Review of manuscript acp-2012-97: “Parameterization of homogeneous ice nucleation for cloud and climate models based on classical nucleation theory”

(submitted for publication in Atmospheric Chemistry and Physics)

by V. I. Khvorostyanov and J. A. Curry

(Anonymous referee)

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1 Overall evaluation

I recommend the editor to accept the manuscript for publication in ACP after – in relation to the quantity and quality of the paper – minor revision.

2 Assessment according to criteria for ACP review and interactive discussion

1. Does the paper address relevant scientific questions within the scope of ACP?
   Yes.

2. Does the paper present novel concepts, ideas, tools, or data?
   Yes.

3. Are substantial conclusions reached?
   Yes.

4. Are the scientific methods and assumptions valid and clearly outlined?
   In general yes, but some points should be more clearly outlined (see specific comments).

5. Are the results sufficient to support the interpretations and conclusions?
   Yes.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?
   In general yes, but I recommend to check some specific parts of the calculus (see specific comments).

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?
   Yes. However, owing to the very expansive calculus I recommend the authors to place references to their own previous works more specifically to the calculus (see specific comments).

8. Does the title clearly reflect the contents of the paper?
   Yes.

9. Does the abstract provide a concise and complete summary?
   Yes.

10. Is the overall presentation well structured and clear?
    Yes.

11. Is the language fluent and precise?
    Yes.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?
    In general yes, some specific comments for revision are given.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?
    No.

14. Are the number and quality of references appropriate?
    Yes.

15. Is the amount and quality of supplementary material appropriate?
    No supplement included.

3 Rationale

The paper contributes to an important subject of atmospheric chemistry and physics, namely the investigation of the atmospheric freezing processes. It is both of high scientifically relevance and of excellent scientific quality! The extensive use of calculus demonstrates the strong theoretical background of the authors. The work presented here satisfies ACP standards for publication.

I would like to emphasise the following results of the study:

- The authors provided a comprehensive theoretical and modelling approach to consider non-steady-state effects in homogeneous freezing (which could, on principle, also be applied to heterogeneous nucleation). The new approach is based on analytical solutions of integro-differential equations governing the time evolution of water vapour supersaturation and ice crystal concentration, i. e., it surrenders the use of the "saturation adjustment approximation", commonly employed in cloud microphysics. Owing to the compact form of the obtained final expressions, the new scheme is very interesting for application in both cloud and climate models.

- The authors demonstrated the importance of water vapour supersaturation for "homeostasis" in atmospheric freezing: ice crystal formation is self-limited by the depletion of vapour supersaturation due to vapour deposition onto ice particles (e. g., time evolution of nucleation rate and polydisperse nucleation rate in Figs. 1d and 1e, p. 6759 ff.). This supersaturation-controlled homeostasis of the ice formation process cannot be obtained by neglecting the dependence of the nucleation rate on water vapour supersaturation and considering temperature dependence only, as assumed in classical nucleation theory (CNT).

- The implications for the radiative properties of cirrus clouds might be very important for climate modelling: the authors showed that the uncondensed water vapour excess is still greater or comparable to the instantaneous ice water content, and that during the first 30 min only less than 50% of the "physically reachable" ice water content has been formed (see also p. 6760, lines 11-15, and Fig. 2f). Therefrom the authors concluded "that optical thickness and emissivity of cirrus clouds at the initial stages of their formation are significantly smaller than predicted in a bulk model" (which is based on "saturation adjustment" and on the assumption of instantaneous condensation of the available water vapour into cloud ice).

- Interesting is also the large sensitivity of the time evolution of different microphysical properties against the vertical velocity of the cloud parcel (comparative simulations have been performed for $w = 4\,\mathrm{cm\,s^{-1}}$ and $20\,\mathrm{cm\,s^{-1}}$). This emphasises the crucial role of an appropriate description/determination of vertical velocities for ice crystallisation.

- In contrast to this, the simulations revealed a remarkable insensitivity against the initial concentration of deliquescent freezing aerosol (haze) (p. 6761, lines 3-6). The authors concluded that there is a kind of "saturation" with respect to the number concentration of deliquescent haze particles, whereat the "saturation" concentration is smaller than the concentrations typically occurring in the upper troposphere.

- The tiny ratio of frozen haze particles to initially (unfrozen) haze particles (in the order of 10^{-4}) is explained by the following processes: (a) the above mentioned self-limitation of the ice freezing by water vapour depletion; (b) much faster crystal growth at given high ice supersaturation than drop growth at small water supersaturation, given at the same time.

- The authors employed an expression for the homogeneous liquid-to-solid nucleation rate, which was obtained from a generalisation of CNT. One interesting feature of the generalised rate expression is the dependence on both supercooling (or temperature T) and water vapour supersaturation (s_w). Considering typical temperature and saturation conditions in the upper atmosphere (ice crystallisation in supercooled haze (solution) particles), the authors obtained (by linearisation) the following inequality: $J_\mathrm{hom}(T, s_\mathrm{w}) \ll J_\mathrm{hom,CNT}(T)$ (see pp. 6767–6768, Eq. (41), Fig. 8). I found this result being interesting and non-trivial.

- The authors demonstrated that $J_\mathrm{f,hom}(T, s_\mathrm{w})$ variations are primarily controlled by variations in s_w, while changes due to the temperature are several orders smaller (p. 6768, lines 15-17). This result is also non-trivial and shows the strong impact of water vapour supersaturation in ice nucleation. This casts also new light of the possible role of turbulence-induced variations of supersaturation in ice nucleation.

- Based on their generalised CNT approach the authors derived a temporal dependence of the homogeneous nucleation rate (see Eqs. (48) and (49)), which was previously hypothesised by Ford (1998), Kärcher and Lohmann (2002a,b), Ren and MacKenzie (2005), and Barahona and Nenes (2008) (see p. 6771, lines 1-13). The authors argued that their generalised approach "allows to express them (i.e., the previous parameterisations) via the fundamental thermodynamic parameters reducing the number of hypothesised relations and quantities." This result can be considered as an *a posteriori* confirmation (a) of previous attempts on ice nucleation, (b) of the reconcilability of previous parameterisations with generalised CNT, and (c) of the predictive power of nucleation theory for atmospheric applications[1].

- The authors derived a useful parameterisation for the time-dependence of the number concentration of ice crystals (Eq. (52)), performed time and length scale analyses, and obtained separate expressions for the linear-growth regime (Eq. (53)) and exponential-growth regime (Eq. (54)) of ice crystals. The latter (Eq.(54)) was presented in a functional form, whose *structure* is practically identical with the parameterisation of the ice crystal concentration proposed by Meyers et al. (1992) for heterogeneous freezing. The generalised CNT gives the empirical parameters a distinct physical meaning, i.e., hitherto empirical parameters can be expressed as physical functions of well defined atmospheric nucleation parameters and observables.

- The authors gave a physically sound theoretical explanation for the greater ice crystal concentrations in cirrus in more polluted Northern Hemisphere than in the cleaner Southern Hemisphere, which "could be caused not only by the heterogeneous ice nucleation mode, but also by a small deposition coefficient in homogeneous nucleation in polluted areas" (see p. 6780, lines 12-22, and Eqs. (93), (94)).

- For the limiting cases of diffusion and kinetic growth, the authors derived the following dependencies for the (maximum) ice crystal concentration N_c

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[1] At this, the role of theoretical predictions like the present one is remindful of the following plea by J. Willard Gibbs: "*It is the office of theoretical investigation to give the form in which results of experiments may be expressed.*"

on vertical velocity w:

$$N_\mathrm{c} \propto \begin{cases} w^{3/2} & \text{Case 1 : diffusion growth limit (Eq.(88))} \\ w^2 & \text{Case 2 : kinetic growth regime : small particle limit (Eq.(93))} \\ w & \text{Case 3 : kinetic growth regime : large particle limit (Eq.(96))} \end{cases}$$

Case 2 was found to be in agreement with Ren and McKenzie (2005), case 3 with Kärcher/Lohmann (2002a,b) and Ren/McKenzie (2004) (see p. 6780, from line 11; p. 6781, lines 9-11.) The parameterisation proposed here delivered results lying within the spread of several previous parcel modelling studies from the literature (being closer to the lower limit and to the parcel simulations by Jensen employing spectral microphysics and explicit supersaturation). The new scheme was found to be in qualitative agreement with Sassen/Benson (2000) and being close to the parameterisation proposed by Kärcher/Lohmann (2002a,b), although it was based on a substantially different approach. I agree with author's conclusions that "this indicates the validity of the new parameterisation based on an extension of the classical nucleation theory and that semi-empirical approaches lead to results that can be derived from the extended classical nucleation theory" (p. 6782, lines 1-9).

4 Style of the paper

The paper is written in a condensed style and represents a systematic advancement of author's previous approach to atmospheric ice formation. In order to assess the quality of the paper, especially the extensive calculus, I have rederived ALL equations (except for Eqs. (12), (13)) therein. This includes also the Khvorostyanov-Curry (KC) theory on heterogeneous freezing of deliquescent mixed CCN and fuerther very meaningful concepts (such as "polydisperse" nucleation rate), which were proposed in Curry and Khvorostyanov (2012), Khvorostyanov and Sassen (1998), Khvorostyanov and Curry (2000), Khvorostyanov and Curry (2004b), Khvorostyanov and Curry (2004a), Khvorostyanov and Curry (2005), Khvorostyanov and Curry (2009), Khvorostyanov and Sassen (1998)[2]. This was

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[2] In order to exclude erroneous expressions (within the framework of the employed theory) I have rederived all parts of the theory from fundamental laws of thermodynamics (entropy equation), Fokker-Planck equation, and transition state theory. The only exception is growth rate according to Eq. (12) in the present paper. I have not yet had the time to check this special formulation, but I trust this expression.

a very exhausting endeavour, but most importantly, I admit to be very impressed about the amazing and nutritive theory behind the present paper.

However, an extensive calculus regularly raises questions, which might be considered by the authors as "matter of course", but which must be answered. Furthermore, in such comprehensive theoretical framework errors and typos are always virulent. Fortunately, **I did not found (logical) errors in the theory (inclusive the underlying physical assumptions)**. Unfortunately, there are a few typos in the cited papers and the present one, which must be removed[3]. In my specific comments I will give some hints, which can hopefully help to make some points more clearly in the final version of the manuscript.

5 Specific (inclusive technical) comments

The present list is an inspection protocol and does not deserve an item-by-item reply by the authors, except for those items, which will be considered by the authors as being wrong!

1. P. 6747, Eq. (1): Write $J_{\mathrm{hom},0} \propto 10^{-X(T)}$ or add units; may be it is better to use ϑ for temperature in degrees Celsius.
2. P. 6747, line 23 (typo/grammar): "... are the depressions of the melting ..."
3. P. 6752, Eq. (7): L_e, L_m, and $L_\mathrm{s} = L_\mathrm{e} + L_\mathrm{m}$ are the specific latent heats of evaporation/condensation, melting/freezing, sublimation/deposition in units of $\mathrm{J\,kg^{-1}}$; I_con, I_dep, and I_fr are the rates of condensation, deposition, and freezing in units of $\mathrm{kg\,m^{-3}\,s^{-1}}$.
4. P. 6753, line 2: The variable ρ_v is the environmental water vapour mass density (mass per volume air), and ρ_ws and ρ_is are the corresponding saturated over water and ice mass densities in units of $\mathrm{kg\,m^{-3}}$ (but not vapour pressure!).
5. P. 6753, Eqs. (8a), (8b), (9), (10): Consider to add reference to Khvorostyanov and Curry (2005, Section 2a therein). Obviously, in the present paper only water vapour deposition onto ice crystals is considered (water vapour condensation onto droplets seems to be not part of the scenario).

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[3] "Typos" occur only locally (not propagating), "errors" non-locally (propagating).

- Later, on page 6757, lines 12-14, the authors wrote: “We consider in this section homogeneous ice nucleation at cold temperatures and not very vigorous updrafts when haze solution particles freeze at water subsaturation, so that drops do NOT form.” Please move this part ahead.
- On page 6758, line 16-18, the authors wrote: “Here, we consider only the homogeneous freezing of deliquescent haze particles.” Hence, there are no water drops present serving as a condensation sink for water vapour (i.e., also no release of latent heat of condensation). The depletion of water vapour is caused by deposition onto freezing haze particles only (accompanied by release of latent heat of deposition). Hence, make the scenario more clearly at the beginning.

The derivations of Eqs. (8a), (8b), (9), (10) are correct, but add here full expressions for Γ_{12} and Γ_2, respectively. Please check in this context also Khvorostyanov and Curry (2005, Eqs. (2.3), (2.5), (2.6) therein), which must be correctly written as:

$$\frac{\mathrm{d}\rho_\mathrm{v}}{\mathrm{d}t} = -(I_\mathrm{dep} + I_\mathrm{con}) + \frac{\rho_\mathrm{v}}{\rho}\frac{\mathrm{d}p}{\mathrm{d}t} - \frac{\rho_\mathrm{v}}{T}\frac{\mathrm{d}T}{\mathrm{d}t} \ ,$$

$$\varrho c_\mathrm{p}\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{\mathrm{d}p}{\mathrm{d}t} + L_\mathrm{e} I_\mathrm{cond} + L_\mathrm{s} I_\mathrm{dep} \ ,$$

$$\frac{1}{1+s_\mathrm{w}}\frac{\mathrm{d}s_\mathrm{w}}{\mathrm{d}t} = \frac{1}{p}\frac{\mathrm{d}p}{\mathrm{d}t}\left(1 - \frac{L_\mathrm{e}}{c_\mathrm{p}T}\frac{M_\mathrm{w}}{M_\mathrm{air}}\right) - \frac{\Gamma_1 I_\mathrm{cond}}{\varrho_\mathrm{v}} - \frac{\Gamma_{12} I_\mathrm{dep}}{\varrho_\mathrm{v}} \ .$$

This form corresponds to the correct equations (8a), (8b), (9), (10).

6. Derivation of Eq. (11) is correct.

7. P. 6754, Eqs. (12), (13): Please add here references to Sedunov (1974), Young (1993), Lin et al. (2002) as was done in Khvorostyanov and Curry (2005, Eqs. (2.9a), (2.9b) therein). I have not rederived Eqs. (12), (13), but dimensional analysis provides correct physical dimensions. Note, that the deposition coefficient, α_d in the kinetic correction ξ_dep is a dimensional property (in contrast to α_d in Khvorostyanov and Curry (2005, Eqs. (2.9a), (2.9b) therein) where it has a dimension or is just a typo).

8. Derivations of Eqs. (14)-(16) are correct. Please add that the mass densities of ice, ρ_i and ρ_is, are assumed to be time independent during integration of Eq. (12), i. e., $c_{3\mathrm{i}} = \mathrm{const.}$

9. Derivations of Eqs. (17), (18a) are formally correct. However, the source term on the right-hand side of Smoluchowski equation (17) deserves a better motivation. I would expect the source term Q_f being a function of both water vapour supersaturation w.r.t. water, s_w, and temperature, T:

$$\frac{\partial f_\mathrm{c}(r_\mathrm{c}(t))}{\partial t} + \frac{\partial}{\partial r}\left(\dot{r_\mathrm{c}} f_\mathrm{c}\right) = Q_f \delta(r_\mathrm{c} - r_\mathrm{c}(t_0)) \;,$$

$$Q_f(s_\mathrm{w}, T) = \frac{\Phi(s_\mathrm{w}, T)}{\mathrm{d}t} = \underbrace{\frac{\partial \Phi}{\partial s_\mathrm{w}}}_{= \Phi_s(s_\mathrm{w}, T)} \frac{\mathrm{d}s_\mathrm{w}}{\mathrm{d}t} + \underbrace{\frac{\partial \Phi}{\partial T}}_{= \Phi_T(s_\mathrm{w}, T)} \frac{\mathrm{d}T}{\mathrm{d}t} \;.$$

In author's approach Φ_s is a function of supersaturation only, and Φ_T a function of temperature only. Is this an *ad hoc* assumption? What is the physical argument underlying the (non-trivial) decomposition of Q_f in two *independent* activity spectra? If this is clear than also the "middle part" of the conservation law for the nucleated crystals, Eq. (18a), is clear to me (I saw, however, that Φ_s and Φ_T are auxiliary variables, which were not explicitly employed later). Later in the paper, the authors performed a tricky separation of functional dependencies.

10. The notion of the "polydisperse" homogeneous freezing nucleation rate, $R_\mathrm{f,hom}$, is clear to me. It appears that this property is again a function of both supersaturation and temperature. However, dependence of nucleation rate on supersaturation (and temperature) is a special feature of author's theory, but is not included in ice crystallisation rate expressions obtained from classical nucleation theory (CNT). As the $R_\mathrm{f,hom}(T, s_\mathrm{w})$ dependence is also non-trivial, make this clear here.

11. Derivation of Eq. (18b) is correct. Please introduce $J_\mathrm{f,hom}$ already here as the homogeneous nucleation rate in units of $\mathrm{m}^{-3}\mathrm{s}^{-1}$.

12. Derivations of Eqs. (18c), (18d) are correct. Add subscript in Eq. (18d):

$$R_\mathrm{f,hom}(t_0) = \frac{\mathrm{d}N_\mathrm{c,hom}}{\mathrm{d}t} = \ldots$$

13. Page 6756, line 5: Add: "... extension of the classical nucleation theory (CNT) as employed below."

14. Derivations of Eqs. (19), (20) are correct. Introduce the effective radius in Eq. (20) as:

$$r_{\mathrm{c,ef}}(t,t_0) = \frac{r_{\mathrm{c}}^2(t,t_0)}{r_{\mathrm{c}}(t,t_0) + \xi_{\mathrm{dep}}} = \dots$$

15. Derivations of Eqs. (21), (22), (23) are correct. Eq. (21) is an integro-differential "equation for the *integral* ice supersaturation", $y_{\mathrm{i}}(t)$ (defined by Eq. (16)). Very nice result.

16. Derivation of Eq. (24) is correct, but expression contains a **typo**. The differential $\mathrm{d}t_0$ appears twice in Eq. (24) for the integral ice supersaturation. Delete the first $\mathrm{d}t_0$. Apart from that, Eq. (24) looks very nutritive. Interesting result!

17. Page 6757, line 14 to page 6758, line 3: This paragraph is unclear to me. We have Eq. (18), the polydisperse nucleation rate $R_{\mathrm{f,hom}}$, which is part of a differential equation for the determination of the crystal concentration N_{c}. What is the expression (25) standing for? Equation (25) does not correspond to Ψ_{fc} given in line 2 (**typo**). Write correctly:

$$\Psi_{\mathrm{fc}} = \frac{\Delta N_{\mathrm{c,fr}}}{\Delta r_{\mathrm{c}}} \cdot \frac{\Delta r_{\mathrm{c}}}{\Delta t}\ .$$

    Please insert here the full differential equation or reference to the governing equation. It looks like a part of author's "full microphysics model". Add sentences.

18. Figure 1a: Only relative humidity is shown here (ordinate annotation is somewhat misleading).

19. P. 6759, lines 10-23: For the interpretation of Figs. 1 and 2, the authors have already employed the notions "critical water and ice supersaturation", "critical radius", and "critical formation energy" before they were introduced/defined. Add sentence here or insert reference to later definition in the paper. Apart from that, I found interpretation physically sound. Most importantly, the simulation reveals the presence of "homeostasis" in atmospheric freezing, controlled by water vapour supersaturation: self-limitation of ice crystal formation by depletion of vapour supersaturation due to vapour deposition onto ice particles (negative feedback mechanism). This supersaturation-controlled homeostasis of the ice formation process cannot be obtained by neglecting the dependence of the nucleation rate on water vapour

supersaturation and considering temperature dependence only (as in CNT expressions). Simulations show the time evolution of nucleation and vapour deposition. Interestingly, the characteristic nucleation time is in the order of 10-20 minutes only.

20. Equation (26) is physically plausible. The value $M_\mathrm{v} = \rho_\mathrm{v} s_\mathrm{i}$ can be considered as the excess in water vapour mass density (absolute humidity), which is still available for ice formation, i.e., which is not yet converted into ice (residual or reservoir absolute humidity). Please replace the symbol M_v by the excess mass density $\Delta\rho_{\mathrm{v}\to\mathrm{i}}$ (or something similar to avoid confusion with molar mass). Fr_con is the fraction of already condensed ice (but not "percentage of uncondensed ice" as written on p. 6760, line 6). If ice supersaturation vanishes than there is no further water vapour available for further condensation, hence $\Delta\rho_{\mathrm{v}\to\mathrm{i}} = 0$ and $\mathrm{Fr}_\mathrm{con} = 1$, i.e., all excess water vapour has been removed and condensation into ice is completed. Please change (misleading) notion "mass of ice supersaturation" (p. 6760, line 8; supersaturation has, of course, no mass ...). However, the idea behind it is sound. The allowance of $\Delta\rho_{\mathrm{v}\to\mathrm{i}} \neq 0$ is an important enhancement in comparison with widely employed "saturation adjustment schemes".

21. Derivations of Eqs. (27), (28), (29) are correct.

22. Equation (30) is only traceable for me (employing from Eq. (29)) with the following assumptions:

$$\overline{J}_\mathrm{f,hom}(t) = \frac{1}{t-t_0}\int\limits_{t_0}^{t} J_\mathrm{f,hom}(t')\,\mathrm{d}t' \; ,$$

$$\rightsquigarrow \quad N_\mathrm{c,hom}(t) \approx \int\limits_{r_\mathrm{min}}^{r_\mathrm{max}} \overline{J}_\mathrm{f,hom}(t) \times (t-t_0) \times v(r_\mathrm{a}) f_\mathrm{a}(r_\mathrm{a}) \mathrm{d}r_\mathrm{a} \; .$$

$$\rightsquigarrow \quad R_\mathrm{f,hom}(t) = \frac{\mathrm{d}N_\mathrm{c,hom}(t)}{\mathrm{d}t} = \int\limits_{r_\mathrm{min}}^{r_\mathrm{max}} v(r_\mathrm{a}) f_\mathrm{a}(r_\mathrm{a}) \underbrace{\left[\frac{\mathrm{d}}{\mathrm{d}t}\left\{\overline{J}_\mathrm{f,hom}(t) \times (t-t_0)\right\}\right]}_{=\chi} \mathrm{d}r_\mathrm{a} \; ,$$

$$\begin{aligned}\chi &= \overline{J}_{\mathrm{f,hom}}(t) + \frac{\mathrm{d}\overline{J}_{\mathrm{f,hom}}(t)}{\mathrm{d}t} \times (t - t_0) \\ &= \overline{J}_{\mathrm{f,hom}}(t)\left[1 + \frac{\frac{\mathrm{d}\overline{J}_{\mathrm{f,hom}}(t)}{\mathrm{d}t} \times (t-t_0)}{\overline{J}_{\mathrm{f,hom}}(t)}\right] \\ &\approx \overline{J}_{\mathrm{f,hom}}(t)\left[1 + \underbrace{\frac{\mathrm{d}\overline{J}_{\mathrm{f,hom}}(t)}{\overline{J}_{\mathrm{f,hom}}(t)}}_{\ll 1}\right] .\end{aligned}$$

Therewith, we can write Eq. (30) as follows:

$$\rightsquigarrow \quad R_{\mathrm{f,hom}}(t) = \frac{\mathrm{d}N_{\mathrm{c,hom}}(t)}{\mathrm{d}t} = \int\limits_{r_{\mathrm{min}}}^{r_{\mathrm{max}}} v(r_{\mathrm{a}}) f_{\mathrm{a}}(r_{\mathrm{a}}) \overline{J}_{\mathrm{f,hom}}(t) \mathrm{d}r_{\mathrm{a}} \ .$$

However, here we have still $\overline{J}_{\mathrm{f,hom}}(t)$. For small t one could argue $\overline{J}_{\mathrm{f,hom}}(t) \approx J_{\mathrm{f,hom}}(t)$. Note that the integration variable in Eq. (30) is the radius of the deliquescent haze particle, r_{a}, but not r_{N} (the authors considered homogeneous nucleation, i. e., there is no catalysing substrate embedded in the haze particle (deliquescence stage)). Add correct subscript:

$$R_{\mathrm{f,hom}}(t_0) = \frac{\mathrm{d}N_{\mathrm{c,hom}}}{\mathrm{d}t} = \ldots$$

23. Derivation of Eq. (31) is correct (rederived from Khvorostyanov and Curry (2009)). In the expression for G_{n}, R denotes the universal (molar) gas constant, M_{w} the molecular weight of water (to be defined here but not only after Eq. (37)), and $L_{\mathrm{m}}^{\mathrm{ef}}$ the specific(!) latent heat of melting.

24. Derivations of Eqs. (32), (33), (34), (35) are correct, the underlying assumptions are plausible.

25. Derivation of Eq. (36) is correct.

26. Derivations of Eq. (37a), (37b) are correct. There is a **typo** in Eq. (37a) (see exponent of water vapour saturation ratio S_{w}), write correctly:

$$\Delta F_{\mathrm{cr}} = \frac{(16\pi/3)\sigma_{\mathrm{is}}^3}{\left\{\rho_{\mathrm{i}} L_{\mathrm{m}}^{\mathrm{ef}}(T) \ln\left[\frac{T}{T_0} S_{\mathrm{w}}^{G_{\mathrm{n}}(T)}\right]\right\}^2} \ .$$

Molecular weight of water, M_{w}, is defined here but employed much earlier.

27. Derivations of Eqs. (38), (39), (40a), (40b) are correct. Separation of the critical formation work into two independent dependencies (T and s_{w}) via linearisation of the denominator in Eq. (37b) is a tricky idea, very nice. (Maybe the authors had this separation in mind with their Eqs. (17), (18), see my comments above). The term $|\kappa_{\mathrm{s}} s_{\mathrm{w}}| \approx |-0.15|$ is lower than one, but not much lower than one (see p. 6767, line 10). Otherwise it must also be neglected in Eq. (39), which is not the case.

28. Derivations of Eqs. (41a), (41b), (42a), (42b) are correct. The non-trivial consequence of authors estimations is the validity of the inequality $J_{\mathrm{hom}}(T, s_{\mathrm{w}}) \ll J_{\mathrm{hom,CNT}}(T)$ at typical nucleation conditions in the upper troposphere.

29. P. 6768, line 14, and caption of Fig. 8: To avoid confusion, please express the incriminated nucleation rate ratio more precisely as $J_{\mathrm{f,hom}}(T, s_{\mathrm{w}})/J^{(0)}_{\mathrm{f,hom}}(T, s_{\mathrm{w}} = 0)$ or better as $J_{\mathrm{f,hom}}(T, s_{\mathrm{w}})/J^{(0)}_{\mathrm{f,hom}}(T)$. The *a posteriori* justification of the linearisation underlying Eq. (41) by numerical parcel simulations is very nice (p. 6768, lines 13-15).

30. Derivations of Eqs. (43a), (43b), (43c) are correct. Use correctly u_{s} (instead of u).

31. Derivations of Eqs. (44), (45) are correct, but make already here clear that $t_0 = t_{\mathrm{cr}} = 0$. The choices of the lower bounds of the coefficients $a_{1\mathrm{i}}$ and $a_{1\mathrm{w}}$ are traceable.

32. P. 6769, line 12: **Typo?** I guess here reference to Eqs. (8a), (8b) (instead of (2.2a), (2.2b)) is meant.

33. Derivations of Eqs. (46a), (46b), (47a), (47b), (47c) are correct. Equation (47c) follows (47b) only for $t_0 = t_{\mathrm{cr}} = 0$. Otherwise, the initial time $t_0 \neq 0$ must be explicitly treated in the preceding expressions.

34. Derivations of Eq. (47d) (see PK97, Eq. (4-86)) and (47e) are correct.

35. Derivations of Eqs. (48), (49) are correct. Replace G therein with G_{n} (as introduced before).

36. Derivations of Eqs. (50), (51), (52) are correct.

37. Derivations of Eqs. (53), (54) are correct. The separation of the time evolution of the crystal concentration into a linear and exponential growth period is a very interesting result. Maybe, it is of interest for the authors that similar considerations have been performed by Slezov and Schmelzer (1999) (I have rederived their calculus and found the outcome very nutritive).

38. Derivations of Eqs. (55), (56a), (56b), (56c) are correct. Please write the lhs of Eq. (55) still as $N_\mathrm{c}(s_\mathrm{w})$. Replacement of supersaturation w.r.t. to water by supersaturation w.r.t. ice comes in the next step. However, argumentation is sound. Physical interpretation of Meyers' approach is a very fine result.

39. Derivations of Eqs. (57), (58), (59), (60a), (60b), (60c) are correct, except for **typos** in (59) and (60c). The last term on the rhs of Eq. (59), and the lhs of Eq. (60c) reads correctly $r^{(3)}_\mathrm{c,ef}(\ldots)$ (superscript!).

40. In order to understand Eqs. (63)-(68), it was necessary to evaluate the **Appendix**:

    - Derivations of Eqs. (A1), (A2), (A3), (A4), (A5) are correct, except for **typo** in Eq. (A4). The lhs of Eq. (A4) reads correctly $r^{(3)}_\mathrm{c,ef}(\ldots)$ (superscript!). Add reference to Eq. (58) for B_i on p. 6785, line 1.
    - Transformation of variable and subsequent derivations of Eqs. (A6a), (A6b) are correct.
    - Transformation of variable and subsequent derivation of Eq. (A7) is correct.
    - Transformation of variable and subsequent derivations of Eqs. (A8), (A9) is correct, but contains a **typo** in Eq. (A9) (it must be c_3i in the denominator therein instead of c_1i). Write correctly:

    $$\lambda = \ldots = \frac{(u_\mathrm{s} c_\mathrm{1w} w)(\xi_\mathrm{dep} + r_0)^2}{2 c_\mathrm{i3} s_\mathrm{i,cr}} \; .$$

    - Derivations of Eqs. (A10a), (A10b), (A11) are correct. Very elegant way! Alternatively, one could also have employed the *generalised* incomplete Euler's gamma function:

    $$\Gamma(\mu, \lambda_1, \lambda_2) \;=\; \int\limits_{\lambda_1}^{\lambda_2} x^{\mu-1} \exp(-x) \mathrm{d}x = \Gamma(\mu, \lambda_1, \infty) - \Gamma(\mu, \lambda_2, \infty) \; .$$

- Derivation of Eq. (A12) is correct.
- Derivations of Eqs. (A13), (A14) are correct.
- Derivation of Eq. (A15) is correct.
- Derivations of Eqs. (A16), (A17), and (A20) (by means of textbook expressions (A17a), (A18), (A19)) are correct. (My version of Gradshteyn/Ryzhik (1994) has 1204 pages ... Maybe add: Eq. (A17a) according to GR94 (entry 8.250); Eq. (A18) according to GR94 (entry 8.359); Eq. (A19) according to GR94 (entry 8.356)). Property Ψ_1 not yet defined here (p. 6788, line after (A19)).
- Equation (A27) is obtained by considering the first three(!) summands of the asymptotic representation of $\Phi(\sqrt{\lambda})$ according to GR94 (entry 8.254).
- Equation (A29): I did not arrive at your Eq. (A29). Maybe there is a **typo** ($\sqrt{\pi}$ instead of $\sqrt{x}$?). For very small x I would write (GR94, entry (8.250)):

  $$\begin{aligned}\Phi(x) &= \frac{2}{\sqrt{\pi}} \int\limits_0^x \exp(-t^2)\,\mathrm{d}t \approx \frac{2}{\sqrt{\pi}} \exp(-x^2) \int\limits_0^x \mathrm{d}t \\ &\approx \frac{2}{\sqrt{\pi}}\, x \, \exp(-x^2) \; .\end{aligned}$$

  This form is reconcilable with the series representation of $\Phi(x)$ for $\mathrm{k} = 0$ according to GR94, entry (8.253), first item.
- Equations (A30), (A31) are clear (textbooks).
- Make use of annotation β in all equations compatible with the main text (β vs. β_{i}). As β is not explicitly related to “ice property”, maybe its better to omit subscript “i”.

41. Derivations of Eqs. (63), (64), (65), (66), (67), (68) are correct.

42. Derivation of Eq. (69) is correct with use of Eqs. (45), (51):

    $$s_{\mathrm{w}}(t) = s_{\mathrm{w,cr}} + c_{1\mathrm{w}} w t \; , \quad \beta = u_{\mathrm{s}} c_{1\mathrm{w}} w \; ,$$

    $$\rightsquigarrow \quad u_{\mathrm{s}} s_{\mathrm{w}}(t) = u_{\mathrm{s}} s_{\mathrm{w,cr}} + \beta t \; , \quad s_{\mathrm{w}}(t) = s_{\mathrm{w,cr}} + \frac{\beta}{u_{\mathrm{s}}} t \; .$$

    There is a **typo** on p. 6775, line 1.

43. Derivations of Eqs. (70a), (70b), (71), (72), (73) are correct.

44. Derivation of Eq. (74) is correct.

45. Include Ghan et al. (1993) also in the reference list (cited on p. 6776, line 2). Equation (75) corresponds to Ghan et al. (1993, Fig. 1, p. 202 therein). Good idea!

46. Derivation of Eq. (76) is correct. The obtained expression for Ψ is a very nice result.

47. Equation (77): In the first line of this equation there is a **typo** in the second summand on the rhs. Write correctly:

    $$\frac{\mathrm{d}y_\mathrm{i}'}{\mathrm{d}t} = \ldots - \frac{\Gamma_2}{\rho_\mathrm{is}} I_\mathrm{dep} \ .$$

    Therewith one arrives at the correct second line. In Eq. (77), the term y_i' in the second line and the second summand on the rhs has been *en passant* replaced with $s_\mathrm{i,cr}$ (instead of s_i), which is an approximation, or? Looking at Eq. (78), however, this replacement is not necessary.

48. Derivation of Eq. (78) is correct. Add superscript “s” on the lhs of Eq. (78) (u_s).

49. Equation (79) contains **typos**: Equation (33) reads:

    $$R_\mathrm{f,hom}(t_0) \approx N_\mathrm{a} \overline{v}_\mathrm{a} J_\mathrm{f,hom}(t_0) \ .$$

    From Eq. (41a) we have ($T = T_0$, $t = t_0$):

    $$J_\mathrm{f,hom}(T_0, s_\mathrm{w}(t_0)) = J^{(0)}_\mathrm{f,hom}(T_0) \exp\left[u_\mathrm{s}(T_0) s_\mathrm{w}(t_0)\right] \ .$$

    Furthermore, from Eqs. (45), (51) we have:

    $$s_\mathrm{w}(t) = s_\mathrm{w,cr} + c_\mathrm{1w} w t \ , \quad \beta = u_\mathrm{s} c_\mathrm{1w} w \ ,$$

    $$\rightsquigarrow \quad u_\mathrm{s} s_\mathrm{w}(t) = u_\mathrm{s} s_\mathrm{w,cr} + \beta t \ , \quad s_\mathrm{w}(t) = s_\mathrm{w,cr} + \frac{\beta}{u_\mathrm{s}} t \ .$$

    Therewith Eq. (79) must be correctly written as:

    $$\begin{aligned} R_\mathrm{f,hom}(t_0) &\approx N_\mathrm{a} \overline{v}_\mathrm{a} J^{(0)}_\mathrm{f,hom}(T_0) \exp\left[u_\mathrm{s}(T_0) s_\mathrm{w}(t_0)\right] \\ &\approx N_\mathrm{a} \overline{v}_\mathrm{a} J^{(0)}_\mathrm{f,hom}(T_0) \exp\left[u_\mathrm{s} s_\mathrm{w,cr} + \beta t_0\right] \ . \end{aligned}$$

50. Derivation of Eq. (80) is correct. Please add the assumption employed therein (definition and treatment of the lower integration limit):

$$\begin{aligned} N_{\mathrm{cm}}(t_{\mathrm{m}}) &\approx N_{\mathrm{a}}\overline{v}_{\mathrm{a}}J^{(0)}_{\mathrm{hom}} \int\limits_{t_{\mathrm{th},1}}^{t_{\mathrm{m}}} \exp\left[u_{\mathrm{s}}(T_0)s_{\mathrm{w,cr}} + \beta t_0\right]\,\mathrm{d}t_0 \\ &\approx N_{\mathrm{a}}\overline{v}_{\mathrm{a}}J^{(0)}_{\mathrm{hom}}\beta^{-1}\,\exp\left\{u_{\mathrm{s}}(T_0)s_{\mathrm{w}}(t_{\mathrm{m}})\left[1-\underbrace{\frac{s_{\mathrm{w}}(t_{\mathrm{th},1})}{s_{\mathrm{w}}(t_{\mathrm{m}})}}_{\ll 1}\right]\right\}\,. \end{aligned}$$

51. Derivation of Eqs. (80), (81) are correct (nice approach). From Eqs. (78), (79), (80) to Eqs. (80), (81), however, the authors have *en passant* replaced $s_{\mathrm{i,max}}$ with $s_{\mathrm{i,cr}}$ (give a comment). Add missing superscript "s" to write correctly u_{s} in Eq. (82).

52. P. 6777, lines 10-20: I recommend to introduce a short sequence at the appropriate place in the text, where all employed times and time scales are defined together.

53. Equations (83), (84), (85) are parameterisations (derived from parcel model simulations, "nothing" to rederive here). Useful result.

54. Derivation of Eq. (86) is correct. Add subscript "i" to correctly write B_{i}.

55. Derivation of Eq. (87) is correct with assumption of $\Phi\left(\sqrt{\beta t_{\mathrm{m}}}\right) \approx 1$ according to Eq. (A28) for $\beta t_{\mathrm{m}} \gg 1$ and $\exp(\lambda) \approx 1+\lambda \approx 1$. Add superscript "i" to correctly write B_{i}.

56. The derivation of Eq. (88) is correct, but Eq. (89) is subject of **three typo's**. From Eqs. (81), (82) we have:

$$N_{\mathrm{c}}(t_{\mathrm{m}}) \approx (4\pi D_{\mathrm{v}})^{-1}u_{\mathrm{s}}^{-1}\left(\frac{c_{1\mathrm{i}}}{c_{1\mathrm{w}}}\right)\left(1+s_{\mathrm{i,cr}}\right)s_{\mathrm{i,cr}}^{-1}\Psi^{-1}\,.$$

Replacing therein Ψ from Eq. (87) we obtain:

$$N_{\mathrm{c}}(t_{\mathrm{m}}) \approx \underbrace{\left\{\frac{(4\pi D_{\mathrm{v}})^{-1}u_{\mathrm{s}}^{-1}\left(\dfrac{c_{1\mathrm{i}}}{c_{1\mathrm{w}}}\right)\left(1+s_{\mathrm{i,cr}}\right)s_{\mathrm{i,cr}}^{-1}}{\left(\dfrac{\pi}{2}\right)^{1/2}\left(c_{3\mathrm{i}}s_{\mathrm{i,cr}}\right)^{1/2}u_{\mathrm{s}}^{-3/2}}\right\}}_{=K_{\mathrm{i,diff}}}\times\left(c_{1\mathrm{w}}w\right)^{3/2}\,,$$

By virtue of Eq. (12),

$$c_{3\mathrm{i}} = \frac{D_{\mathrm{v}}\rho_{\mathrm{is}}}{\rho_{\mathrm{i}}\Gamma_2} \ , \quad c_{3\mathrm{i}}^{-1/2} = \left(\frac{D_{\mathrm{v}}\rho_{\mathrm{is}}}{\rho_{\mathrm{i}}\Gamma_2}\right)^{-1/2} = \left(\frac{\rho_{\mathrm{i}}\Gamma_2}{D_{\mathrm{v}}\rho_{\mathrm{is}}}\right)^{1/2}$$

one arrives at Eq. (89):

$$K_{\mathrm{i,diff}} = (2\pi D_{\mathrm{v}})^{-3/2} \underbrace{\left(\frac{\rho_{\mathrm{i}}\Gamma_2}{\rho_{\mathrm{is}}}\right)^{1/2}}_{\text{Check it!}} u_{\mathrm{s}}^{1/2} \underbrace{\left(\frac{c_{1\mathrm{i}}}{c_{1\mathrm{w}}}\right)}_{\text{Check it!}} (1 + s_{\mathrm{i,cr}})\, s_{\mathrm{i,cr}}^{-3/2} \ .$$

Replacing therein u_{s} by use of Eq. (42a),

$$u_{\mathrm{s}} = \frac{2R}{k_{\mathrm{B}} M_{\mathrm{w}} L_{\mathrm{m}}^{\mathrm{ef}}} \, \frac{\Delta F_{\mathrm{cr},0}}{\ln(T_0/T)} \ ,$$

one finally arrives at:

$$\begin{aligned} K_{\mathrm{i,diff}} &= (2\pi D_{\mathrm{v}})^{-3/2} \underbrace{\left(\frac{\rho_{\mathrm{i}}\Gamma_2}{\rho_{\mathrm{is}}}\right)^{1/2}}_{\text{Chek it!}} \underbrace{\left(\frac{2R}{k_{\mathrm{B}} M_{\mathrm{w}} L_{\mathrm{m}}^{\mathrm{ef}}} \, \frac{\Delta F_{\mathrm{cr},0}}{\ln(T_0/T)}\right)^{1/2}}_{\text{Chek it!}} \\ &\quad \times \underbrace{\left(\frac{c_{1\mathrm{i}}}{c_{1\mathrm{w}}}\right)}_{\text{Chek it!}} (1 + s_{\mathrm{i,cr}})\, s_{\mathrm{i,cr}}^{-3/2} \ . \end{aligned}$$

57. Derivation of Eq. (90) is correct (it is a somewhat cruder approximation of $\Phi(\sqrt{\lambda})$ than the Taylor expansion given by Eq. (A27)).

58. Derivation of Eq. (91) is correct, but the first equality in Eq. (91) is subject of a **typo**. We start with Eq. (74):

$$\begin{aligned} \Psi &= \mathrm{e}^{\lambda}\beta^{-1/2}\sqrt{\pi}\,[\Delta\Phi]\,\Xi \\ &\quad + B_{\mathrm{i}}^{1/2}\beta^{-3/2}\left[\lambda^{1/2} - (\lambda + \beta t)^{1/2}\exp(-\beta t)\right] \\ &\quad + 2\xi_{\mathrm{d}}\beta^{-1}\left[\exp(-\beta t) - 1\right] \ , \\ \Xi &= \left(\frac{1}{2}\right) B_{\mathrm{i}}^{1/2}\beta^{-1} + \xi_{\mathrm{d}}^2 B_{\mathrm{i}}^{-1/2} \ , \\ \Delta\Phi &= \Phi(\sqrt{\lambda + \beta t}) - \Phi(\sqrt{\lambda}) \ . \end{aligned}$$

We employ the following approximations:

$$\begin{aligned}
\Phi(\sqrt{\lambda+\beta t}) &\rightarrow 1\ , \\
\Phi(\sqrt{\lambda}) &\approx 1-\frac{1}{\sqrt{\pi}}\lambda^{-1/2}\mathrm{e}^{-\lambda}\left(1-\frac{1}{2\lambda}\right) \\
\rightsquigarrow \quad \Delta\Phi &\approx \frac{1}{\sqrt{\pi}}\lambda^{-1/2}\mathrm{e}^{-\lambda}\left(1-\frac{1}{2\lambda}\right)\ , \\
\exp(-\beta t) &\rightarrow 0\ .
\end{aligned}$$

Therewith we can write:

$$\Psi = \mathrm{e}^{\lambda}\mathrm{e}^{-\lambda}\pi^{1/2}\pi^{-1/2}\lambda^{-1/2}\underbrace{\left(1-\frac{1}{2\lambda}\right)}_{\approx 1}\beta^{-1/2}\Xi + B_{\mathrm{i}}^{1/2}\beta^{-3/2}\lambda^{1/2} - 2\xi_{\mathrm{d}}\beta^{-1}$$

Finally, we arrive at the first equality in Eq. (91):

$$\Psi \approx \lambda^{-1/2}\beta^{-1/2}\Xi + B_{\mathrm{i}}^{1/2}\beta^{-3/2}\underbrace{\lambda^{1/2}}_{\text{Check!}} - 2\xi_{\mathrm{d}}\beta^{-1}\ .$$

59. Derivation of the second part of Eq. (91) is correct again (hence, the typo in the first part is indeed a "typo" only, but not an error).

60. Use unique annotation: $\xi_{\mathrm{dep}} \leftrightarrow \xi_{\mathrm{d}}$ (p. 6780, lines 2-3, and later).

61. Derivation of Eq. (92) is correct. Add subscript "i" to correctly write B_{i}.

62. Derivations of Eqs. (93), (94) are correct. The agreement of the relation $N_{\mathrm{cm}} \propto w^2$ with Ren and McKenzie (2005) is a very nice and interesting result, which was not *a priori* expectable. Also the relation of $N_{\mathrm{cm}} \propto \alpha_{\mathrm{d}}^{-1}$ and its implications are very interesting (physical interpretation of empirical findings).

63. Derivation of Eq. (95) is correct.

64. Derivation of Eq. (96) contains an **error** ($N_{\mathrm{cm,l}}$ has incorrect physical dimension here) (it is your last equation, thus I cannot check wether it is just a typo). We start with (81), (82)

$$N_{\mathrm{c}}(t_{\mathrm{m}}) \approx (4\pi D_{\mathrm{v}})^{-1}u_{\mathrm{s}}^{-1}\left(\frac{c_{1\mathrm{i}}}{c_{1\mathrm{w}}}\right)(1+s_{\mathrm{i,cr}})\,s_{\mathrm{i,cr}}^{-1}\Psi^{-1}\ .$$

Replacing therein Ψ from Eq. (95) we obtain:

$$\begin{aligned}
N_\mathrm{c}(t_\mathrm{m}) &\approx (4\pi D_\mathrm{v})^{-1} u_\mathrm{s}^{-1} \left(\frac{c_\mathrm{1i}}{c_\mathrm{1w}}\right) (1+s_\mathrm{i,cr})\, s_\mathrm{i,cr}^{-1} \times \left(\frac{\beta}{r_0}\right) \\
&\approx (4\pi D_\mathrm{v})^{-1} u_\mathrm{s}^{-1} \left(\frac{c_\mathrm{1i}}{c_\mathrm{1w}}\right) (1+s_\mathrm{i,cr})\, s_\mathrm{i,cr}^{-1} \left(\frac{u_\mathrm{s} c_\mathrm{1w}}{r_0}\right) \times w \\
&\approx (4\pi D_\mathrm{v})^{-1} \left(\frac{c_\mathrm{1i}}{r_0}\right) (1+s_\mathrm{i,cr})\, s_\mathrm{i,cr}^{-1} \times w \ , \\
&\propto w \ .
\end{aligned}$$

Therewith, N_cm is given in correct unity of m^{-3}.

65. P. 6781, line 10: Write correctly McKenzie.
66. P. 6782, line 2: Add full reference to Jensen in the reference list.
67. Please check completeness of your reference list.

6 Final comment

A great and nutritive work, congrats to the authors! I enjoyed to evaluate this very interesting and instructive paper on freezing theory and modelling.

Anonymous reviewer

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