

We thank Reviewer 1 for his/her thoughtful comments. We reproduce the reviewer's comments in black and our responses in blue. Line numbers refer to the revised, marked up manuscript.

In the presented study the ice nucleation properties of two types of kaolin minerals are investigated, which are chemically identical but have different morphologies. While kaolinite forms flat platelets and has a more constrained ice nucleation behaviour (e.g., freezing onset temperature in the range of 243.3 K to 244 K using freshly prepared samples), halloysite has a variety of different morphologies, e.g. tubes, and shows a more diverse ice nucleation activity with ice onset temperatures ranging from 238.2 K to 244.9 K. To better understand the role of morphology, the samples are milled and reveal a clear decrease in the ice nucleation ability of halloysite, while kaolinite samples are rather unaffected. By determining the pore size distributions and pore volumes of the samples before and after milling, it is shown that the halloysite tubes are destroyed, and thus it is suggested that they are likely involved in ice nucleation processes. The authors provide a detailed discussion about the surface type of the mineral causing the ice formation and conclude that hydroxylated particle edges are the most likely location for ice nucleation.

The study is well conceived and I enjoyed reading the manuscript which is generally very well written. I only have minor comments.

General comments:

Do you have suggestions for further studies to test your hypothesis that the hydroxylated edges of the kaolin minerals are causing the ice nucleation, e.g., molecular dynamics studies, or other laboratory studies?

Molecular dynamics would certainly be an interesting option. Usually, molecular dynamics studies rely on the regular surface lattice of the respective mineral as the starting point for simulations of ice nucleation. The structure of the edges, in contrast, are not well defined, and the specific features that serve as nucleation sites are unknown. Therefore, in a first step, different surface edge structures would need to be defined before their ability to nucleate ice could be assessed.

As direct experimental observation of the nucleation process is not feasible, only circumstantial evidence can be gathered. To this end, the IN activity of still other clay minerals could be assessed and correlated with surface and stacking properties, as we have already done in the recent study by Kumar et al. (2022), where we correlate the IN activity of different montmorillonites with exchangeable cations and stacking thickness. This study is still in discussion in *Atmos. Chem. Phys. Discuss.*

Abstract: You might want to consider mentioning that the milling leads to an increase in specific surface area.

We modify the abstract starting from line 17:

“To interpret these findings, the freezing experiments were complemented by dynamic vapour sorption, BET (Brunauer, Emmett, Teller) surface area measurements, pore ice melting experiments with slurries, and transmission electron microscopy (TEM) before and

after milling. These measurements demonstrate an increase in surface area and the destruction of tubes by milling “

Line 65: An early study by Vonnegut (1947) should be referenced here as well.

Added

Section 2: It might be helpful to include a figure showing the structure of kaolinite and halloysite.

We added a figure that illustrates the kaolinite and halloysite structures as the new Fig. 1 and renumbered the other figures accordingly.

Lines 182 and 197: Shouldn't other studies next to Klumpp et al. (2022) be referenced here as well?

We added additional references on line 177 of the revised manuscript as suggested by the reviewer.

Lines 210 to 211: Is there a reason why those halloysite samples were chosen for milling (e.g., the content of impurities)?

We chose the halloysite samples to cover different morphologies and different DSC curve types of the untreated samples. The amount of halloysite sample available also played a role.

Lines 230 to 232: Can you explain in more detail this equation, and also provide uncertainty estimates for your measurements for pore volume distributions?

For a detailed explanation of the equation, we refer to Kocherbitov and Alfredsson (2007) and their references in the manuscript. Unfortunately, uncertainty quantification is impossible given the different assumptions incorporated in Eq. 1. However, we are confident that for the purpose of comparison between the samples measured and analysed in this study the relative uncertainty is small since the systematic uncertainties are kept constant.

Line 278: The description of the experiments using ammonia/ammonium is missing in the methods.

We added a reference to the experiments performed in ammonia solution by revising the sentence starting on line 185:

“Suspensions of kaolinite and halloysite with 0.2 and 1 wt % in pure water (molecular bioreagent water, Sigma Aldrich) or in 0.2 M ammonia solution (prepared from Merck 25 % ammonia aqueous solution) were prepared and sonicated for 5–10 minutes. “

Line 358 and following: You might want to consider referencing Fig. 2 here.

Following the reviewer, we added the reference to Fig. 2 on line 383 after “240 K” by adding “see Fig. 2”, on line 387 after “~243 K”, and on line 391 after “maxima”.

Section 4.3: I recommend naming this section slightly differently, to indicate that this is a discussion and not a results section (e.g., “likely location of ice nucleation”).

We follow the suggestion of the reviewer and rename the section “Likely location of ice nucleation”.

Figs 7, 8, 11 and 12: Could you indicate the uncertainties in your measurements by error bars?

Following the reviewer, we add error bars for temperatures and F_{het} . Given the high precision of the DVS technique (explained in Section 3) and the difficult treatment of systematic uncertainties we do not indicate error bars for values derived from DVS.

Technical comments:

Line 3: “1” is missing in the authors’ name for their affiliation.

Added.

Figs. 4 and 5: While in Fig. 4 the untreated samples are labeled “pure”, there are not specifically labeled in Fig. 5.

We changed to “untreated” in both cases.

Lines 446 and 448: A bracket is missing at the end of the sentence.

Bracket added.

Reference

Vonnegut, B.: The Nucleation of Ice Formation by Silver Iodide, J. Appl. Phys., 18, 593-595, 10.1063/1.1697813, 1947.