

The authors thank Reviewer#2 for this comprehensive review of the paper. We address below each comment individually (in blue color). Line numbers refer to the original manuscript.

My main concern is related to the lack of a proper justification for the proposed parameterizations. The authors base their development on CNT which accuracy for heterogeneous ice nucleation is still matter of debate, although it has been applied before. However the authors make some assumptions that need to be justified. Ice nucleation is assumed to occur mainly in the deposition mode or by immersion in solution. As mentioned by another reviewer only expressions for deposition ice nucleation are used. Moreover, why are these considered the main paths of ice nucleation in the stratiform clouds? Droplet freezing is probably more significant. If not, the authors should show some evidence or at least reports suggesting otherwise. Also, a control simulation where CNT is used but with no acidity dependency considered should be added to discriminate the effect of the later.

We thank Reviewer#2 for this comment, similar to points stressed by Reviewer#1. We copy here the detailed answer to that comment.

Our objective in developing the new parameterization was to represent the formation of ice crystals in the particular conditions of Arctic TIC clouds. In these conditions, it is mainly the deposition mode that occurs for the heterogeneous nucleation of ice, i.e. the air mass is in water-subaturated regime. Kulkarni et al. (2014) showed that, except for quartz, acid-coated dusts are less effective INPs in the deposition mode but have similar effectiveness in the immersion-freezing mode, i.e. in water-supersaturated regime. Based on X-ray diffraction analyses, they argued that acid treatment caused structural deformations of the surface dusts, and the lack of structured order reduced the ice nucleation properties of coated particles in the deposition mode. Moreover, they suggested that, at water-supersaturated conditions, surface chemical reactions might not change the original ice nucleating properties permanently because coating material could be removed by dissolution. For kaolinite, Panda et al. (2010) concluded that sulfuric acid-treated particles could result in the formation of aluminum sulfate that can be easily dissolved in water. Considering these recent findings, and our objective to develop a simplified parameterization to limit computational time, we chose to use the CNT formula for deposition mode but with a specific factor, the neutralization fraction, indicating the degree of acidity of the coating of dust particles. Several passages of the text have been modified to clarify the conditions of the parameterization:

Line 55: “In Keita et al. (2019), the parameterization of Girard et al. (2013) for water-subaturated conditions based upon CNT approach was implemented in the online Weather Research and Forecasting model coupled with chemistry (WRF-Chem) (Grell et al., 2005). This parameterization is suitable to represent the formation of ice clouds in Arctic. It assumes that INPs are mainly mineral dust particles, which is consistent with recent results from the NETCARE (Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments) project (Abbatt et al., 2019).”

Line 66: “In this paper, we investigate for the first time the ice heterogeneous nucleation in a fully coupled aerosol and chemistry parameterization.”

Line 78: “The new scheme for ice crystals formation by heterogeneous nucleation in the deposition mode is implemented...”

Line 153: “Moreover, the condensation-freezing mode, as discussed in Vali et al. (2015), is quite uncertain.”

Line 157: “The new parameterization focuses on the heterogeneous ice nucleation for uncoated INPs and for sulphuric acid coated INPs in the deposition mode i.e. in water-subsaturated conditions. In this approach, INPs are assumed to be mineral dust particles following Girard et al. (2013). For contact freezing and immersion freezing from supercooled cloud droplets, the parameterizations remain unchanged. As condensation-freezing is uncertain (Vali et al., 2015), this process is not longer included in the model.”
Line 207: “For instance, Kulkarni et al. (2014) showed that, except for quartz, acid-coated dusts are less effective INPs in the deposition mode but have similar effectiveness in the immersion-freezing mode, i.e. in water-supersaturated regime. Based on X-ray diffraction analyses, they argued that acid treatment caused structural deformations of the surface dusts, and the lack of structured order reduced the ice nucleation properties of coated particles in the deposition mode. Moreover, they suggested that, at water-supersaturated conditions, surface chemical reactions might not change the original ice nucleating properties permanently because coating material could be removed by dissolution. Panda et al. (2010) concluded that sulfuric acid-treated kaolinite particles could result in the formation of aluminium sulfate that can be easily dissolved in water. Considering these recent findings, and our objective to develop a simplified parameterization to limit computational time, we choose to use the CNT formula for deposition mode but with a specific factor, the neutralization fraction f_n , indicating the degree of acidity of the coating of dust particles.”

Please, note that the immersion freezing of raindrops and cloud water droplets still follows the parameterization of (Bigg, 1953) but is not activated due to the absence of liquid drops in the simulated TIC clouds, except for some few exceptions in the lower part of clouds.

The authors have already performed several control simulations where CNT is used but with no acidity dependency, i.e. with a prescribed contact angle. Those results have been presented in Keita et al (2019) and compared to the same vertical cloud profiles obtained during ISDAC. The simulated vertical profiles of IWC, R_i and N_i found in Keita et al. (2019) for a contact angle of 12° or 26° turn out to be extreme cases of the new profiles described in the current paper. The new parameterization based on prognostic aerosols from WRF-Chem has the ability to distinguish polluted and non polluted air masses in the Arctic and to assess the ice crystal nucleation rate with a contact angle between 12° (clean air mass) and 26° (acidic air mass).

Lines 16-19. Please split this sentence.

Done

Line 26. Should be “specific”.

For the sake of clarity, this paragraph has been thoroughly revisited:

“The detailed process of ice nucleation in cold clouds is complex and remains a major challenge for parameterization in atmospheric models. This is especially the case for polar ice clouds, where the paucity of observations is a serious limitation (Curry et al. 1996; Kanji et al. 2017; McFarquhar et al. 2017). For instance, instead of assuming that cloud particles are distributed homogeneously, to investigate model response and climate sensitivity, some models have based their parameterization on in situ observations (Kay et al., 2016, Cirisan et al, 2020). However, the strong coupling between clouds and state variables, particularly temperature and moisture or relative humidity, requires a dynamic coupling of the cloud microphysics interactively with the atmospheric state variables.

Among these coupling processes, the efficiency of ice nuclei particles (INPs) to activate cloud formation is critical, given the rarity of INPs in the pristine atmosphere. Two approaches are used to treat the INPs efficiency; a singular and deterministic method, or a stochastic method (Pruppacher and Klett, 1997). While the singular approach assumes nucleation to occur at specific relative humidity and temperature (e.g. Wheeler and Bertram 2012; Murray et al. 2012), the stochastic method allows for time-dependent state variables following the classical nucleation theory (CNT) (Pruppacher and Klett, 1997; Cirisan et al, 2020). It is also our approach in this study, where we assume that freezing occurs at any location on the INP surface with equal probability. This is one attempt to represent best in situ observations, yet still not fully physically comprehensive, but one exploration step. The ultimate general method is still a matter of intense research (Vali, 2014; Wright and Petters, 2013).”

Line 29. Remove the comma.

Please refer to the answer to line 26 above.

Line 33. Number density is however a function of temperature.

Please refer to the answer to line 26 above.

Line 34. CNT is not a requirement of the stochastic hypothesis. Please rephrase.

Please refer to the answer to line 26 above.

Lines 36-39. Most atmospheric models use time-independent formulations. In fact, all of these references correspond to time-independent formulations.

There was a typo here. We had written “time-dependent” instead of “time-independent”. This has been removed in the revised version.

Line 41. Please explain the significance of the contact angle. Also isn't this the approach used in this work? A single contact angle, dependent on the acidity?

In the CNT model, a crucial fitting parameter is the contact angle (θ), quantifying the wettability of a solid particle surface by ice via the Young-Dupré equation. It is generally described as a single contact angle for an entire aerosol population, which does not work well for predicting the fractions of INPs on dust aerosol or on particles that have heterogeneous surfaces (Hoose and Möhler, 2012). In this paper, the contact angle is a function of the neutralization fraction, which in turn depends on the variable aerosol composition. It has been precised in the revised version of the manuscript.

Line 45. Say INP (ice nucleating particle) instead of IN.

Done

Line 55. Is dust internally mixed with sulfuric acid?

Yes. In the model description, the MOSAIC module is briefly introduced: MOSAIC uses a sectional approach to represent aerosol size distributions by dividing up the size distribution for each species into several size bins (8 used in this paper) and assumes that the aerosols are internally mixed in each bin.

Line 103. Is this assumption appropriate for small ice particles?

Yes.

Line 133. Why is immersion freezing of cloud droplets (which is likely the dominant path of ice formation) not treated in a more rigorous way?

We thank the reviewer for this comment.

Please refer to the answer to your main concern above.

Line 159, Eq. 13. Is this the total surface area? Shouldn't this equation be weighted by the aerosol size distribution? Also, when applying this to the immersion case, shouldn't it be only valid for the dust particles immersed within the haze aerosol droplets?

Yes, A_d is the total surface area of the aerosol particles. The number concentration of nucleated ice crystals could have been computed per size bin, but it has not been done in this paper. As a consequence, the total number of aerosol particles is used and their total surface area takes into account a weighting by the size distribution. The parameterization is only valid for the deposition mode.

Line 170. This seems wrong. Is it maybe 10^{26} ?

It was indeed a typo. We change 10^{-26} to 10^{26} .

Line 174. The surface tension between ice and vapor is a function of temperature.

Also, this would be invalid for immersion within haze particles.

This is right but the formulation of the parameterization only refers to the deposition mode.

Line 176. This is not the expression for an infinite plane surface. This is in fact the expression for small INP when the size is comparable to the size of the ice germ.

We agree, this is the expression for a curved substrate. It has been corrected.

Line 203, Eq. 19. Is this for the dust particles internally mixed with sulfate and nitrate, or the overall composition? The latter would not seem very rigorous. Please explain.

Yes, in the MOSAIC aerosol module, dust particles are assumed to be internally mixed with sulfate, nitrate and ammonium. The other components of the aerosol composition are not of interest in this study.

Line 215-220. What is the rationale behind the proposed functional forms in Eqs. 20 and 21? Why would the contact angle depend on the acidity?

We thank Reviewer#1 for this comment.

Keita and Girard (2016), after analysing the slope between the nucleation rate and the saturation over ice for TIC1 and TIC2 clouds (cf. Fig. 16 in Keita and Girard (2016)) observed for a given S_i that: (1) the slope is the largest for the smallest accessible contact angles; (2) the decrease of the slope with the increasing contact angle is very non-linear. These results are consistent with laboratory experiments (Sullivan et al., 2010) showing a rapid increase of the contact angle with acidity on coated IN. These results motivated us to parameterize the contact angle θ as a function of the aerosol neutralization fraction under a concave form. Simple concave functions follow power law: $\theta = 26 - 14 \times f_n^p$ with p larger than 1. We have chosen a quadratic ($p=2$, MYKE2 simulation) form for simplicity. We have besides added a sensitivity simulation (MYKE4) under a biquadratic form ($p=4$) for simplicity to test the influence of the exponent p on the concave form of the contact angle with the neutralization fraction.

Line 233. There are no equations 20a and 20b.

We have replaced them by Eq. 20 and Eq. 21.

Line 265. If ice nucleation occurs at cloud top why would it be on haze aerosol instead of cloud droplets immersed with dust?

Please see the response to your main concern above.

Line 285. Is this the total aerosol number for all species?

Yes.

Line 327. This is a confusing sentence? What do the authors mean by the same f ?

We rephrase it by “Results obtained with the MYKE2 and MYKE4 using the same value of the neutralization fraction are very similar.”

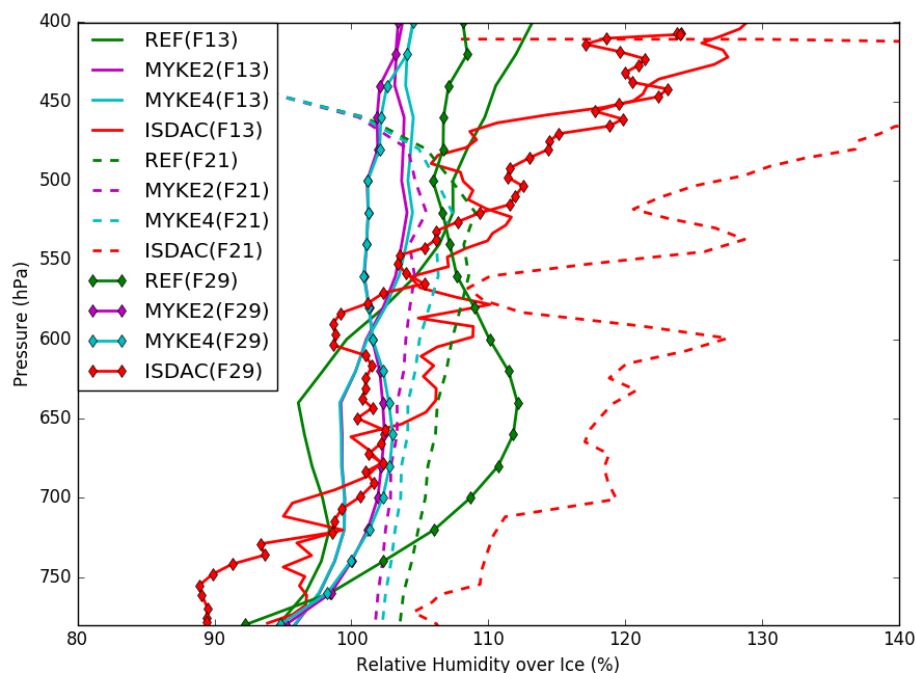
Line 349-350. Can you show this in a plot?

Here we just mention the general behaviour of the nucleation of ice crystals in the CNT as a function of the contact angle. The critical free energy is proportional to the reduction factor $f(\cos\theta)$ (Eq. 15), a monotonic decreasing function of the cosine of the contact angle (Eq. 16). Since the cosine is also a monotonic decreasing function between 0° and 90° , the energy barrier is a monotonic increasing function of the contact angle. As a consequence, a smaller contact angle in the simulation tends to decrease the critical Gibbs free energy to form ice embryos (Eq.15), hence leading to a higher nucleation rate of ice crystals (cf. Fig. 16 in Keita and Girard (2016)) and higher IWC. This explains the differences between MYKE2 and MYKE4.

Lines 384-385. Please show this.

Here is the figure showing the RH_i as a function of altitude (in pressure levels) for the different simulations on the three cases.

As these results were already shown in Keita et al. (2019) (cf. Figure 4), we choose to not show them again in the present manuscript.



Line 402-403. What about using no f , i.e., Just a fixed contact angle?

This has already been done in Keita et al (2019). The current paper presents the big advantage to calculate a contact angle that adjusts to the acidity of the air mass. The spatial and temporal heterogeneities of air masses and ice clouds are thus better represented.

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