

Dear Martina,

Thank you for your continued work on our manuscript. Please find below details of the manuscript changes in response to the second round of reviewer comments.

Best wishes,

Rachel Hawker.

Replies to Report #2

In this work the authors investigate the effect of the parameterization of ice nucleation particles (INP) on the development of convective clouds and their radiative properties. The authors perform simulations of a tropical convective event using a set of different INP parameterizations with diverse dependency on aerosol concentration and temperature. Their work demonstrates that ice initiation may affect strongly the radiative properties of these systems, and in contrast to previous work, such an effect is not buffered by secondary ice production. This is a well-written paper, relevant to the atmospheric community. I'd recommend its publication in ACP after a few minor clarifications.

Thank you for your work and your helpful comments on our paper.

Comments.

This paper has gone already through a review process, which resulted in an improved work. Hence, I just have a minor comment. The authors must emphasize that the INP parameterizations refer only to immersion freezing and provide some information (probably in section 2.1.2) on the other ice formation mechanism in the model. I am referring specifically to contact ice nucleation in the convective cloud, and the in situ formation of cirrus by heterogeneous ice nucleation by deposition, and, by the immersion and homogeneous freezing of haze droplets at $T < 235$ K. The latter could be quite relevant since in situ cirrus may interact with anvil particles and may either exacerbate or negate the observed radiative impact.

Change to paper: The following text has been added to Section 2.1.2: *"The INP parameterisations tested in this study represent only immersion freezing. Heterogeneous ice nucleation by deposition and contact nucleation are not represented. Other mechanisms of heterogeneous ice formation should be tested and included in future studies but was beyond the scope of this work. However, immersion freezing is expected to be the dominant mechanism of heterogeneous ice formation in convective clouds (Ansmann et al., 2008; De Boer et al., 2011; Kanji et al., 2017) and therefore the simulations presented here should capture the majority of heterogeneous ice nucleation relevant for cloud properties. Immersion and homogeneous freezing of haze droplets are not represented, but it is unlikely that they contribute significantly to ice crystal number concentration in the main anvil cloud derived from mixed-phase cloud regions. However, the importance of these mechanisms on anvil cloud properties should be investigated in future work."*

Replies to Report #3

In this study, the authors test the sensitivity of a mix of Eastern Atlantic Tropical cloud types to INP parametrizations with different temperature dependencies as well as the Hallett-Mossop process. They show that the slope (temperature dependence) of INP parametrizations has a significant impact on the TOA outgoing radiation for the clouds studied. Additionally, even in the presence of the Hallett-Mossop process, the INP parametrizations still produce significant results, indicating that correctly parameterizing the temperature-dependence of INPs is important to accurately simulate the radiative properties of clouds.

The manuscript is well written and the explanations are well thought through and explained nicely. The authors have also done a nice job responding to the previous reviewer comments. Nevertheless, I still think some points could be better explained as well as have a few additional comments listed below. The line and figure numbers correspond to the line numbers in the track changes version of the manuscript with the reviewer comments and author responses.

Thank you very much for your constructive comments on our paper.

General/minor comments:

As previously mentioned, it is not immediately clear which hydrometeor classes are able to gain mass through riming and therefore produce secondary ice through the Hallett-Mossop process (HMP). Please clarify this as in several places, ice crystals are mentioned specifically but no other ice phase hydrometeor classes (snow and graupel) are mentioned. In response to a previous reviewer comment it is stated that ice crystals are meant to represent all cloud ice but for clarity it might be easier to replace ice crystals with ice phase hydrometeors or something more inclusive in the text when referring to the HMP. See specific line numbers below.

Reply: Ice crystals, snow and graupel can all gain mass through riming. However, only riming from snow and graupel species produce splinters via the Hallett-Mossop process. This is the case because the majority of riming occurs on snow and graupel particles with a negligible contribution from cloud ice or small ice crystals which are converted to graupel before substantial riming, and therefore rime-splintering, can occur. We appreciate this was not clear previously and have clarified this in the locations in the text highlighted in your minor comments below. When we refer to ice crystals initiating the Hallett-Mossop process, we are referring to an indirect initiation by allowing the earlier formation of snow and graupel in the cloud, and therefore driving the subsequent riming and splinter production that follows snow and graupel formation.

As previously mentioned by a reviewer, it is stressed that INPs are not scavenged in the text. But what does this actually mean? In the response, the authors state that as long as a threshold number of droplets is present and based on the ICNC already present and T , a freezing rate is calculated. This makes me wonder if the ICNC in a grid box is already above the predicted number of ice crystals from a parametrization due to settling (lifting) of ice from above (below), is no new ice formed? Please clarify this and also discuss what impacts this might have on the influence of the INP parametrizations used and the resulting TOA radiation if this is the case.

Reply: The reviewer is correct that if the ICNC in a grid box is already above the predicted number of ice crystals from a parameterisation due to settling (lifting) of ice from above (below) no new ice is formed. This is the main caveat of this study resulting from the exclusion of aerosol scavenging. The INP parameterisation computes the change in ICNC resulting from INP at the timestep after the ice nucleation occurs. This means that in the case of strong updraft, such as in the deep convective core, it is our expectation that the representation of heterogeneous freezing accounts for scavenging in a primitive way and should come relatively close to what we would expect if the background aerosol was being removed by ice nucleation. In the case of strong

sedimentation of ice crystals from above, the lack of scavenging would lead to an underestimation of heterogeneous freezing. However, as the terminal velocity of ice crystals is very low, this would not be expected to play a large role except in very weak updrafts outside of the deep convective core. Scavenging can have counterintuitive effects on the cloud properties making speculation on its effect difficult but we agree that this is an important question to address in future work.

Change to paper: The following bold text has been added to the limitations section of the paper (Section 4) to make clearer the limitations of excluding aerosol scavenging: “The effect of INP parameterisation choice on convective cloud field properties should also be examined with the inclusion of aerosol scavenging but this was beyond the scope of this study. ***Aerosol scavenging would allow the aerosol number concentration to be reduced by cloud droplet activation and the number of dust particles within cloud droplets to be tracked and depleted when frozen heterogeneously. In the simulations presented here, the heterogeneous freezing rate is calculated using the interstitial aerosol number concentration and the ICNC of the gridbox in question meaning that ice crystals advected into the gridbox will reduce the heterogeneous nucleation rate even if they were frozen elsewhere in the domain.***”

Similarly, a threshold number of cloud droplets is used. But what is this number? Is it large enough so that there are always enough cloud droplets such that the number of formed ice crystals does not exceed the number of cloud droplets? Please clarify this and discuss the impacts on the results if this is not the case.

Reply: The gridbox conditions (temperature, cloud droplet number, ICNC, and aerosol concentrations) are inspected and the number of heterogeneously formed ice crystals is calculated if the cloud droplet number concentration exceeds $1.0\text{e-}6 \text{ kg}^{-1}$. If the number of heterogeneously formed ice crystals calculated by the parameterisation exceeds the cloud droplet number concentration, all the cloud droplets in a gridbox are frozen. The number of formed ice crystals cannot exceed the number of cloud droplets.

“Hallett Mossop” should be changed to “Hallett-Mossop” throughout the text.

Change to paper: As suggested.

It is not always clear if the results are referring to the TOA changes in radiation over the entire daylight period (10-17 UTC) or the entire simulation (10-24). Please make that clearer in the text and the figure captions rather than just mentioning it in the Methods.

Change to paper: The following figure captions have been altered to make clear what time period the results depict:

Caption Figure 4: “Effect of INP and secondary production on top of atmosphere (TOA) **daytime (10:00-17:00 UTC)** outgoing radiation.”

Caption Figure 5: “INP and TOA outgoing **daytime (10:00-17:00 UTC)** radiation from cloudy regions.”

Caption Figure 6: “Outgoing **daytime (10:00-17:00 UTC)** radiation from cloudy regions and INP parameterisation slope.”

Caption Figure 8: “Vertical composition of cloud. 2D distribution of cloud type at 20:00 for all six SIP_active simulations (a-f), as well as anvil and domain cloud fraction change **(10:00-24:00 UTC)** due to INP”

Caption Figure A4. “Effect of INP on domain-mean TOA outgoing daytime **(10:00-17:00 UTC)** shortwave and longwave radiation.”

Additionally, the following sentences at the start of the relevant results sections have been altered to make clear that they are discussing the daytime outgoing radiation:

Start of Section 3.1: “We first examine the effect of INP parameterisation on the **TOA outgoing daytime (10:00-17:00 UTC)** radiation relative to the simulation where the only source of primary ice production was through homogeneous freezing (NoINP).”

Start of Section 3.2: “Here we discuss the changes in **daytime** outgoing radiation from cloudy regions only due to INP parameterisation choice.”

Start of Section 3.3: “Overall cloud fraction is increased by INP for all INP parameterisations and these increases in cloud fraction contribute about as much to changes in domain-mean **daytime** radiation as the changes in outgoing radiation from cloudy regions”

I find the equations nice to explain the different calculations that have been conducted. However, there is no return to the acronyms used in the equations and therefore it is not immediately clear which values are calculated with which equation. Consider integrating the equation acronyms into the text and figures. Although, in general I find the text explanations quite clear, but also mentioning the acronyms, especially in the figures might make things clearer as how things were calculated.

Change to paper: The following figure captions have been altered to refer back to the terms calculated in Section 2.1.3.:

Caption Figure 4: “In (a), the change from the NoINP simulation is shown ($INP - NoINP$) with SIP active. In (b), the change from SIP_active to SIP_inactive is shown ($SIP_active - SIP_inactive$). A positive value indicates more outgoing radiation when INP or SIP are active. In (a) and (b), the relative contributions of changes in outgoing radiation from cloudy regions (left) (i.e. ΔRad_{REFL} from Eq. (1)) and cloud fraction (middle) (i.e. ΔRad_{CF} from Eq. (2)) to the total radiative forcing (right) (i.e. ΔRad_{s-r} from Eq. (5) with simulation *s* referring to simulations with INP active in (a) and to the SIP_active simulations in (b) and simulation *r* referring to the NoINP simulation in (a) and to the SIP_inactive simulations in (b)) are shown (calculation described in Sect. 2.1.3).”

Caption Figure 5: “Absolute change in outgoing shortwave, longwave and total radiation from cloudy regions relative to the NoINP simulation (i.e. ΔRad_{cl} used in Eq. (1)) (a)”

Caption Figure A4: “Effect of INP on domain-mean TOA outgoing daytime (10:00-17:00 UTC) shortwave and longwave radiation. The change from the NoINP simulation is shown ($INP - NoINP$). A positive value indicates more outgoing radiation when INP are present. The contributions of changes in outgoing radiation from cloudy regions (left) (i.e. ΔRad_{REFL} from Eq. (1)) and cloud fraction (middle) (i.e. ΔRad_{CF} from Eq. (2)) to the total radiative forcing (right) (i.e. ΔRad_{s-r} from Eq. (5) with simulation *s* referring to the simulations with INP active and simulation *r* referring to the NoINP simulation) are also shown (calculation described in Sect. 2.1.3).”

It is stated in the manuscript that there was little sensitivity to the time frame (day versus day and night) considered for the impacts on TOA outgoing radiation. However, I find this quite surprising especially in light of the results in the convective anvils (high clouds). As at night when no shortwave radiation is reflected and the longwave is the only outgoing radiation, the cirrus anvil extent will likely have a large impact on the TOA outgoing radiation. Was this investigated and see comment below?

Reply: The effect of the INP parameterisation on the outgoing radiation in the night-time was not investigated in detail due to the short length of our simulations. When the spin-up period is excluded from the analysis period, less than a full 24 hours of simulation time remains. Thus we don't have data from a full night-

time period. As such, we decided to focus on the daytime hours for which we have simulation data for all hours in question to avoid an arbitrary bias in the results due to simulation length. However, the effect of including the night-time hours (10:00-23:45 UTC) on the outgoing longwave radiation and the cloud fraction was tested and found not to have a large effect on the stated change in the outgoing longwave radiation due to the inclusion of heterogeneous ice nucleation, particularly relative to the very large changes in the outgoing shortwave radiation. This is what we refer to when we say there was little sensitivity to the time frame. The overall outgoing radiation will of course be sensitive to the inclusion of the night-time hours owing to the absence of outgoing shortwave at night-time. This has been clarified in Section 2.1.3.

Change to paper: The following bold text has been inserted in Section 2.1.3. ***“The sensitivity of the outgoing longwave radiation and the cloud fraction to time period selection was tested and found to have little impact. The overall outgoing radiation (shortwave + longwave) will be sensitive to the time period selection owing to the absence of outgoing shortwave radiation at night-time. We focus on the radiation during daylight hours only because our simulation is only 24 hours in length owing to computational restrictions and therefore when the spin-up period is excluded from the analysis, less than 24 hours of simulation data remains with much of the night-time hours removed with the spin-up period.”***

As I am unfamiliar with the SOCRATES radiation scheme, do the clouds ever become saturated in the amount of radiation that they can emit? I would think this would occur quite quickly in the cirrus anvils.

Reply: Clouds do not become saturated in the amount of radiation they can emit. Even for high optical thicknesses, e.g. at the centre of the anvil over the convective core, there should be changes (however small) in the cloud reflectivity due to the cloud properties.

The authors do a nice job of showing that the slope of the INP parameterization used influences the strength of the changes in TOA outgoing radiation. However, as the A13 parameterization has the steepest slope at temperatures above ~-26 C but also no slope at colder temperatures, how does that impact the argument that the slope of the parameterization is critical? Perhaps it is better to state that the number of INPs at cold temperatures is more important than at warmer temperatures in the MPC cloud regime?

Reply: The reviewer is correct that at high dust concentrations the slope of the N12 and A13 parameterisation is flat owing to the plateau in INP number concentrations that occurs once the parameterisations reach the number concentration of dust represented in the model gridbox in question and therefore the number concentrations of aerosols capable of nucleating ice and the parameterisation slope are not decoupled in this work. The relative importance of these two variables is being investigated in current, as yet unpublished, work, and we have added a paragraph detailing this caveat to the limitations section (Section 4).

Change to paper: The following text has been added to Section 4 of the paper: *"Furthermore, while many cloud macro- and microphysical were correlated with INP parameterisation slope, the slope of the parameterisation at low temperatures for the A13 and N12 parameterisations can be flat because the parameterisations plateau once they reach the number concentration of dust represented in the model gridbox in question. This means that at high dust concentrations, the slope of the INP parameterisation correlates with the INP concentration at temperatures between -25 and -35°C (Figure 2). This means that the absolute number concentration of aerosols capable of nucleating ice is not decoupled from the INP parameterisation slope in some INP parameterisations and that some cloud responses attributed to changes in the INP parameterisation slope may have in fact been caused by the absolute INP number concentration at cold temperatures. The relative importance of the INP parameterisation slope and the absolute number concentration of aerosols capable of nucleating ice will be investigated in future work. However, whether the INP number concentration plateaus at cold temperatures is determined in part by the INP parameterisation slope, and correlations with INP parameterisation slope are evident at both warm and cold cloud altitudes indicating the importance of the INP parameterisation slope."*

It is mentioned clearly that more studies are needed to investigate what the impacts of adding INPs has over a longer period. However, is it possible to hypothesize on what impacts the increase in TOA outgoing radiation has on subsequent cloud development on subsequent days? More specifically would the reduction in surface temperature reduce the ability of convective clouds to form and ultimately over a long period offset any changes to the TOA outgoing radiation?

Reply: It is certainly possible that reductions in surface temperature due to the radiative changes reported in the paper could affect cloud formation beyond the simulation length. However, due to the complex interactions between multiple hydrometeor and cloud types in these simulations, we don't feel able to give a reliable prediction of the effect of the changes in radiation presented on cloud formation beyond the time period analysed. For example, as noted in our results, while our simulations show an increase in domain cloud fraction within our analysis period due to the inclusion of INP, the anvil cloud fraction is substantially reduced. As anvil cloud can persist in the atmosphere longer than the convective cloud that forms it, it is possible that the reductions in anvil cloud could become more important to the overall cloud signal over a longer time period. Many factors will contribute to changes in the cloud formation in the day(s) after the simulation end including changes in the moisture and temperature profiles, and convergence lines due to large scale flow making speculation difficult. Furthermore, as our domain is over the ocean, the surface temperature will not react very quickly to changes in the radiation.

Minor comments:

Line 170: Is the rime mass calculated for the snow hydrometeor class or only for the ice crystal class? Please clarify here as well as in the following comments on this.

Reply: The rime mass is calculated from snow and graupel species. We appreciate this was not clear previously and have clarified this wherever necessary.

Change to paper: "The rate of splinter production per rimed mass is prescribed with 350 new ice splinters produced per milligram of rime at -5°C. **Splinters are produced from rime mass of snow and graupel.**"

Line 190: Here it is stated that the radiative cloud properties are not affected by changes in ice or snow number or any changes to rain and graupel. However, based on the following lines it sounds like the cloud radiative properties are sensitive to the mass of these hydrometeor classes. If that is the case, please specify that in this sentence as the mass is sensitive to the number/size and therefore this sentence is potentially misleading.

Change to paper: “It does not **explicitly consider** changes in ice crystal or snow number concentration or size (though changes in number and size will affect mass concentrations which are considered), and does not consider any changes to rain or graupel species.”

Line 221: Please clarify why the change in radiation from clear sky areas is only multiplied by the clear sky fraction of the sensitivity run (s) and not the change in the clear sky fraction from the sensitivity run and reference run (s-r), as is done for the cloudy sky fraction.

Reply: The equation in question is actually the combination of two interaction terms. The first ($\Delta Rad_{cl} \times \Delta cf$) is the change in radiation caused by changes in cloud albedo that occurs in regions where there was previously no cloud, i.e. the change in domain-mean outgoing radiation that can be attributed to regions of new cloud in simulation s having a different albedo to the cloud that was present in simulation r. The second ($\Delta Rad_{cs} \times (1 - cf_s)$) is any change in clear sky outgoing radiation (a kind of clear sky albedo), and is applied to all clear sky areas, which accounts for any small bit of cloud mass that may be excluded from our cloudy regions if it falls below the cloud mass threshold. Both of these terms are near zero and negligible to the overall change in outgoing radiation so they were combined into one interaction term. They have now been separated into two different terms now shown in Eq. 3 and Eq. 4.

Change to paper: Section 2.1.3 has been altered with the previous Eq.3 now split into Eq. 3 and Eq.4 to calculate the contribution of the interaction between cloud fraction and cloud albedo changes (ΔRad_{INT}) and the contribution of clear sky albedo (ΔRad_{CSKY}) changes separately:

“There is interaction between the outgoing radiation from cloudy regions and cloud fraction changes (ΔRad_{INT}) which is calculated in Eq. (3).

$$\Delta Rad_{INT} = \Delta Rad_{cl} \times \Delta cf \quad (3)$$

The contribution of changes in the outgoing radiation from clear sky areas (ΔRad_{CSKY}) can be calculated as shown in Eq. (4).

$$\Delta Rad_{CSKY} = \Delta Rad_{cs} \times (1 - cf_s) \quad (4)$$

Where ΔRad_{cs} is the change in mean outgoing radiation from clear sky areas between simulations s and r and cf_s is the cloud fraction of simulation s.

The total outgoing radiation difference between simulations s and r (ΔRad_{s-r}) is therefore as shown in Eq. (5).

$$\Delta Rad_{s-r} = Rad_s - Rad_r = \Delta Rad_{REFL} + \Delta Rad_{CF} + \Delta Rad_{INT} + \Delta Rad_{CSKY} \quad (5)$$

The interaction term ΔRad_{INT} and the clear sky term (ΔRad_{CSKY}) were found to be negligible and are therefore ignored for the purposes of this paper.”

Line 246: Was cloud base always at temperatures above freezing i.e. was the melting layer always within cloud? If not then omitting rain alone may not be enough to ignore falling precipitation. Please clarify this here.

Reply: Cloud base height distribution shows that cloud bases of low and mixed phase clouds occur predominately between 2.5 and 5 km. Figure 1c in the paper indicates that 0°C occurs at around 5 km. Therefore,

most cloud base heights are at temperatures above freezing, but it is possible as the reviewer notes that some falling precipitation is included in our ‘in-cloud’ values. We felt it was important to include snow and graupel in the cloud mass calculation for determining where cloud was due to their importance in the deep and dynamic convective clouds represented in the domain. The determination of what qualifies as ‘in-cloud’ is uncertain and not well defined in the literature. In order to address this, a number of cloud mass threshold for determining ‘in-cloud’ values were tested and these were not found to notably affect the sensitivity of the clouds to the INP parameterisation choice. The inclusion of rain in the in-cloud threshold determination also had a negligible effect. The discussion and results presented in general do not depend strongly on the exact classification of ‘in-cloud’ values because the radiation calculations refer to the domain-wide calculations of top-of-atmosphere outgoing radiation and precipitating areas will be below cloud and so not likely to affect this. However, we agree that the classification of what qualifies as in-cloud should be explored and its effects examined in future work.

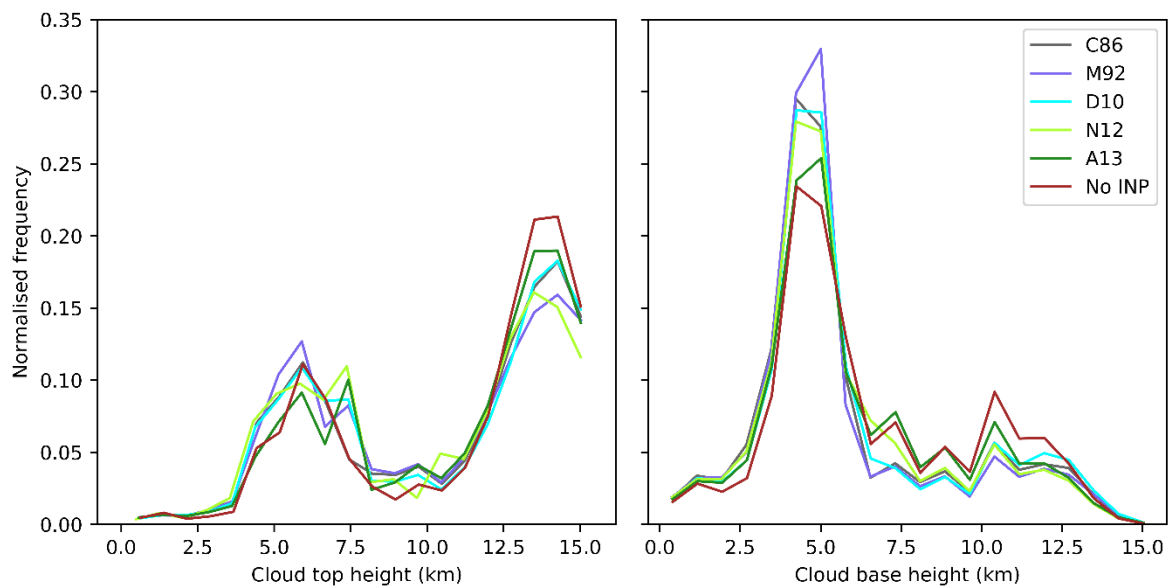


Figure 1. Histograms of cloud top and cloud base height distributions throughout the simulations.

Line 257: please remove the additional “our” before “one of our”

Change to paper: As suggested

Line 274: define DMT ie. Droplet Measurement Technologies

Change to paper: As suggested.

and please provide the specifics on the CDP and CIP e.g. size range of measurements.

Change to paper: The following changes in bold have been made to Section 2.2: “The aircraft cloud droplet number concentration (CDNC), measured using a Droplet Measurement Technique (DMT) cloud droplet probe (**which allows measurement of the cloud droplet size distribution for particles with diameters between 3 and 50 μm (Lloyd et al., 2020)**), falls predominantly in the regions of parameter space most highly populated by model data when plotted against vertical wind speed (Fig. A2b).”

“The observed ICNC was derived from measurements using the DMT Cloud Imaging Probes (CIP-15 and CIP-100, **photodetector widths of 15 and 100 μm respectively, both with 64 detector elements**)”

Line 281: should SODA2 also include a reference?

Change to paper: We are unaware of a reference for the open source SODA2 code but we have now referenced a recent monograph describing the OAP processing. The following change in bold has been made to Section 2.2: “*using the SODA2 (**System for OAP (optical array probe) Data Analysis**) processing code (McFarquhar et al., 2017)*”, and the relevant citation has been added to the reference list.

Line 290-291: Please clarify here that all ice-phase hydrometeors contribute to HMP.

Change to paper: “*Ice crystals formed via homogeneous freezing **and sedimented to lower levels can initiate ice production via the Hallett-Mossop process once converted to snow or graupel***”

Line 311-312: Same here.

Change to paper: “*Bear in mind that SIP was active (SIP_active) in the simulations summarised in Fig. 4a, including in the NoINP simulation in which the Hallett-Mossop process can be initiated by settling **ice-phase hydrometeors (either by settling homogeneously frozen ice crystals subsequently converted to snow or graupel, or by settling snow or graupel formed from homogeneously frozen ice crystals at upper cloud levels),** indicating that these cloud systems are sensitive to INP even in the presence of SIP.*”

Line 334: please add a “to” in “due a reduction”

Change to paper: Sentence has been altered to: “*When INP are included in a simulation, snow and cloud droplet water path are enhanced, causing increases in total cloud condensate, despite decreases (in all except A13) in ice crystal water path due to a reduction in ice crystal number and mass concentrations **caused by a** reduction in the availability of cloud droplets for homogeneous freezing.*”

Line 342-345: Here it is stated that the increase in snow and graupel production due to heterogeneous freezing increases sub-cloud evaporation of rain. However, when looking at Figure A5, the snow mass and graupel mass suggest that the A13 parametrization would lead to the largest amount of available melted mass to increase below cloud humidity. Yet, when looking at Figure A6, A13 has one of the lowest increases in sub-cloud humidity. How is this justified?

Reply: In Figure A4, we can see that A13 has an in-cloud concentration of graupel lower than the other 4 parameterisations. In our CASIM simulations, the density and fallspeed of graupel is higher than that of snow, and therefore graupel is likely to make the largest contributions to falling precipitation. Therefore the lower concentration of graupel in A13 relative to the other INP active simulations may explain why it has the lowest increase in sub-cloud humidity.

Change to paper: The parameters used for particle fallspeed and density are listed in Miltenberger et al. (2018) and this is now referenced in Section 2.1.2: “*The parameters used in the representation of the size distribution, density and terminal fall speed velocities of each of the five hydrometeor classes represented by CASIM are shown in Table 2 of Miltenberger et al. (2018).*”

Line 352-360: In line with my general comment about the slope of the A13 parameterization, I generally agree that the A13 will produce less ice and therefore remove fewer cloud droplets at low levels in the cloud (below 5

km). However, the number of ice crystal produced at this low-level is quite insignificant to the number of cloud droplets. This can be clearly seen in Fig. 2 where at -6 C (Approx 5 km, assuming dry atmospheric lapse rate) the shallower sloped INP parameterizations (with significantly higher INP concentrations than A13 (~3 orders of magnitude)) still only predict < 1 INP per liter while in Fig. 6c, the concentration of cloud droplets is between 3 and 5 per cm³. Thus, the low number of cloud droplet activation due to ice nucleation, and following loss due to riming and depositional growth is likely insignificant to the number of cloud droplets observed at this height and that can be advected to higher regions of the cloud. Rather, is it more likely, that the increase in relative humidity of the low-level air mass due to evaporation of rain that is increased due to enhanced ice nucleation due to the higher INP concentrations of the steep parameterizations at higher levels of the MPC responsible for the observed changes in cloud droplet number? Additionally as HMP is rather unimportant on the CDNC for the steeper sloped parameterizations (N12 and A13) could it be argued that the higher INP concentrations predicted at colder temperatures are much more important as discussed below? If yes, then again, perhaps the slope of the INP parameterization is not as important as presented, especially as A13 is flat starting at -26 C.

Reply: As noted by the reviewer above, A13 has the lowest increase in sub-cloud humidity of the parameterisations tested. Therefore, we feel it is unlikely that the higher cloud droplet number concentrations at lower mixed-phase levels in A13 is exclusively caused by the changes in the relative humidity, or that the changes in sub-cloud relative humidity are caused by the ICNC at upper mixed-phase levels (as if that were the case A13 would have the highest increase in sub-cloud humidity). While the reviewer is correct that the HMP is less important for steeper parameterisations, this relative unimportance is a result of the lower heterogeneous ice production rates in lower mixed-phase levels of the cloud and thus a direct result of the chosen INP parameterisation. Therefore, we feel it is fair to say that the CDNC at the lower mixed-phase levels is affected by parameterisation slope. However, the number concentration of INP at colder temperatures may be more important than the slope of the parameterisation for some of the results presented and we were not able to decouple these two effects in this work. This caveat is now clearly stated in our limitations section (as also stated above).

Change to paper: The following text has been added to Section 4 of the paper: *“Furthermore, while many cloud macro- and microphysical were correlated with INP parameterisation slope, the slope of the parameterisation at low temperatures for the A13 and N12 parameterisations can be flat because the parameterisations plateau once they reach the number concentration of dust represented in the model gridbox in question. This means that at high dust concentrations, the slope of the INP parameterisation correlates with the INP concentration at temperatures between -25 and -35°C (Figure 2). This means that the absolute number concentration of aerosols capable of nucleating ice is not decoupled from the INP parameterisation slope in some INP parameterisations and that some cloud responses attributed to changes in the INP parameterisation slope may have in fact been caused by the absolute INP number concentration at cold temperatures. The relative importance of the INP parameterisation slope and the absolute number concentration of aerosols capable of nucleating ice will be investigated in future work. However, whether the INP number concentration plateaus at cold temperatures is determined in part by the INP parameterisation slope, and correlations with INP parameterisation slope are evident at both warm and cold cloud altitudes indicating the importance of the INP parameterisation slope.”*

Line 367-371: Again here it is mentioned that adding evaporating precipitation acts to increase humidity. But again A13 which has the highest snow and graupel mass, has lower humidities outside of cloud. Please clarify this. Additionally, when discussing the invigoration of updraft velocity due to enhanced latent heat release due to riming, it is clear that A13, which has the largest snow mass also has the highest updrafts. However, as the updraft increase is occurring primarily in a region where A13 predicts fewer INPs than the other parameterizations, is it really the slope of the INP parameterization at these temperatures that is important, or the concentration of INPs predicted by the parameterization at colder temperatures that settle to these altitudes?

Reply: As stated above, in Figure A4, we can see that A13 has an in-cloud concentration of graupel lower than the other 4 parameterisations, likely due to the lower concentrations of INP and therefore lower ICNC at low mixed-phase altitudes and the resultant reduction in the Hallett-Mossop ice particle production rates. In our CASIM simulations, the density and fallspeed of graupel is higher than that of snow, and therefore graupel is likely to make the largest contributions to falling precipitation. Therefore the lower concentration of graupel in A13 relative to the other INP active simulations may explain why it has the lowest increase in sub-cloud humidity.

In response to the question about the updraft velocity, A13 has one of the lowest updraft velocities below ~6 km likely due to the lower graupel concentrations seen in Figure A4. The lines in question refer to the lower mixed-phase cloud fraction at ~5 km which would be affected by the lower out-of-cloud humidity and lower updrafts speeds. The changes at upper levels in the cloud may well be more sensitive to the low temperature INP concentrations rather than the INP parameterisation slope and the fact that these two parameters are not decoupled in our simulations is now stated clearly in our limitations.

Change to paper: The parameters used for particle fallspeed and density are listed in Miltenberger et al. (2018) and this is now referenced in Section 2.1.2: *“The parameters used in the representation of the size distribution, density and terminal fall speed velocities of each of the five hydrometeor classes represented by CASIM are shown in Table 2 of Miltenberger et al. (2018).”*

The following text has been added to Section 4 of the paper: *“Furthermore, while many cloud macro- and microphysical were correlated with INP parameterisation slope, the slope of the parameterisation at low temperatures for the A13 and N12 parameterisations can be flat because the parameterisations plateau once they reach the number concentration of dust represented in the model gridbox in question. This means that at high dust concentrations, the slope of the INP parameterisation correlates with the INP concentration at temperatures between -25 and -35°C (Figure 2). This means that the absolute number concentration of aerosols capable of nucleating ice is not decoupled from the INP parameterisation slope in some INP parameterisations and that some cloud responses attributed to changes in the INP parameterisation slope may have in fact been caused by the absolute INP number concentration at cold temperatures. The relative importance of the INP parameterisation slope and the absolute number concentration of aerosols capable of nucleating ice will be investigated in future work. However, whether the INP number concentration plateaus at cold temperatures is determined in part by the INP parameterisation slope, and correlations with INP parameterisation slope are evident at both warm and cold cloud altitudes indicating the importance of the INP parameterisation slope.”*

Line 374: The idea of reducing homogeneous freezing by removing condensate via heterogeneous freezing has been well studied before in several geoengineering papers. Perhaps it is worthwhile citing some of these papers here (e.g Gasparini et al., 2020 and references within).

Change to paper: The following has been added to the paragraph in question “The ability of heterogeneous freezing to reduce the availability of moisture for homogeneous freezing has been previously observed (e.g. Gasparini et al., 2020; van den Heever et al., 2006; Kärcher and U. Lohmann, 2003; Lohmann and Gasparini, 2017; Phillips et al., 2005, 2007; Storelvmo et al., 2013).”

The citations for the additional references have been added to the reference list.

Line 390: There is no discussion about how anvils are classified in section 2.1.4, just how clouds are defined as high/mid or low. Also, it seems a bit misrepresentative to constrain anvil extent to only areas where no cloud is found below. I agree that the anvil (high cloud) is linked to the mid-level and low-level cloud below. However, the anvil should absolutely be considered when it extends over low-level clouds only. When looking at the example of the cloud classifications in Fig. 8, it looks like this does not really occur. Nevertheless, it might be more important at other times.

Reply: Anvil cloud is classified as regions where cloud is present above 9 km only. It is marked as high cloud (or H) in Figure 8 because using high cloud made the classification mixed category columns (e.g. columns with cloud in mid and high areas (MH)) easier for plotting purposes. We agree that technically anvil cloud should include cloud that extends over low clouds only. However, the inclusion of this low/high cloud category in the results was tested and found to have no effect because it comprises so little of the domain cloud. We therefore classified anvil cloud as high cloud only because it greatly simplified the discussion and made the explanations more accessible with no notable effect on the results.

The previous point also raises the question as to why 20 UTC was selected as the example of the cloud classifications to the cloud field in Fig. 8, rather than 1330 or 1030 when the comparison between the satellites and the model were conducted? Especially since the 20 UTC does not fall within the daytime calculations.

Reply: 20 UTC was selected as the example of the cloud classifications to the cloud field because the changes to the anvil cloud are likely to become more important the longer the simulation is run for due to the persistence in the atmosphere of the anvil cloud beyond when the convective cloud has decayed. This is stated clearly in Section 3.4 (*"it is possible that the effect of INP and INP parameterisation choice on anvil cloud fraction, and the contribution of anvil cloud to overall cloud fraction and radiative changes, would become larger with a longer analysis period."*). 20 UTC also has a very obvious and well developed convective cloud system which clearly shows all the different cloud categories. However, quantitative differences in the cloud development are obvious across the entire simulation.

Line 394-396: Such a drastic decrease in cirrus anvils between the noINP and INP simulations must have large impacts on the domain averaged outgoing longwave radiation at nighttime. Perhaps it is worthwhile showing this even if it is not the same amount of time that is compared, as mentioned in a response to one of the reviewers. That being said, with such a reduction in cirrus cloud fraction, does the addition of INPs makes the nighttime outgoing longwave radiation significantly higher than reported?

Reply: The change in the longwave radiation over the entire simulation period excluding spin-up (10:00-24:00 UTC) is shown in Figure 1 below for high (i.e. anvil) cloud and for the entire domain. We see a large reduction in outgoing radiation from anvil cloud indicating that the anvil cloud when INP are active are occurring at warmer temperatures. This is partly compensated by the reduction in anvil cloud cover that occurs in the simulations with active INP but the net result is still a reduction in the outgoing longwave radiation over the whole domain (total in Figure 2).

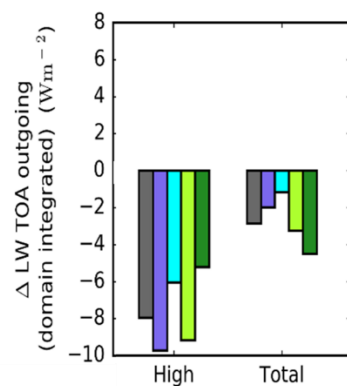


Figure 2. Change in TOA outgoing longwave (10:00-24:00 UTC) radiation for high (anvil) cloud and the entire simulation domain due to the inclusion of INP (INP-NoINP). A positive value indicates more outgoing radiation when INP are active.

Line 396-399: Again it might be worthwhile to cite previous studies here that have investigated this as well e.g references in Gasparini et al, (2020).

Change to paper: As suggested: “*The reduction in anvil extent in the presence of INP is caused by increased liquid consumption at all mixed-phase levels, due to heterogeneous freezing, enhanced SIP and increased graupel and snow production, reducing the availability of cloud droplets for homogeneous freezing (Fig. A4b), reducing ICNC at cloud-top, and reducing cloud anvil extent (Fig. 8g), in agreement with previous studies (e.g. Gasparini et al., 2020; van den Heever et al., 2006; Kärcher and U. Lohmann, 2003; Lohmann and Gasparini, 2017; Phillips et al., 2005, 2007; Storelvmo et al., 2013).*”

Line 419: The review paper by Korolev and Leisner, (2020) could be included here.

Change to paper: As suggested.

Line 439-440: What effect does the formation of the “anvil” occurring at warmer temperatures have on the outgoing longwave radiation? This may be important as outgoing radiation is related to the temperature to the 4th power.

Reply: As noted in the manuscript results, the outgoing longwave radiation in C86, M92 and D10 is enhanced by the inclusion of the Hallett-Mossop process and decreased in N12 and A13. The lines in question relate to the change in hydrometeor size in the anvil due to changes in the altitude of complete cloud glaciation (which occurs at lower altitudes for N12 and A13). Therefore the lines in question do not directly relate to the anvil forming at warmer temperatures. We did not look into this issue in detail because the changes in shortwave radiation were so much larger than those of longwave radiation, and so changes in cloud reflectivity and cloud fraction was where we directed our focus. The changes in outgoing longwave radiation due to INP parameterisation slope should be examined in more detail in future work.

Figure 2: It is mentioned in the caption that the INP parametrizations shown are for an aerosol concentration of 8 cm^{-3} . As three of these parameterizations are surface area dependent or at least size dependent (D10 e.g. aerosols $> 500 \text{ nm}$) what sized particles are assumed here?

Reply: In Figure 2, the assumed mean particle radius is $0.7 \mu\text{m}$, which is calculated from the number and mass concentrations of the assumed insoluble dust profile shown in Figure 1. In the model, the surface area of the particles is calculated using the available dust particles and the particle size distribution of CASIM. For the D10 parameterisation, all particles are assumed to be over $0.5 \mu\text{m}$ since dust particles are relatively large and the INP number concentration is calculated using the coarse dust size mode in CASIM.

Change to paper: The following sentence has been added to the caption of Figure 2: “*N12 and A13 are calculated assuming a mean dust particle radius of $0.7 \mu\text{m}$. In D10, all particles are assumed to be larger than $0.5 \mu\text{m}$.*”

Also, a concentration of 8 cm^{-3} seems to be an unfair comparison to the observations from the Welts et al, (2018) study as the ambient surface aerosol concentration were likely significantly higher when these INP measurements were conducted. Indeed the modelled and measured surface aerosol are approximately 2 orders of magnitude higher than at the 4 km level. This would put the parameterizations, with the exception of A13, significantly higher than the observations by Welts et al (2018). Or are you relying on the values of the modeled insoluble aerosol concentration to justify the comparison with the Welts et al, (2018) data? This should be expanded upon or at least mentioned. Additionally, if the values are normalized to surface area then this should also be mentioned.

Reply: Comparison is made between the modelled INP and the Welts et al. (2018) INP concentrations because the Welts dataset is from the Cape Verde region where our simulations are based. The modelled insoluble dust profile shown in Figure 1b, which is based on a Met Office Unified Model run with the CLASSIC dust scheme, indicates that much of the aerosol measured at the surface is likely not dust. However we have clarified in the figure legend that the Welts data is measured at the surface and the Price data is measured from aircraft.

Change to paper: The following sentence has been added to the caption of Figure 2: *“Note that the Welts et al. (2018). Note that the Welts et al. (2018) dataset is from surface INP measurements at Cape Verde while the Price et al. (2018) dataset is measured from an aircraft flown from Cape Verde.”*

Furthermore, how was the data from Price et al, (2018) presented? Was it scaled to the surface area of the modelled insoluble aerosol or are these just the absolute concentrations per liter of air reported from the airborne samples?

Reply: Shown is the absolute concentrations per liter of air reported from the airborne samples. However, the calculation of the INP number concentrations was tested using a range of aerosol concentrations (additional to that shown in Figure 2) from the modelled profile and in all cases the parameterisations agree relatively well with the Price et al (2018) data.

Also please fix the Welts et al, (2017) to (2018) in the figure legend.

Change to paper: As suggested.

Lastly, I am not sure it makes sense to include the information that D10 was linearized for the correlation analysis in the figure caption here. Perhaps this is better suited in the text or in the first slope correlation plot.

Change to paper: As suggested, the statement regarding the linearization of D10 has been moved to Section 2.1.4.

References:

Gasparini, B., McGraw, Z., Storelvmo, T. and Lohmann, U.: To what extent can cirrus cloud seeding counteract global warming?, Environ. Res. Lett., 15(5), 054002, <https://doi.org/10.1088/1748-9326/ab71a3>, 2020.

Korolev, A. and Leisner, T.: Review of experimental studies on secondary ice production, Atmospheric Chem. Phys., 1–42, <https://doi.org/10.5194/acp-2020-537>, 2020.