

The authors thank Reviewer#1 for this comprehensive review of the paper. We address below each comment individually (in blue color). Line numbers refer to the original manuscript.

The first major comments concern the condition of the paper presentation. Usually this would be part of the minor comments section. However, several presentation errors popped up while reading through the paper, which is the reason why it is already mentioned here. My impression is that the paper was not checked at the end for typos, bracket errors, consistency of parameter notation/writing style (regular vs. italic), description of abbreviations, etc. The authors apologize for those typos and errors in the style. We have carefully corrected all of them.

I will give some examples: Abstract, line 1, 6 and 10: What do the abbreviations TIC, WRF-Chem and IWC stand for?

Those abbreviations had been originally detailed in the text of the paper but, as pointed by Reviewer#1, we have forgotten to repeat them in the Abstract. They have now been clarified in the Abstract as well.

Line 25: It should read “. . . understand fundamental processes. . .”.

Done.

Line 26: Delete one “are” in “. . . which are are particularly. . .”.

Done.

Line 239 and 240: There are no Eqs. (20a) and (20b), only Eqs. (20) and (21).

Done. These equations become Eq. 19 and Eq. 20 in the revised manuscript.

Line 273: There is no Table 5. It should be Table 4.

Done.

Line 352: There is a “10⁻³” too much between -8.1 10⁻³ g/kg. There are more of such errors. Please check the paper thoroughly.

We checked the entire paper for such errors and corrected all of them.

If you use a citation as a constituent part of the sentence (e.g. grammatical subject) then please check your brackets. For example, line 52: It should read “In Keita et al. (2019), the parameterization of Girard et al. (2013) based upon CNT. . .”

Done, we also checked all citations.

Some of the parameters used are not / differently introduced or differently written, etc. Some examples:

N_i vs N_i , S_i vs S_i , f vs f , U vs U , etc. ‘ R_i ’ is not introduced (from the context it is ice particle radius), but in the model description you use diameter.

Physical quantities and variables are now typeset in italic font, as indicated by the recommendations of GMD journal. Abbreviations from 2 letters are typeset in roman font (e.g. Rh_i). Vectors are identified in bold italic font and matrices in bold roman font.

Line 145, N_i has been changed to $N_{m,i}$. R_i stands for the mean ice crystal radius.

Line 368 we have added “the mean ice crystal radius (R_i)”.

Line 201 has been rewritten as: “ The coupling is done by expressing θ as a function of the aerosol neutralization fraction f_n in dust particles internally mixed with sulphate, nitrate and ammonium (Zhang et al., 2007; Fisher et al., 2011), which is between 0 and 1 and is defined as:”

Line 101 -103: Please avoid the brackets around ‘ αx ’, ‘ λx ’, etc.

We suppressed the brackets around all variables in the manuscript.

Please also check your equations. For example Eq. (2): There is the wrong ‘dot’ in the scalar product. And what does ‘TURB’ stand for or in other words how does this term including ‘TURB’ look like?

We disagree with Reviewer#1 as Eq. (2) includes a divergence. We have also added a dot to Eq. (1) according to Milbrandt and Yau (2005a) and Ferrier (1994).

We checked all the equations. From Eq. (4) and (6) we removed (D) from $N_{Tx}(D)$. From Eq. (5) and (4), $dN_x(D)$ has been replaced by $N_x(D)$.

According to Milbrandt and Yau (2005a), Ferrier (1994) and Khvorostyanov and Curry (2014, pages 171-172), the TURB term is:

$$\text{TURB} = \frac{\partial}{\partial x} k_x \frac{\partial}{\partial x} + \frac{\partial}{\partial y} k_y \frac{\partial}{\partial y} + \frac{\partial}{\partial z} k_z \frac{\partial}{\partial z}$$

where k_x , k_y and k_z are the components of the turbulent exchange coefficient. Eq. 1 and Eq. 2 have therefore been rewritten to include the turbulent diffusion matrix.

The second major comments concern the new parameterization and some of the results. You mention on page 6, starting line 151: “The new parameterization focuses on deposition ice nucleation for uncoated IN and to immersion freezing of sulphuric acid coated IN, i.e. IN immersed in an acid aqueous solution.” Here I wonder that for ΔG in the presentation of the nucleation theory you only consider the case of deposition nucleation. If you look into the literature, e.g., Lamb and Verlinde (2010, pages 313-318) you can see that there is a difference between the Gibbs free energies between the case of deposition nucleation and immersion freezing due to e.g. the differences in the interfacial free energies. Finally, you would end up with different nucleation rates even if identical contact angles would be used in the nucleation rate equation. What about the freezing point depression when the particle is immersed in an aqueous solution? Could you please comment on that?

The authors thank Reviewer#1 for these comments.

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 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 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.

Finally, the term “pre-exponential factor” at line166 has been replaced with “kinetic coefficient” in coherence with Fletcher (1958).

Concerning the parameterization of the contact angle: Why do you use the quadratic and biquadratic forms? Eastwood et al. (2008) show the ice nucleation behavior of various minerals and the respective contact angles. Why did you choose the contact angle of kaolinite? Note that Eastwood et al. (2008) used a different (simplified) equation of the reduction factor in contrast to your Eq. (17). What are the consequences when using contact angles based on Eastwood et al.? Have you also considered checking other papers for contact angles? For my impression the contact angles given in Eastwood et al. are smaller compared to other studies of kaolinite (e.g. Welti et al. (2012), with $\theta \approx 90^\circ$ for kaolinite particles in the immersion freezing mode)

The authors thank Reviewer#1 for these comments.

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 INP. 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.

Kaolinite represents a significant component of mineral dust (Glaccum and Prospero, 1980). It is also found to be efficient ice nuclei in the deposition mode, requiring relative humidity with respect to ice (RH_i) below 112% in order to initiate ice crystal formation (Eastwood et al. 2009). This is a typical microphysical condition found in Arctic ice clouds. Recent studies from Kumar et al. (2018; 2019a; 2019b) showed that: (1) the relevance of quartz particles as atmospheric INPs is uncertain; (2) INP activity of dust particles not only depends on their composition but also on their chemical exposure history; (3) the exposition of dust particles to acidic air masses decreases their INP activity. Thus, using kaolinite as a proxy of dust particles in our parameterization is reasonable in the current state of knowledge on dust particles composition in the atmosphere, and in particular in the Arctic atmosphere where our parameterization applies.

All this discussion has been added into the revised version of the paper.

The simplified form of the reduction factor used in Eastwood et al. (2008) is appropriate for their experimental conditions where the radius of the INP is larger than the radius of the ice embryo. Unlike previous studies using the CNT approach (Keita and Girard, 2016; Keita et al., 2019; Girard et al., 2013; Khvorostyanov and Curry, 2009; Morrison et al. 2005; Liu et al., 2007; Hoose et al., 2010; Chen et al., 2008), the INP radius varies within the aerosol module in our parameterization. As a consequence, we choose the general form of the reduction factor from Fletcher (1958) including the effect of the curvature of the INP. As dust particles are mostly in the accumulation and coarse modes of the aerosol size distribution, using the simplified form of the reduction factor in our parameterization might only show small discrepancies in the results. Moreover, a typo error found in Eq. (17) is now corrected as:

“The function $f(\cos\theta)$ is a decreasing function of the cosine of the contact angle θ as defined by Pruppacher and Klett (1997) for a curved substrate:

$$f(\cos\theta) = \frac{1}{2} \left\{ 1 + \left(\frac{1 - q \cos\theta}{\phi} \right)^3 + q^3 \left[2 - 3 \left(\frac{q - \cos\theta}{\phi} \right) + \left(\frac{q - \cos\theta}{\phi} \right)^3 + 3q^2 \cos\theta \left(\frac{q - \cos\theta}{\phi} - 1 \right) \right] \right\}, \quad (17)''$$

Most studies giving values of the contact angle for kaolinite focused on the immersion-freezing mode, i.e. under water-supersaturated regime. This is the case for Welti et al. (2012) for instance. This is why we consider that values of the contact angle found in these studies are irrelevant for our parameterization that concerns the water-subaturated regime.

Concerning the representation of the IWC by MYKE2 and MYKE4: Looking on figure 6, the IWC for F29 is well reproduced by MYKE2 and MYKE4. However, looking on Figs. 7 and 8, which show the vertical distribution of the ice particle number concentration and the ice particle radius (the combination of both at the end leads to IWC), you can see that MYKE2 and MYKE4 overestimate ice particle number concentration and underestimate ice particle radius for F29. Putting these two factors now together lead to a good IWC, however, to my impression just by chance. Actually, for the vertical distribution of the ice particle number concentration alone, REF does a better job. In my view, MYKE2 and MYKE4 are not able to correctly represent the TIC2 microphysical characteristics. Could you please comment on that?

We thank the reviewer for this comment.

For F29 case, no liquid droplets are present inside the simulated cloud for both REF and MYKE. ISDAC observations showed very low liquid water content not exceeding 10^{-3} g/kg with a mean value around 10^{-4} g/kg. Such value cannot explain the observed IWC shown on Fig. 6. Among ice phase microphysical processes, the IWC is determined mainly by the ice nucleation and the solid condensation in a pure ice-phase cloud. The only difference between

REF and MYKE is the parameterization of heterogeneous nucleation of ice by deposition. In both schemes, N_i is first computed and the IWC is deduced assuming the same mass of nucleated ice crystal. As in MYKE, N_i is greater than in REF, the IWC is greater too with values in the same order of magnitude than ISDAC observations. We can deduce that, for F29, MYKE fails to simulate a correct cloud with an overestimation of N_i and an underestimation of R_i and that, even if MYKE could simulate proper value for N_i and R_i , then IWC would be underestimated in comparison with observations. However, the F29 case seems particular in comparison to others TIC clouds observed during ISDAC. In Jouan et al. (2012), where it was analysed based on flight track above Barrow (instead of Fairbanks in Keita et al., 2019 and the present study), it was classified as a TIC1 cloud ($N_i > 10 \text{ L}^{-1}$) with mean N_i of 33 L^{-1} . For F29, Keita et al. (2019) showed a great difference between N_i observed from ISDAC over Fairbanks and N_i deduced from DARDAR observations in the upper part of the cloud (cf. Fig. 13 above 500hPa) whereas it was not the case for F21. Thus it is not clear considering results from Jouan et al. (2012) and Keita et al. (2019) if the cloud corresponding to F29 flight is a TIC1 or a TIC2. Moreover, the order of magnitude of simulated N_i with MYKE for F29 is comparable to N_i deduced from DARDAR.

The discussion of results for F29 lines 365-368 have been rewritten considering the above discussion:

“However, it is reasonably close to satellite observations as analysed by (Keita et al., 2019). Their analysis revealed a large discrepancy of N_i between ISDAC flights and satellite estimations for F29 in the upper part of the cloud. We can notice here that the order of magnitude of N_i for F29 estimated from satellite can question the classification of F29 as a TIC2 especially as Jouan et al. (2012), using flight track above Barrow instead of Fairbanks, classified this cloud as a TIC1. This discrepancy between airborne measurements, simulated results and satellite observations can be due to the small sampling domain taken during ISDAC versus the low resolution of satellite products and of the model grid.”

Following the recommendation of Vali et al. (2015), I would suggest to use “ice nucleating particles (INPs)” instead of “ice nuclei (IN)”.

We agree with the reviewer and we now use INP instead of IN.

The year in the citation “Pruppacher and Klett (1998)” should either be 1997 or 2010. 1998 refers to a review of that book.

Done.

Line 95 and Eq. (4): Why is this equation explained and written in such a complicated way? For me it looks like to simply be density times volume for the hydrometeors considered:
 $m_x(D) = \rho_x V_x = \pi/6 \rho_x D^3$

We agree with the reviewer but we would like to highlight the density approximation used in the MY05 microphysics scheme.

Line 124 -124: You only consider homogeneous ice nucleation of pure supercooled water droplets. What about haze droplets and the resulting freezing point depression?

Indeed, homogeneous ice nucleation is possible for haze droplets but only for high value of S_i . For instance, using Barahona and Nenes (2009) parameterization, at a temperature of -40°C , S_i have to be superior to 1.46 for the homogenous freezing to occur. Vertical profiles of temperatures and relative humidity over ice for the three simulated cases (see Fig. (3) and (4) in Keita et al. (2019)) show that S_i is almost always under this threshold value both for simulated results and observations.

Eqs. (20) and (21) and Fig.1: Could you please make clear in the text, when introducing Eqs. (20) and (21), that Eq. (20) belongs to MYKE2 and Eq. (21) to MYKE4? It is mentioned in Fig. 1 but not in the text at the end of section 2.1.1.

The sentence “Both formulations are implemented in MY05 and tested hereafter.” at Line 217 was changed for:

“Both formulations referred to MYKE2 (Eq. (20)) and MYKE4 (Eq. (21)) are implemented in MY05 and tested hereafter.”

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