

# ***Interactive comment on “Modelling of cirrus clouds – Part 2: Competition of different nucleation mechanisms” by P. Spichtinger and K. M. Gierens***

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## **1 Reply to reviewer 1**

### **1.1 Ad General comments:**

Ad heterogeneous nucleation parameterisation:

The main concern of the reviewer is the very simple treatment of heterogeneous nucleation as a threshold phenomenon. First, we want to stress that we do not claim in any way that nature behaves like that. But there are indications that nature sometimes is not far from this kind of behaviour, see below. Anyway, for a process study like ours

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it is an advantage to have as little free parameters as possible. The simple threshold is one parameter, the concentration of IN is the other. Every more advanced threshold formulation would require more free parameters and would render interpretation of the results more complicated, perhaps even non-unique. Even if our approach here may look a bit academic we are willing to pay this price because we think we can learn the underlying mechanisms more clearly.

As heterogeneous nucleation is still a “closed book” in many respects, we also think that simple formulations are warranted. Nevertheless, in principle, we could use much more sophisticated formulations, for instance including dependencies of heterogeneous nucleation on aerosol size, temperature and other environmental conditions (e.g. Möhler et al., 2006; Spichtinger and Cziczo, 2008), but with increasing complexity of parameterisations the main impact of heterogeneous IN on cloud evolution could get entangled with the effects of the parameterisations.

Furthermore, it is not at all clear whether a complex formulation describes nature more accurately than our simple one. Laboratory experiments are inconclusive in this respect, and some lab and field measurements seem to be more consistent with a “threshold phenomenon” (D. Cziczo, pers. comm.). Thus, our simple parameterisation may be not so academic as it appears.

In a sensitivity study the impact of a parameterisation like that of Möhler et al., 2006 (i.e. increase in activation of the heterogeneous IN until all aerosols are activated) was tested. In a qualitative sense, this did not lead to results different from those shown in our paper. Hence we are quite confident that our simulations convey a lore that will stay robust when we employ more advanced parameterisations of heterogeneous nucleation.

The impact of fluctuations on more sophisticated nucleation parameterisations is considered below in 4. The limiting factor for the possible effects is the very limited number of available IN.

Ad figures: Some unreadable figures have been improved.

## 1.2 Ad Specific comments:

### Introduction:

1. We add “Koop (2004)” as an appropriate reference for homogeneous freezing. As a matter of fact, homogeneous freezing below the spontaneous freezing limit of pure water (about  $-38^{\circ}\text{C}$ ) is freezing of aqueous solution droplets and needs substantial supersaturation in excess of 45% in order to rarefy the solute molecules sufficiently that formation of an ice crystal lattice is possible. As Koop et al. (2000) have shown, onset of freezing occurs for many solutions at a *certain* temperature dependent water activity (which is relative humidity when updraught is slow). Hence, words like “on average” do not appropriately describe the process.
2. DeMott et al. (2003) describe measurements indicating generally low IN concentrations in the order of tens per liter. Moreover we are very cautious with our wording, saying “generally believed” and “while this may be so”. Models for heterogeneous nucleation sometimes use the old Fletcher curve (with *ad hoc* corrections) to predict the number concentration of IN. These paramterisations often predict high numbers of IN. However, the Fletcher curve was derived from measurements down to  $-30^{\circ}\text{C}$ , which is not representative for the upper troposphere, and we do not use it therefore.
3. Boxmodel and parcel model are synonyma to our understanding.
4. (See also above under general comments).  
It is true, we can say we did not investigate how a continuous heterogeneous nucleation process would affect the cloud evolution. However, we stress that this was done on purpose. As we say in section 2:

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...details are unimportant, since our question is: what happens to homogeneous nucleation and the cloud evolution when a number of ice crystals is already formed by *any* mechanism at a lower relative humidity threshold. For this purpose we can apply a very simple parameterisation ...

In principle we can prescribe any form of heterogeneous nucleation behaviour. But any other than a simple threshold introduces more degrees of freedom which renders the interpretation of the results more complicated. Therefore, for the present paper we do not intend to investigate more complex heterogeneous nucleation behaviour. We clarify this further in the revised version.

### Setup:

1. We add an explanation for the abbreviation ISSR = Ice SuperSaturated Region and a reference (Gierens et al., 1999).

### Discussion of results:

1. The difference between 5a and 5b is obvious, we hope. Fig 4b is intermediate between the cases of fig 5 (bigger figures in the final version will help).
2. We agree, but see above. Hence this should be the topic of another paper, perhaps Part 3.
3. We agree that the explanation is too simple. While the nucleation rate decreases indeed with increasing temperature (see our Fig. 8), this is only one part of the explanation. Less crystals mean less competition for excess vapour and additionally at higher temperature there is higher absolute humidity. Hence crystals grow quicker and fall faster through the supersaturated layers. Hence they don't have much of an effect there. We add a few sentences to complete the explanation.

4. Heterogeneous nucleation is always limited by the given number concentrations of ice nuclei. According to measurements (DeMott et al., 2003) the usual number concentrations in upper troposphere are quite low (in order of tens per liter). That is, regardless of how the heterogeneous freezing mechanism works, fluctuations cannot produce more ice crystals than the available IN allow. The question is then whether fluctuations in combination with a continuous  $RH_i$  dependence of heterogeneous freezing could produce *less* crystals. In order to achieve that it would be necessary that enough crystals that are produced first get mixed into their neighbourhood and start to grow such quickly that they could inhibit the further increase of supersaturation there. Since heterogeneous nucleation does only produce small numbers of ice crystals they are simply not numerous enough to produce such an effect. Moreover, if such a quenching effect would arise from fluctuations, it would be strongest in the case considered here (i.e. with fixed nucleation threshold) since then the first nucleation event produces the full number of ice crystals (equal to the number of IN) that thereafter can spread etc. With a continuous freezing model the first nucleation event does not produce the full number of ice crystals, hence the effect would be even weaker.
5. Relaxation means the approach of equilibrium, i.e. ice saturation. It might be better if we write "Apart from crystal growth".
6. We are referring to exotic mechanisms in microphysics with evidence from lab measurements:
- cubic ice formation at low temperatures  $T < 200$  K (Murray et al., 2005; Murray, 2008a),
  - suppressing homogeneous nucleation via organic compounds (Kärcher and Koop, 2005),
  - glassy states of aqueous solutions that inhibit freezing (Zobrist et al., 2008; Murray et al., 2008b),

The remaining part of the question again supposes that there are much higher IN concentrations in the UT than assumed in our simulations. Again we state, that measurements by DeMott give number concentrations of the order ten per liter. Simulations sometimes use the old Fletcher curve or derivatives from it (to prevent much too high IN concentrations in the UT), or the parameterisation of Meyers et al. (1992), which parameterises the number of IN as a function of  $RH_i$ . While the original Fletcher curve was derived from data in the lower part of the troposphere (see above), the derivatives seem not to be based on measurements in the UT as well. Meyers parameterisation is valid in a temperature range  $T > -20^\circ\text{C}$ . Hence these parameterisations do not apply to cirrus clouds in the UT.

### 1.3 Ad Technical corrections:

1. “boxmodel” → “box model” done.
2. Page (9063) line (20): done.
3. Page (9064) line (15): “dual” was meant in its mathematical sense. We could say “is assigned to”.
4. Page (9071) line (15-19): “Increase rate” is indeed a bit ugly. What we mean is the rate (change per time unit) of increase of supersaturation ( $dSi/dt$ ). This is very long. So we now write this long piece of text once and then always use  $dSi/dt$ .
5. Page (9071) line (26): done
6. Page (9073) line (3): done
7. Page (9075) line (15-19): see above ( $dSi/dt$ )
8. Page (9076) line (10-11): see above ( $dSi/dt$ )

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9. Page (9076) line (4): done (“surface area concentration”, which is area per unit volume)
10. Page (9080) line (15-17): We change the sentence to “This impeding effect ... is strong in particular when  $RH_{i_{het}}$  is little below  $RH_{i_{hom}}$ .” The suggested word “even” would turn the sense of the statement into its contrary.
11. Figure 6, figure caption: done
12. Figure 8, figure caption: done
13. Figure 12, figure caption: corrected.

## 2 Reply to reviewer 2

### 2.1 Ad General comments:

The word “dominant” in our paper refers to the ice production process, i.e. heterogeneous or homogeneous nucleation. It does not refer to any of the quantities. Therefore it is not possible to define the regime in terms of measurable quantities. We admit that text pieces like “homogeneously dominated cloud” are a bit misleading. We have reformulated such pieces and say now, for instance, “clouds predominantly formed by homogeneous freezing”.

The critical number concentration of IN is a strong function of several parameters and therefore expression like “low”, “medium”, “high” would indeed be inappropriate if they would be used in a general way. However, in the paper we use these expressions only in the context of the respective situation, i.e. “low” means less IN than the critical concentration and so on.

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The number concentration of liquid aerosol particles is  $300 \text{ cm}^{-3}$  (given in par 2 of the setup section). Hence it needs a lot of nucleation events to deplete this reservoir. The impression of a constant amount of  $n_c = 50 \text{ L}^{-1}$  comes from plotting the data - the isolines are labelled in  $50 \text{ L}^{-1}$  intervals.

Kajikawa and Heymsfield (1989) measured the number density of aggregates in ice clouds warmer than  $-44.8 \text{ }^\circ\text{C}$ , which is warmer than our case. Less than one percent of the crystals they could detect were aggregates in the coldest cloud layer they considered. They could not detect smaller crystals than  $75 \text{ }\mu\text{m}$ , so that small fraction of aggregates is rather over- than underestimated. They determined a collection efficiency of about 0.03. Under colder conditions it is probable that the collection efficiency is even smaller (although we do not know any measurements of this).

For gravitational collection we derive the following formula for the timescale using relations taken from Pruppacher and Klett (1997), assuming a collection efficiency of about 0.03:

$$\left(\frac{\tau_{grav}}{s}\right) \approx 2 \times 10^9 \left[\frac{(r_1 + r_2)}{\mu\text{m}}\right]^{-2} \left(\frac{\Delta v}{\text{cm/s}}\right)^{-1} \left(\frac{N_i}{\text{cm}^{-3}}\right)^{-1}$$

The formula shows that small sedimentation time scales can be achieved when large crystals are present together with high number concentrations. Generally, we find large crystals in the lower part of the cloud while high number concentrations prevail in the upper part of the cloud. This all makes it very improbable that aggregation would play an important role in clouds like those we simulated.

Let us illustrate this with some numbers. We calculate aggregation timescales using two columnar ice crystals of lengths  $L_1 = 200 \text{ }\mu\text{m}$  and  $L_2 = 400 \text{ }\mu\text{m}$  as typical for the lower part of the cloud assuming  $r_i = L_i/2$ . Terminal velocities are as derived by Heymsfield and Iaquinta (2000) (described in Spichtinger and Gierens, 2008). The number concentration of such large crystals is of the order  $0.001 - 0.01 \text{ cm}^{-3}$ . This gives  $\tau_{grav}$  between  $2 \times 10^4$  and  $2 \times 10^5 \text{ s}$ . In the upper part of the cloud we take a

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maximum concentration of  $n_c = 0.2 \text{ cm}^{-3}$ , crystal lengths of 50 and 10  $\mu\text{m}$ , and the corresponding  $\Delta v$  from Heymsfield and Iaquinta (2000). This again leads to  $\tau_{grav}$  of the order  $10^5 \text{ s}$ . For comparison, a fall time for a  $L = 300 \mu\text{m}$  crystal through the 1 km deep ISSR is 2000 s.

We added a subsection on size distributions, i.e. sec. 4.7.

## 2.2 Ad Specific comments:

- Page (9063) line (9): This is true. They specify critical IN concentrations as functions of vertical velocity (and other parameters). Corrected.
- Page (9065) line (6): There is no intended physical implication with this choice. It is merely the simplest possible assumption. It needs only 2 degrees of freedom (the threshold value and the number of IN), and the results of process studies can therefore be interpreted relatively easily and unambiguously. Every other choice needs more degrees of freedom, and would render the interpretation more complex and perhaps ambiguous. Has been clarified further. See also reply to referee 1, ad general comments.
- Page (9065) line (5): corrected.
- Page (9065) line (10): yes. It is 0.5 for both crystal classes. Specification added.
- Page (9066) line (15): Strictly speaking, there is no explicit size distribution for this aerosol type in our simulation since the parameterisation does not need it. We will slightly reformulate the sentence (“not specified” instead of “irrelevant”).
- Page (9067) line (8): Done.
- Page (9068) line (24): It is well known that the homogeneous nucleation rate is a very steep function of supersaturation. Usually, a nucleation threshold is defined

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when the nucleation rate exceeds one per cubic centimeter and second. It is not necessary to explicitly give a formulation of such a threshold, but one can find one in Kärcher and Lohmann (2002), Gierens (2003), Ren and Mackenzie (2005), e.g.

$$S_{cr} = 2.583 - T/207.83 \text{ K. (Kärcher and Lohmann, 2002)}$$

The word overshooting includes the whole  $S_i(t)$  during the time where the humidity is above the threshold, but we agree that the reduction of the peak supersaturation is certainly the main effect (due to the above mentioned steepness of the nucleation rate).

- Page (9069) line (10): corrected
- Page (9070) line (5): corrected
- Figure 4: We prefer to leave the panel c in the figure. Even if it is not explicitly discussed in the paper, it helps the interested reader to see how the cloud properties change when the number of IN is increased.
- Figure 5: This is simply a plotting problem: our resolution of the plot is 10 min in the x-axis, but 10m on the y-axis - the kink in the isolines is due to the fact that in the upper layers already at time  $t_0$  heterogeneous nucleation took place, while in the lower parts the heterogeneous threshold is surpassed only 10 min later.
- Page (9071) line (20): The relative difference between 200 and 250 is 25% (taking the lower value as the base), while the relative difference between 100 and 180 is 80%. Hence, in a relative sense the impact of the heterogeneous nucleation threshold is larger in the middle of the cloud than at cloud top.
- Page (9072) line (6): corrected.

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- Page (9072) line (9): We add in the text of the figure caption the specification of the threshold. The implication of this is: in equilibrium with ambient water vapour an aerosol particle with radius  $0.25 \mu\text{m}$  freezes homogeneously within 1 s (Koop et al. 2000).
- Page (9073) line (6): When the uplift is strong enough, homogeneous nucleation will occur for sure, and produce much more ice crystals than heterogeneous nucleation has produced before. The stronger the uplift, the more crystals are produced homogeneously while the number of heterogeneously formed crystals does not depend on uplift strength. Hence the further evolution of the cloud *in all respects* is less and less affected by the presence of a smaller and smaller portion of heterogeneously formed crystals with increasing uplift strength. Sentence will be reformulated.
- Page (9073) line (23): corrected.
- Page (9075) line (25): We replace “most polluted situation” with “situation with the highest concentration of IN”.
- Figure 8: This figure presents crystals formed homogeneously with no IN in the background. Caption will be amended.
- Page (9076) line (15): Yes.
- (disagree with statement): See reply to referee 1, discussion of results, point 4.
- Page (9076) line (25): We mean variability in units of RH<sub>i</sub>. Will be clarified.
- Page (9077) line (21): When we replace “fluctuations” with “variations” or “variability” in this sentence, it should be clearer.
- Page (9078) line (13): corrected

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- Page (9078) line (23): see reply to referee 1, discussion of results, point 6.
- Page (9080) line (9): Agreed.
- Page (9081) line (23): corrected.

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