". . ..concentration is was. . .". Please reword.
Answer to R2.8 We rewrote the related paragraph.

Minor comment R2.9 Line 202: "was adjusted"
Answer to R2.9 Spelling corrected as suggested.

Minor comment R2.10 Line 203: "represents"
Answer to R2.10 Spelling corrected as suggested.