

# Responses to reviewer 2: discussion (acp-2020-136)

September 15, 2020

The authors would like to thank the anonymous referee for taking the time to review the manuscript and for addressing various important issues which helped us improving the quality of our work. We have decided to implement substantial changes to our manuscript based on his remarks, the details of which are addressed point by point below.

## 1 Major remarks

**R2-Ma1.** In the simulation, the surface temperature of the hydrometeor was fixed at  $0^{\circ}\text{C}$ . The ambient temperature was varied between  $-20^{\circ}\text{C}$  and  $-40^{\circ}\text{C}$ . Can the authors justify the choice of ambient temperatures for this study? Can the authors cite observations that detect wet growth at such low temperatures? There is a comprehensive experimental study by Greenan and List (JAS, 1995) on the surface temperature of hydrometeors at different conditions. It is unlikely that wet growth would occur at such low temperatures.

We agree with the reviewer that the temperature gap investigated in this work exceeds the range of values which are to be reasonably expected for natural clouds. The intention behind this was to demonstrate that extreme conditions are required in order to produce significant wake-induced ice enhancement, and that these conditions are unlikely to be observed in nature. However, this point apparently has not been communicated clearly enough. Furthermore, the decision to portray the contour plots at  $T_{\infty} = -30^{\circ}\text{C}$  seems unfortunate. We have therefore decided to vary the ambient temperature in a smaller range ( $-15^{\circ}\text{C} < T_{\infty} < 0^{\circ}\text{C}$ ) according to the experimental observations by Greenan and List (1995) and depict the contours at  $T_{\infty} = -15^{\circ}\text{C}$ .

*Adjustments to the manuscript:* The adjusted versions of the affected figures can be found in section 4 of this document. The text in the manuscript will be adjusted accordingly.

**R2-Ma2.** In section 3.3 the authors define a parameter called ice enhancement factor to quantify the effects of enhanced supersaturation. This parameter is justified, but the expression used for finding NIN is not. This expression is used in Baker 1991, but none of the recent work on ice nucleation use this expression (to the best of reviewer's knowledge). Such a power law relationship between the number concentration of ice nuclei and supersaturation seems physically inconsistent. For example, barring the effects of wettability/chemical composition, as the supersaturation is increased, the size of the aerosols that is activated is reduced. For ice nucleation, the size of the nucleus is an important parameter, and as the size of the nucleus is reduced, its ice nucleating efficiency is also reduced. So, the number concentration of ice nuclei may not increase with supersaturation like a power law with such high exponents as mentioned in this paper. Furthermore, such a power law may not even be applicable to CCN concentrations when the supersaturation is quite high (Q. Ji and G. Shaw 1998 GRL). So, the applicability of such a power law to ice nuclei concentration is highly questionable. Can the authors comment/justify the applicability of the expression for NIN, as the whole of section 3.3 and the most important conclusion in the paper is based on this expression? This comment needs to be addressed in detail to support the conclusion. If this issue

cannot be addressed satisfactorily, the authors can consider presenting their arguments based on fractional cloud volume (like in section 3.2) that is exposed to the enhanced supersaturation due to the falling wet hydrometeors.

The power-law equation for  $N_{IN}$  indeed appears to be rarely used in recent literature. We have therefore decided to replace it by the exponential law provided by Meyers, DeMott, and Cotton (1992) which has been obtained from continuous-flow diffusion chamber (CFDC) measurements of natural aerosols. The constitutive relation is a parametrization of both the deposition and condensation-freezing mechanisms of ice nucleation and reads

$$N_{IN} = 0.528 \exp(12.96s_i) \text{ m}^{-3}. \quad (1)$$

It is reported to be strictly valid for the following parameter range (the range of the CFDC data).

$$-20^\circ\text{C} < T < -7^\circ\text{C}, \quad 2\% < s_i < 25\%, \quad -5\% < s_w < 4.5\% \quad (2)$$

The temperatures of interest in the current work (after making the adjustments stated in R2-Ma1) fall well into the range of validity. The distribution of  $s_i$  in the wake is shown in fig. 3 (an updated version of fig. 6 of the discussion paper). For most ambient temperatures,  $s_i$  does not exceed 25%. At  $T_\infty = -15^\circ\text{C}$ , regions where  $s_i$  is slightly larger than 0.25 exist, but only occupy a small volume within the domain, and hence imprecisions are likely to be insignificant for integral quantities. When looking at the volumetric distribution of  $s_w$  in the wake in fig. 4, it can be seen that water supersaturation exceeds the CFDC data range in significant portions of the domain when  $T_\infty \lesssim -10^\circ\text{C}$ . Due to these relatively large supersaturations w.r.t. liquid, eq. (1) might underestimate the contribution of the condensation-freezing mode, as this mode shows increased activity under these conditions as has been demonstrated by Schaller and Fukuta (1979) for various substances. However, we are not aware of any parametrization of condensation-freezing nucleation for natural aerosols which can be directly applied under these conditions.

The main conclusion of this work is not affected by the substitution of the nucleation law. The global ice enhancement factor computed with eq. (1) behaves similar to the power law estimation with  $\alpha \approx 3$  as can be seen when comparing fig. 7 of this document to fig. 9 of the discussion paper.

*Adjustments to the manuscript:* All affected figures have been updated and the text will be adjusted accordingly. The range of validity will be justified similar to the response above. Figure 4 will be added to the manuscript as it raises awareness concerning the applicability of eq. (1) and provides information on the supersaturation w.r.t. liquid which may be interesting for the reader.

**R2-Ma3. The analysis in section 3.3 can be recast as the cloud volume that is exposed to very high supersaturation in the wake. This analysis concludes that the fraction of the cloud volume exposed to the high supersaturation in the wake is insignificant. There is a similar study published recently (Prabhakaran et al 2020 (GRL)). Their analysis concluded that a significant fraction of the cloud volume can be exposed to the high wake supersaturation during the lifetime of the cloud. Can the authors comment about the difference between these two analyses?**

It is true that the current manuscript investigates the instantaneous exposure of a cloud subvolume to meteor-induced supersaturation, while the analysis presented in Prabhakaran, Kinney, Cantrell, Shaw, and Bodenschatz (2020) focuses on the volume swept by the meteors. The latter approach is reasonable since history effects in ice nucleation should be taken into account, i.e. it should be taken into account that ice nuclei which have been activated in the wake of a hydrometeor may stay activated once they are not exposed to the wake anymore. However, the difference in the two analyses can be regarded as two limiting cases of the nucleation rate, namely one which is limited by the rate of renewal of fluid in the wake (analysis of Prabhakaran et al. (2020)) and one which is limited by the time scale of nucleation (our analysis), as will be demonstrated in the following.

Following Prabhakaran et al. (2020), the time required for a significant volume of air to be sampled by hydrometeor wakes is estimated by

$$\tau_{sweep} = \left( \int_0^\infty \bar{N}_{met}(D) \dot{V}_{sweep}(D) dD \right)^{-1}, \quad (3)$$

where  $\bar{N}_{met}(D)$  denotes the number concentration density of ice particles and  $\dot{V}_{sweep} = \epsilon v_p D^2 \pi / 4$  is the volumetric flow rate of air which is swept by a hydrometeor with diameter  $D$  and velocity  $v_p$ . The unknown factor  $\epsilon$  is assumed to be of the order of unity. Using eq. (10) of the discussion paper for  $\bar{N}_{met}(D)$  and the terminal velocity for smooth spheres, we obtain  $\tau_{sweep} \approx 110\text{s}$  at  $T = -15^\circ\text{C}$ , which fits the estimation of Prabhakaran et al. (2020) well. The use of an empirical law for the terminal velocity of frozen hydrometeors of natural shape leads to longer time scales, however, they are found to be of similar order of magnitude. This analysis tells us that even though the cloud volume which is instantaneously exposed to high supersaturations is very small, it does not take a long time to expose a significant volume because the rate at which air is swept by the meteors is high.

In the following we attempt to quantify the nucleation rate of INP, henceforth denoted as  $j_{met}$ , from our simulation data and the swept-volume argument. Under the assumption that nucleation occurs sufficiently fast to achieve the INP concentrations predicted by eq. (1), the nucleation rate can be estimated from the number of INP activated in the wake and the time it takes to replenish the volume of fluid affected by high supersaturations. The former is obtained directly from our simulation data by computing the volume integral  $\int_{\Omega(D)} (N_{IN}(\mathbf{x}) - N_{IN,\infty}) d\mathbf{x}$  while the latter is difficult to define objectively. We propose to estimate the time scale of wake renewal by

$$\tau_{expo} = \frac{V_{aff}}{\dot{V}_{sweep}} \quad (4)$$

where  $V_{aff} = \gamma D^3 \pi / 6$  is the volume affected by the wake of a hydrometeor of diameter  $D$ , which should be proportional to the volume of the hydrometeor. The prefactor  $\gamma$  is currently unknown, but might be related to the concept of supersaturated volume defined in the manuscript. The time scale  $\tau_{expo}$  may be regarded as the characteristic time a fluid volume is exposed to high supersaturations, and hence the subscript. From the definitions of  $V_{aff}$  and  $\dot{V}_{sweep}$  it follows that

$$\tau_{expo} \propto \frac{D}{v_p}, \quad (5)$$

with the constant of proportionality being referred to as  $C_{expo}$  hereafter. This new constant contains both unknown coefficients  $\epsilon$  and  $\gamma$  and might be interpreted as the non-dimensional streamwise length of the wake. Again, this length is difficult to define rigorously due to the asymptotic decay of supersaturation. However, judging from fig. 5 it is likely that  $C_{expo} = \mathcal{O}(10)$  which results in exposure times of the order of  $\tau_{expo} \approx 5\text{ms}$  for all diameters of interest. The swept-volume limited nucleation rate for an ensemble of meteors is then given by

$$j_{met}^{expo} = \int_0^\infty \bar{N}_{met}(D) \frac{1}{\tau_{expo}(D)} \int_{\Omega(D)} (N_{IN}(\mathbf{x}) - N_{IN,\infty}) d\mathbf{x} dD \quad (6)$$

under the assumptions that the rate of activation of INP is sufficiently fast, a sufficient number of interstitial aerosol particles are present and that those are homogeneously distributed within the wake.

The exposure time estimated previously is substantially shorter than the time scales usually relevant for cloud modelling (few milliseconds compared to minutes). As eq. (1) has been developed for cloud modelling, the validity of the assumption that INP activation can be regarded as instantaneous at the time scales considered should be brought into question. Indeed, classical nucleation theory (Fletcher, 1958) suggests that nucleation is a time-dependent process until the activated fraction of AP approaches unity. From concentrations of INP obtained from continuous-flow diffusion chamber (CFDC) experiments, the nucleation rate may be estimated by taking into account the residence time in the apparatus  $\tau_{nucl}$  (Hoose and Möhler, 2012). Since eq. (1) is based on CFDC data, we make the conjecture that the local nucleation rate may be approximated by the relationship

$$j_{IN}(\mathbf{x}) \approx N_{IN}(\mathbf{x}) / \tau_{nucl}. \quad (7)$$

In Hoose and Möhler (2012) residence times ranging from 1.6s to 120s are reported for various CFDC experiments. The primary data used to obtain eq. (1) also suggests that the peak concentration  $N_{IN}$  is achieved with residence times of approximately 10s (Al-Naimi and Saunders, 1985, fig. 6) and that

shorter residence times lead to lower concentrations (in accordance to the arguments stated before). As can already be seen,  $\tau_{nucl} \gg \tau_{expo}$ , and hence, the supposition that the INP concentrations predicted by eq. (1) are achieved within the exposure time is disproved. The rate-limited nucleation rate of the ensemble of hydrometeors is then given by

$$j_{met}^{nucl} = \int_0^\infty \bar{N}_{met}(D) \frac{1}{\tau_{nucl}} \int_{\Omega(D)} (N_{IN}(\mathbf{x}) - N_{IN,\infty}) d\mathbf{x} dD, \quad (8)$$

under the assumption that interstitial AP are entrained sufficiently fast into the wake (which is reasonable given the arguments by Prabhakaran et al. (2020)) and that they are distributed homogeneously within the wake.

Figure 8 shows the meteor-induced nucleation rate for both limiting cases. We assume that the most likely values for the tunable parameters are  $C_{expo} = 10$  and  $\tau_{nucl} = 10\text{s}$  (solid lines), but also investigate the range  $C_{expo} \in [1, 100]$  and  $\tau_{nucl} \in [1, 100]\text{s}$  in order to pay regard to the uncertainties associated with these quantities (shaded area). The swept-volume limited estimation  $j_{met}^{expo}$  is at least two orders of magnitude higher than rate-limited estimation  $j_{met}^{nucl}$ . A high relevance of the wake-induced nucleation is indicated by  $j_{met}^{expo}$ , as it would only take around 40s for the number concentration of wake-activated INP to match the concentration of primary meteors at  $T_\infty = -10^\circ\text{C}$ . In contrast, it would take around 82h to achieve this concentration with the rate-limited estimation, which suggests that this process is of little relevance in clouds. The large disparity between the results is explained by the differences in time scales, i.e.  $\tau_{expo} = \mathcal{O}(10^{-3}\text{s})$  while  $\tau_{nucl} = \mathcal{O}(10^1\text{s})$ , as has been stated earlier. Physically this implies that the time a fluid volume is exposed to high supersaturations is too short to create considerable concentrations of INP.

This result can be linked to the ice enhancement factor introduced in the manuscript, as this quantity directly relates to the rate-limited estimation of the nucleation rate:

$$\langle f_i \rangle_{\mathcal{V}} = \frac{j_{met}^{nucl}}{N_{IN,\infty}/\tau_{nucl}} + 1. \quad (9)$$

Furthermore, it is straightforward to show that  $\tau_{expo} \propto \tau_{sweep}$  for a given meteor concentration, which implies that as soon as a significant cloud volume is swept quickly at low volume fractions of ice, the transient exposure of a cloud fluid element to the wake will be short.

In order for wake-induced ice nucleation to be a relevant SIP, the nucleation rate in the wake needs to be significantly higher than what has been estimated in this work. If the conjectures presented in the above analysis hold, the most feasible way to accomplish this is that the overall concentration of AP is higher than what has been assumed in this work implicitly through eq. (1), i.e. this mechanism may gain importance in clouds with a high number of possible nucleation sites. It might also be conceivable that the AP concentration is locally enhanced in the wake due to flow-induced clustering, i.e. AP may be preferentially located in highly supersaturated region as opposed to the ambient, although this mechanism unlikely leads to the required concentrations. Nonetheless, we suggest that an analysis of individual AP trajectories may be beneficial in the future to clarify the importance of this SIP, as such an analysis would allow for a more rigorous assessment of the nucleation rate by providing access to the actual residence times of AP and by enabling the use of more fundamental constitutive laws for ice nucleation (such as classical nucleation theory).

*Adjustments to the manuscript:* Subsection 3.3 will be fully revised and extended by the discussion above. Figure 8 will be added to the manuscript. The notation concerning ice nuclei concentration and the concentration density of primary hydrometeors will be adjusted.

## 2 Comments

**R2-Co1.** In lines 108-109, the authors state that buoyancy contributions to momentum due to the variations in temperature and water vapor is negligible. Can the authors justify this statement briefly (a few lines) by quoting the value of the relevant parameter, e.g. Richardson number, along with the reference to Chouippe et al 2019? Would it be insignificant when the temperature difference between the ambient and the drop is  $40^\circ\text{C}$ ?

**Similarly, in lines 118-119, can the authors justify briefly why the variations in the vertical velocity is not important in the present context?**

The Richardson number for a freely falling heated sphere has been defined in Chouippe, Kraye, Uhlmann, Dušek, Kiselev, and Leisner (2019) as

$$Ri_T = \frac{1}{\left(\frac{\rho_p}{\rho_\infty} - 1\right)} \frac{T_p - T_\infty}{T_\infty}, \quad (10)$$

where  $T_\infty$  is given in Kelvin (see (Chouippe et al., 2019, Appendix A) for the derivation). In accordance to the value stated in the discussion paper, we assume  $\rho_p = 600\text{kg m}^{-3}$  and  $\rho_\infty = 1\text{kg m}^{-3}$ . For an ambient temperature of  $T_\infty = -40^\circ\text{C} = 233.15\text{K}$  and a particle temperature of  $T_p = 0^\circ\text{C} = 273.15\text{K}$ , we obtain

$$Ri_T \approx 3 \cdot 10^{-4}. \quad (11)$$

In (Chouippe et al., 2019, fig. 7) it is documented that the recirculation length of the wake, a quantity which is shown to be sensitive to buoyancy effects, does not differ significantly from passive scalar transport when  $Ri_T = 1 \cdot 10^{-3}$ , which is a value significantly higher than what is investigated in the present manuscript.

The Richardson number due to variations in water vapor content is defined as

$$Ri_{n_v} = -\frac{1}{\left(\frac{\rho_p}{\rho_\infty} - 1\right)} \frac{M_w - M_d}{N_A \rho_\infty} (n_{v,p} - n_{v,\infty}), \quad (12)$$

where  $N_A$  is the Avogadro constant,  $M_w$  the molar mass of water and  $M_d$  the mixture molar mass of dry air (Chouippe et al., 2019, eq. (45)). Using the temperatures stated in the previous paragraph and the vapor boundary conditions stated in the manuscript, we obtain

$$Ri_{n_v} \approx 5 \cdot 10^{-6}, \quad (13)$$

which is orders of magnitudes smaller than  $Ri_T$ . Please note that eq. (4) in the manuscript is incorrect and should read

$$n_v = e/k_b T. \quad (14)$$

This will be corrected in the revised version of the manuscript.

Fluctuations in  $\mathbf{v}_p$  are only important in the context of this manuscript if they lead to modifications in the structure of the wake. The equations of motion for the spherical particle suggest that the time scale of particle acceleration is proportional to  $\rho_p/\rho_\infty$  (Chouippe et al., 2019, eq. (8)). If this time scale is much larger than the observation time of interest, which in our case is  $L_x/v_p$  with  $L_x$  being the length of the simulation domain, the wake will have a structure similar to that of particle falling through a fluid at rest with constant velocity. In other words the structure of the wake is only altered if the particle changes its falling direction significantly during the observation time. In (Chouippe et al., 2019, § 3.3) it is reported that a freely falling particle with  $\rho_p/\rho_\infty = 10$  already behaves very similar to a fixed particle ( $\rho_p/\rho_\infty \rightarrow \infty$ ) e.g. in terms of centerline temperature evolution and half-width of the thermal wake. Since we assume a much larger density ratio of 600, it seems unlikely that fluctuations in particle velocity have an impact on the shape of the wake.

*Adjustments to the manuscript:* Equation (4) of the discussion paper will be corrected. A sentence will be added to the manuscript stating that the wake of a freely falling hydrometeor behaves similar to that of a fixed particle due to the high value of the density ratio.

**R2-Co2. In a deep convective cloud, the hydrometeors are falling through a turbulent environment. Can the authors comment about the role of turbulent fluctuations in the ambient? How would the volume of the supersaturated region change with turbulence intensity in the ambient? There are some heat transfer studies from a heated sphere in a turbulent environment (Bagchi and Kottam 2008, Phys of Fluids). Can this be extended to the current study? It might be worthwhile to briefly discuss this as a part of future**

**work.**

The study of Bagchi and Kottam (2008) is very helpful when the effect of ambient turbulence on the supersaturated volume and ice enhancement is discussed. The parameter range investigated in their work corresponds reasonably well to the scenario of a hydrometeor with a diameter of a few millimeter settling under atmospheric conditions, where the largest flow scales are expected to be  $\mathcal{O}(100\text{m})$  and the smallest scales around  $\mathcal{O}(1\text{mm})$  (Lehmann, Siebert, and Shaw, 2009).

We know from simple mixing parcel models (e.g. (Chouippe et al., 2019, fig. 17) or (Prabhakaran et al., 2020, fig. 4)) that the highest supersaturations occur in regions where the temperature of the mixture is roughly halfway between  $T_\infty$  and  $T_p$ , i.e.  $\tilde{T} \approx 0.5$  with  $\tilde{T}$  being the non-dimensionalized temperature as introduced in the manuscript. In (Bagchi and Kottam, 2008, fig. 17) it is demonstrated that the centerline temperature in the wake decays significantly faster if the background flow is turbulent, especially when  $\tilde{T} \lesssim 0.4$ . Therefore it is to be expected that supersaturation decays faster in the wake than it does for a uniform inflow, and thus, the supersaturated volume as well as the ice enhancement are most likely smaller if the ambient is turbulent.

However, Bagchi and Kottam (2008) also investigated the effect of turbulence on the heat and mass transfer coefficient. While the mean value of the Nusselt number remains mostly unaffected, strong fluctuations in its value can be observed. This presumably leads to a more intermittent behavior of the temperature and vapor fields. The role of intermittency on ice nucleation activity still needs to be investigated more thoroughly, especially when the distribution of aerosol particles is explicitly considered (a point which was suggested to be investigated as part of future work). If regions of strong supersaturation coincide with regions where AP are preferentially located, intermittency might promote ice nucleation as supersaturation and nucleation rate are non-linearly linked to the temperature/vapor fields.

*Adjustments to the manuscript:* A paragraph discussing the influence of turbulence will be added to the manuscript.

*Note:* This response is the same as the response to remark R1-Co2 raised by Alexei Korolev, due to the strong similarity of the remarks.

**R2-Co3. Can the authors comment on how the supersaturated volume would be affected in the presence of cloud droplets and ice particles in the ambient?**

If a second riming ice particle or warm droplet approaches the settling ice particles, their thermal/vapor wakes will interact, which probably leads to reduced heat and mass transfer as temperature/vapor gradients are dampened in the boundary layer. Therefore the supersaturated volume induced by two nearby hydrometeors is likely to be smaller than their sum.

If a cloud droplet or ice particle which is colder than the riming meteor enters the wake, e.g. a hydrometeor at ambient temperature, water vapor may be removed from the gas phase by diffusional growth of the secondary hydrometeor. This presumably leads to a depletion of supersaturation, and hence, the supersaturated volume decreases.

### 3 Minor remarks

**R2-Mi1. Excess supersaturation - notation difference between Eq. 15 and Fig 4 caption. Fig 4 caption has a “\*” on top of “s”.**

We thank the reviewer for pointing out this mistake, which will be corrected in the revised version of the manuscript.

## 4 Figures

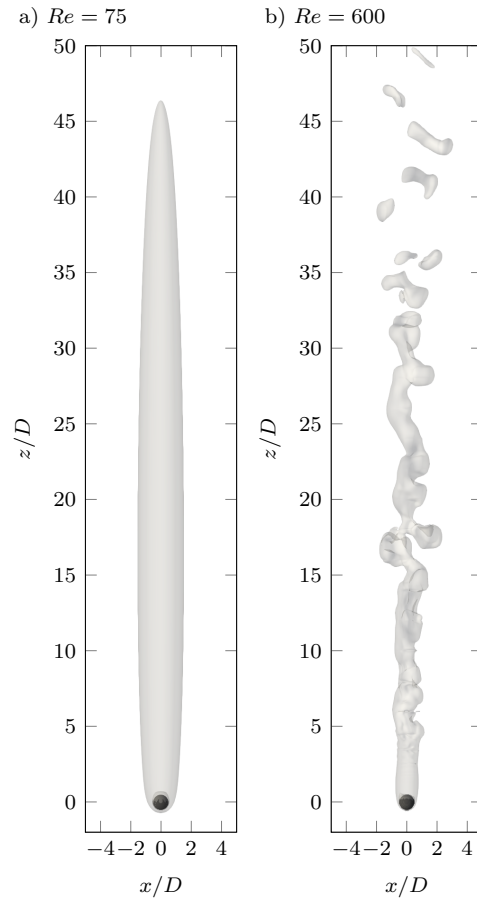


Figure 1: Isosurfaces of supersaturation in the wake at  $T_\infty = -15^\circ\text{C}$ . The value of the isocontour is  $\tilde{s}_i^* = 0.02$ , i.e. two percentage points higher than the ambient supersaturation. Two different wake regimes are depicted, which correspond to two different hydrometeor sizes in our framework. (a) axisymmetric regime at  $Re = 75$ , (b) chaotic regime at  $Re = 600$ .

Changelog: changed ambient temperature to  $T_\infty = -15^\circ\text{C}$ ; changed isocontour threshold to  $\tilde{s}_i^* = 0.02$ ; adapted caption accordingly

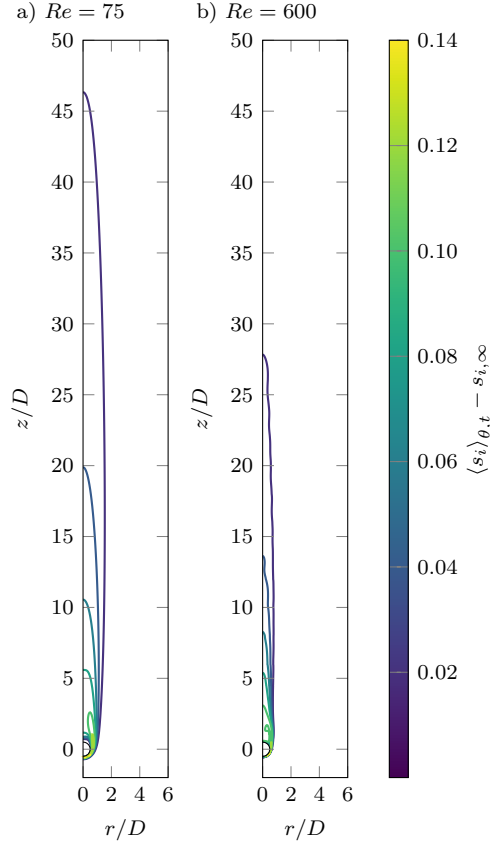


Figure 2: Contours of excess supersaturation in the wake, averaged over time and azimuthal direction at  $T_\infty = -15^\circ\text{C}$ . (a) axisymmetric regime at  $Re = 75$ , (b) chaotic regime at  $Re = 600$ .  
 Changelog: changed ambient temperature to  $T_\infty = -15^\circ\text{C}$ ; adapted caption accordingly

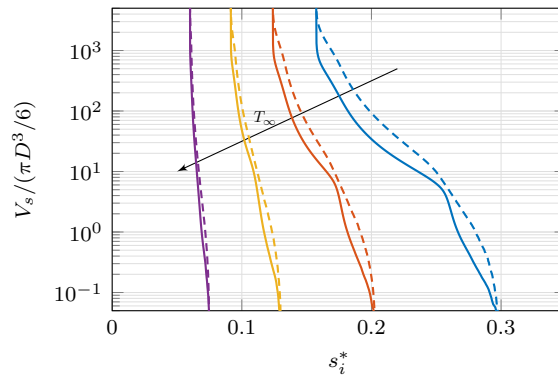


Figure 3: Volume of air where supersaturation w.r.t. ice exceeds a given threshold as a function of the threshold. The volume is normalized by the volume of the ice particle and four different ambient temperatures are shown:  $T_\infty = -6^\circ\text{C}$  (—),  $T_\infty = -9^\circ\text{C}$  (—),  $T_\infty = -12^\circ\text{C}$  (—),  $T_\infty = -15^\circ\text{C}$  (—). Solid lines correspond to  $Re = 600$  (chaotic regime), while dashed lines show the data obtained for  $Re = 75$  (axisymmetric regime).

Changelog: changed ambient temperature range; added  $T_\infty$  indicator; adapted caption accordingly



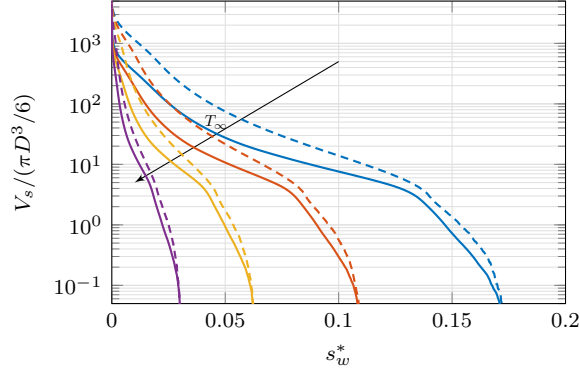


Figure 4: Volume of air where supersaturation w.r.t. liquid exceeds a given threshold as a function of the threshold. The volume is normalized by the volume of the ice particle and four different ambient temperatures are shown:  $T_\infty = -6^\circ\text{C}$  (—),  $T_\infty = -9^\circ\text{C}$  (—),  $T_\infty = -12^\circ\text{C}$  (—),  $T_\infty = -15^\circ\text{C}$  (—). Solid lines correspond to  $Re = 600$  (chaotic regime), while dashed lines show the data obtained for  $Re = 75$  (axisymmetric regime).

Changelog: new figure

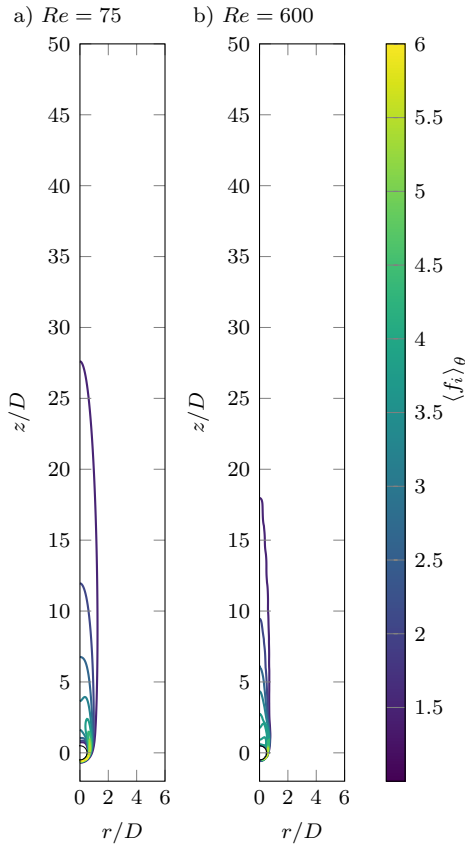


Figure 5: Contours of local ice enhancement factor in the wake, averaged over time and azimuthal direction at  $T_\infty = -15^\circ\text{C}$ . (a) axisymmetric regime at  $Re = 75$ , (b) chaotic regime at  $Re = 600$ .

Changelog: changed ambient temperature to  $T_\infty = -15^\circ\text{C}$ ; contour lines are now linearly spaced; ice enhancement computed according to deposition nucleation law provided by Meyers et al. (1992); adapted caption accordingly

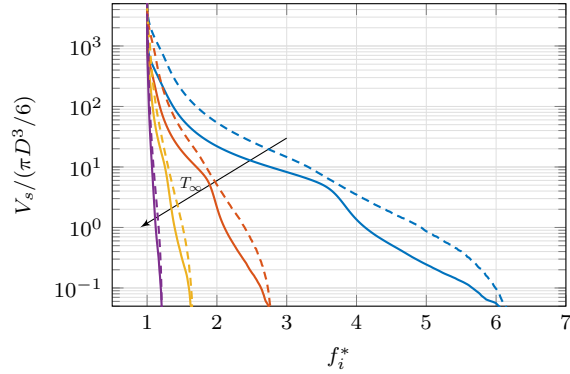


Figure 6: Volume of air with supersaturation above a given threshold as a function of the ice enhancement factor. The volume is normalized by the volume of the ice particle and four different temperatures are shown:  $T_\infty = -6^\circ\text{C}$  (—),  $T_\infty = -9^\circ\text{C}$  (—),  $T_\infty = -12^\circ\text{C}$  (—),  $T_\infty = -15^\circ\text{C}$  (—). Solid lines correspond to  $Re = 600$  (chaotic regime), while dashed lines show the data obtained for  $Re = 75$  (axisymmetric regime).

Changelog: changed ambient temperature range; ice enhancement computed according to deposition nucleation law provided by Meyers et al. (1992); x-axis now linearly spaced; added  $T_\infty$  indicator; adapted caption accordingly

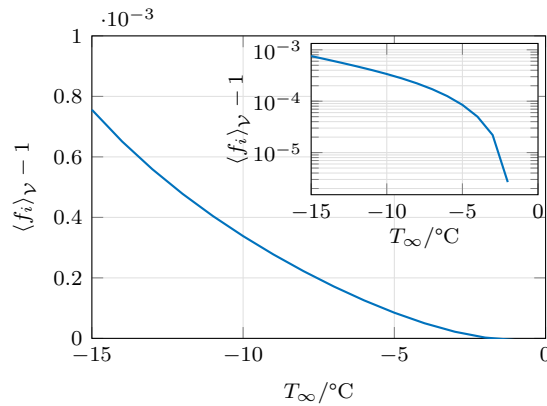


Figure 7: Global ice enhancement factor as a function of cloud temperature. The inset shows the same data, but in semi-logarithmic scale.

Changelog: changed ambient temperature range; ice enhancement computed according to deposition nucleation law provided by Meyers et al. (1992); adapted caption accordingly

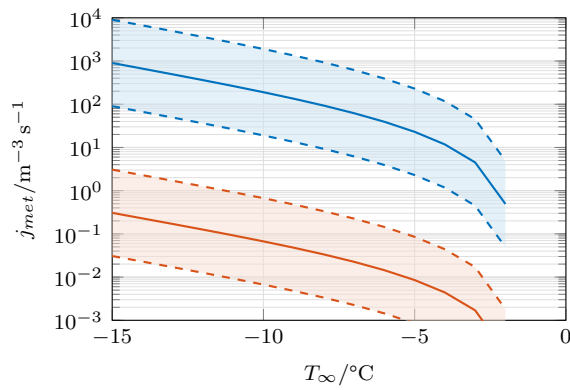


Figure 8: Limiting cases for the nucleation rate  $j_{met}$ . The swept-volume limited estimation based on the considerations of Prabhakaran et al. (2020) is shown for  $C_{expo} = 10$  (—) with the shaded area depicting the values obtained for  $1 < C_{expo} < 100$ . The exposure-time limited estimation, which is directly linked to the ice enhancement factor defined in the manuscript, is shown for  $\tau_{nucl} = 10s$  (—) and the range  $1s < \tau_{nucl} < 100s$  (shaded area).

Changelog: new figure

## References

- Al-Naimi, R. and Saunders, C. P. R.: Measurements of Natural Deposition and Condensation-Freezing Ice Nuclei with a Continuous Flow Chamber, *Atmospheric Environment* (1967), 19, 1871–1882, [https://doi.org/10.1016/0004-6981\(85\)90012-5](https://doi.org/10.1016/0004-6981(85)90012-5), 1985.
- Bagchi, P. and Kottam, K.: Effect of Freestream Isotropic Turbulence on Heat Transfer from a Sphere, *Physics of Fluids*, 20, 073 305, <https://doi.org/10.1063/1.2963138>, 2008.
- Chouippe, A., Krayner, M., Uhlmann, M., Dušek, J., Kiselev, A., and Leisner, T.: Heat and Water Vapor Transfer in the Wake of a Falling Ice Sphere and Its Implication for Secondary Ice Formation in Clouds, *New Journal of Physics*, 21, 043 043, <https://doi.org/10.1088/1367-2630/ab0a94>, 2019.
- Fletcher, N. H.: Size Effect in Heterogeneous Nucleation, *The Journal of Chemical Physics*, 29, 572–576, <https://doi.org/10.1063/1.1744540>, 1958.
- Greenan, B. J. W. and List, R.: Experimental Closure of the Heat and Mass Transfer Theory of Spheroidal Hailstones, *Journal of the Atmospheric Sciences*, 52, 3797–3815, [https://doi.org/10.1175/1520-0469\(1995\)052<3797:ECOTHA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1995)052<3797:ECOTHA>2.0.CO;2), 1995.
- Hoose, C. and Möhler, O.: Heterogeneous Ice Nucleation on Atmospheric Aerosols: A Review of Results from Laboratory Experiments, *Atmospheric Chemistry and Physics*, 12, 9817–9854, <https://doi.org/10.5194/acp-12-9817-2012>, 2012.
- Lehmann, K., Siebert, H., and Shaw, R. A.: Homogeneous and Inhomogeneous Mixing in Cumulus Clouds: Dependence on Local Turbulence Structure, *Journal of the Atmospheric Sciences*, 66, 3641–3659, <https://doi.org/10.1175/2009JAS3012.1>, 2009.
- Meyers, M. P., DeMott, P. J., and Cotton, W. R.: New Primary Ice-Nucleation Parameterizations in an Explicit Cloud Model, *Journal of Applied Meteorology*, 31, 708–721, [https://doi.org/10.1175/1520-0450\(1992\)031<0708:NPINPI>2.0.CO;2](https://doi.org/10.1175/1520-0450(1992)031<0708:NPINPI>2.0.CO;2), 1992.
- Prabhakaran, P., Kinney, G., Cantrell, W., Shaw, R. A., and Bodenschatz, E.: High Supersaturation in the Wake of Falling Hydrometeors: Implications for Cloud Invigoration and Ice Nucleation, *Geophysical Research Letters*, 47, e2020GL088 055, <https://doi.org/10.1029/2020GL088055>, 2020.
- Schaller, R. C. and Fukuta, N.: Ice Nucleation by Aerosol Particles: Experimental Studies Using a Wedge-Shaped Ice Thermal Diffusion Chamber, *Journal of the Atmospheric Sciences*, 36, 1788–1802, [https://doi.org/10.1175/1520-0469\(1979\)036<1788:INBAPE>2.0.CO;2](https://doi.org/10.1175/1520-0469(1979)036<1788:INBAPE>2.0.CO;2), 1979.