

We thank Reviewer 1 for his/her constructive comments. We reproduce reviewer's comments in *blue* and our responses in black.

General Comment:

This is the third manuscript of a series of studies that aimed to understand the ice nucleating abilities of mineral dust particles with an especial focus on feldspars. Part 3 reports the ice nucleating abilities of different aluminosilicate particles in the immersion freezing mode in pure water and aqueous solutions. The authors investigated the effect of surface ion exchange, NH₃ or NH₄⁺ adsorption, and surface degradation on the ice nucleating abilities of kaolinite, sanidine, andesine, muscovite, biotite and gibbsite. The ice nucleating abilities were enhanced in some cases and reduced in others. The increase/decrease of the ice nucleating abilities was found to depend on the type of mineral and also on the exposure time of the surfaces to water and solutes.

This is a very interesting topic of high relevance for the atmospheric chemistry and physics communities. The experiments from the current study were carefully designed and performed. The current results brings our understanding one step forward and it helps the ice nucleation community to better understand why mineral dust particles are good ice nucleating particles. Although the reviewer did not find any major point in terms of the scientific content and the drawn conclusions, the structure, length, and readability of the manuscript needs to be significantly improved before it can be accepted for its publication in ACP.

Major comments:

The manuscript is unnecessary long and different information is repeated several times along the text. This makes the manuscript difficult to read and confusing in some parts.

We have made appropriate changes throughout the manuscript in order to remove the repeated information.

The structure of the manuscript is not the best with too many subsections and mixing information from sections 3 and 4.

We have restructured the manuscript by combining the results and discussion sections in a “*Results and discussion*” section, which is now ordered according to minerals.

The usage of qualitative terms (e.g., strong decrease, remarkable enhancement, strong enhancement) is pronounced and not appropriate.

We have made appropriate changes throughout the manuscript in order to reduce the usage of such terms.

Minor comments:

Abstract: I suggest to reduce its length and to focus on the main results only. Leave out unnecessary or theoretical information for the Introduction section.

The 1st part of the abstract summarizes the results of Part 3 and the 2nd part gives the synopsis of the whole paper series. We would like to keep both parts since it provides a quick summary of the series for the readers. We have shortened the abstract slightly by removing some less important text.

Introduction: It is missing why the authors focused on immersion freezing and what is the importance of mixed-phase clouds. Also, why are mineral dust particles important on a global or regional scale, and why this specific aerosol type is important in comparison to biological or organic particles?

We have added the following statements in the *Introduction* to support the points highlighted by the reviewer.

“Moreover, mixed-phase clouds are responsible for precipitation formation via the Wegener-Bergeron-Findeisen process (Rogers and Yau, 1989; Korolev and Field, 2008).” (Lines 38-39)

“The most important ice nucleation mechanisms in mixed-phase clouds are viewed to be immersion and condensation freezing (Hoose et al., 2008; Ansmann et al., 2009; Twohy et al., 2010; de Boer et al., 2011).” (Lines 50-51)

“The impact of mineral dusts on cloud properties has been shown in several observational and modelling studies (Lohmann and Diehl, 2006; Hoose et al., 2010; Seifert et al., 2010).” (Lines 54-55)

There are too many statements/paragraphs without a proper citation(s).

Specific comments:

Line 38: Add a reference after “properties”.

Sun and Shine (1994); Sassen and Benson (2001) have been added as references. (Line 37)

Line 41: Add a reference after “crystalline ice”.

Vali et al. (2015) has been added as reference. (Line 42)

Line 46: Add a reference after “deposition nucleation”.

Vali et al. (2015) has been added as reference. (Line 47)

Lines 52-53: Either clarify that this studies refer to immersion freezing only or add studies for other ice nucleation modes using feldspar.

Suggested lines have been modified to: “Feldspars have been reported to be the most IN active minerals although the individual members of the feldspar group exhibit very different IN efficiencies in immersion freezing mode.” (Lines 55-57)

Line 56: Add the Kanji et al. ACPD (2018) study.

Kanji et al. (2018) has been added. (Line 60)

Line 58: Add a reference after “humidity”.

Augustin-Bauditz et al. (2014) has been added. (Line 62)

Line 62: Add a reference after “function of aw”.

The references to this statement are mentioned 2 lines below “...INPs can indeed be approximated by such a water-activity-based description (Archuleta et al., 2005; Zobrist et al., 2006; Zobrist et al., 2008; Koop and Zobrist, 2009; Knopf et al., 2011; Knopf and Forrester, 2011; Knopf and Alpert, 2013; Rigg et al., 2013).” (Lines 68-70)

Line 64: Add a reference after “solute”.

Zobrist et al. (2008) has been added. (Line 68)

Line 73: Add “previous” after “our” and before “freezing”.

Added (Line 78)

Line 84: Why immersion freezing? Why is this heterogeneous freezing mode important?

We have added the importance of immersion freezing in the *Introduction* as previously suggested by the reviewer. (Line 50)

Lines 90-101: This paragraph deserves more than a single reference.

Two more references have been added: Brown and Parsons (1989) and Hofmeister and Rossman (1983) (Lines 163, 166)

Line 103: Add a reference after “time”.

This paragraph has been removed.

Line 107: Add a reference after “crust”.

Murray (1991) added. (Line 311)

Line 111: Add a reference after “edges”.

Bibi et al. (2016) added. (Line 316)

Line 129: Add a reference after “dusts”.

Boose et al. (2016) and Kaufmann et al. (2016) added.

Line 167: Add a reference after “1000 particles”.

Kumar et al. (2018a) added. (Line 132)

Lines 192: Given that the authors separated the Results from the Discussion of the Results in different sections, there are several parts which are repeated. It would be ideal to reduce this where possible. I suggest combining the Results and Discussion sections to avoid redundancy along the manuscript and to reduce its length.

We have combined the results and discussion sections in a “*Results and discussion*” section, which is now ordered according to minerals. As suggested by the reviewer, we have also removed less important text to shorten the *Results* and *Discussion* section.

Line 201: Add a reference after “ice”.

Negi and Anand (1985) added. (Line 180)

Line 219, 228: What is strong? I suggest to report this in a more statistical fashion.

We have replaced the whole sentence with: “In contrast to $\text{NH}_3/\text{NH}_4^+$ -solutions, freezing experiments in the presence of Na_2SO_4 as a non- NH_4^+ solute show a decrease in T_{het} below $T_{\text{het}}^{\Delta a_w}(a_w)$ by ~ 1.7 K and ~ 2.4 K for sanidine and andesine, respectively, at $a_w \approx 0.99$. No discernible heterogeneous freezing signal was observed for higher Na_2SO_4 concentrations ($a_w < 0.99$).” (Lines 198-200)

Lines 223-224, 241: “Remarkable enhancement”. Please be more specific.

We have replaced the whole sentence with: “In dilute NH_4^+ -containing solutions, sanidine shows an enhancement in F_{het} up to ~ 0.75 at $a_w \approx 0.99$ compared to the suspension in pure water ($F_{\text{het}} = 0.42$ at $a_w = 1$).” (Lines 202-203)

Line 241: “Strong enhancement”. Please be more specific.

We revised it to: “Interestingly, kaolinite shows a strong enhancement of F_{het} to $0.75 - 1.00$ compared to the pure water case ($F_{\text{het}} = 0.52$) in the presence of NH_3 and NH_4^+ -solute over the complete investigated concentration range ($a_w = 1 - 0.88$).” (Lines 333-335)

Line 293: “because of the high bonding energy involved”. On the other of?

Nash and Marshall (1957) reported that a part of the ammonium ions is firmly fixed to the surface because they could not be exchanged by other cations. However, they did not give any number for the bonding energy. We re-phrase the sentence to follow better Nash and Marshall (1957): “Ammonium ions not only have a strong preference for cation exchange with K-feldspars and (Na-Ca)-feldspars but part of them remain fixed to the surface in non-exchangeable form.” (Lines 244-246)

Line 330: “a thick amorphous surface”. How thick?

We have re-phrased the line to: “...we hypothesize that amorphous surface layers exceeding few nanometers in depth hamper the IN activity of feldspars.” (Lines 280-281)

Lines 383-384: Given that there are many studies on this topic, I suggest to include review papers only where most of previous studies are included (e.g., Hoose and Mohler (2012), Murray et al. (2012), and Kanji et al. (2017)).

Citations have been modified to Hoose and Mohler (2012), Murray et al. (2012) and Kanji et al. (2017). (Line 344)

Line 385: Is this sample only used by ETH research groups?

KGa-1b has been used by several research groups. It is a relatively pure kaolinite provided by Clay Mineral Society and has a low density of crystal lattice defects. We make this clear in the revised manuscript by stating: “... while others used the KGa-1b kaolinite from the Clay Mineral Society with high mineralogical purity (96 %, minor impurities of anatase, crandallite, mica, and illite) (Murray et al., 2011; Pinti et al., 2012; Wex et al., 2014; Kaufmann et al., 2016)”. (Lines 345-347)

Line 385: For consistency also mentioned relevant studies using the KGa-1b sample.

Murray et al. (2011); Pinti et al. (2012); Wex et al. (2014); Kaufmann et al. (2016) added. (Lines 347)

Lines 390-391: Why this should not be the case if both studies used the same sample and the same instrument?

This is indeed what is expected, nevertheless, the IN efficiency could change due to storage over longer time periods depending on storage conditions.

Lines 391-395: I do not get the point or contribution of this paragraph.

We have deleted the suggested part to shorten the manuscript.

Lines 410-444: This is a nice review of recent studies and discoveries on this topic, but this is unnecessary long and I believe that it does not contribute too much to the manuscript. I suggest to summarize this and to focus on the papers that are strictly necessary to explain the present results.

Lines 410 – 433 (lines 360-394 in the revised manuscript) characterize the surface functional groups present on the different kaolinite surfaces. We think that this is relevant information summarized here from studies comprising a broad range of sources, which should become accessible to our community.

Lines 434 – 448 (lines 384-398 in the revised manuscript) summarize molecular dynamics studies carried out with different kaolinite surfaces, which might be deleted but we think that readers that are not interested could also just skip this part.

Line 368: This belongs to section 3 instead to section 4.

Sections 3 and 4 from the previous manuscript are now merged together into one section “*Results and discussion*”.

Line 486: Line 201: Add a reference after “activity”.

Kumar et al. (2018b) added. (Line 434)

Lines 510-549: Unnecessary long. Please be more concise and specific.

We prefer to keep this part since it summarizes surface properties of micas collected from various studies. We think that all available information about mineral surfaces need to be considered as long as it is not clear which are the relevant ones for ice nucleation.

Lines 550-634: This is a super long summary and it repeats many things already mentioned in the Abstract and Section 4. If the authors would like to keep this section focus on the main results/discoveries/conclusions only.

This section is a synopsis of the main findings of Parts 1 – 3 of this series and brings them in a broader context. It shows e.g. that milling is highly relevant for silica but not for feldspars and that the enhancing effect of $\text{NH}_3/\text{NH}_4^+$ pertains to feldspars and clay minerals but not to silica. We think that it is highly relevant to show that results obtained for one mineral should not be generalized to all minerals. We critically went through the whole section and shortened some parts, while we would like to keep most of it.

Figures 2 and 3: Open symbols are too small.

The size of open symbols in Figures 2 and 3 have been increased in the revised manuscript.

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