

Response to comments from Referee 1. Referee comments in italics

Current version of manuscript – ACPD version

LE – low exposure

HE – high exposure

Ka – Kaolinite

ATD – Arizona Test Dust

Revised manuscript page/line references in **bold text**.

Interactive comment on “Laboratory studies of immersion and deposition mode ice nucleation of ozone aged mineral dust particles” by Z. A. Kanji et al.

Anonymous Referee #2, Received and published: 7 June 2013

Kanji et al. present new laboratory studies of immersion and deposition ice nucleation of aged mineral dust particles. Chosen proxies for mineral dust are Arizona Test Dust and Kaolinite. The novelty of this work is that particles were aged using various levels of O₃ exposure. Relatively minor changes in freezing temperatures were observed.

This is a timely study that is of interest to the readers of Atmospheric Chemistry and Physics. The methods are sound and the data are analyzed appropriately in terms of measurement uncertainty. The modeling and parameterization is commensurate with our current understanding of ice nucleation and the results are placed into the context of the currently ongoing debate in the IN community. My main concern is the writing style. In this respect, the manuscript needs to be substantially improved prior to publication. The structure convolutes results and discussion, the writing is often repetitive and explores tangential points that do not contribute relatively little value to the main theme of the work. I therefore recommend that the revised manuscript should be significantly shortened. Furthermore, while the results are interesting and important and certainly worthy to be published, my personal interpretation of the raw data is that the shifts are insignificant relative to current measurement uncertainties, and probably also relative to its impacts on the atmospheric aging of dust.

We thank Referee 2 for the positive comments on the manuscript. We have made changes in the revised manuscript to remove repetition and points that are tangential to the main theme of the discussion (See response to Referee 1 comments). In addition we remove Figs 2 and 3. These changes should shorten the manuscript as per your suggestion.

Comments:

I don't understand why so much space is devoted to the O₃ uptake results. It is sufficient to demonstrate briefly that O₃ is taken up and that gamma values are approximately consistent with previous studies. A couple of paragraphs would suffice.

We agree, that a lot of space is dedicated to this section of the work. However, this is the first time we perform such measurements in our laboratory and group, we think it would be best to report how this is done to show that we are able to do this in a reproducible fashion with our new set up. This would also imply that for future planned studies with uptake of trace gasses we can refer to this paper and in fact follow Referee 2's recommendation of describing the uptake in a couple paragraphs.

There is significant overlap between the introduction and discussion the material should be consolidated and presented only once.

In the revised manuscript, we have removed parts of the discussion (section 4) that were repetitive. In addition we remove from the introduction on page 8708 line 25-29 description of the Salam et al. (2008) study since we discuss it later section 4.5. We have also shortened sections 3 and 5 (see comment below).

I question the comparison with field experiments. The closure calculations performed for CRYSTAL-FACE and PACDEX are too poorly constrained to warrant inclusion here. It is encouraging to see that the results broadly make sense in the context of ambient measurements. Nevertheless, closure attempts that use properties from this aging study that are combined with poorly constrained aerosol composition and non-MD contributions to IN are premature. If the authors feel that such closure studies can be attempted they should devote a separate paper to it. Certainly even the most optimistic interpretation of a 3K shift attributed to O₃ ageing is too small to result in meaningful changes in predicted-vs-observed ambient IN comparisons. The results presented here stand well on its own and should focus simply on the observed results rather than making a giant leap to past field campaigns. The resulting statements regarding the parameterization conclusion stated in the abstract needs to be removed.

We have now shortened this section in the revised manuscript and only report one comparison to the field campaign CRYSTAL-FACE to make the relation between laboratory studies and field measurements. We have removed any comparisons from the ageing study to the field campaigns. We therefore remove the comparison of parameterization from the O₃ aged particles to the PACDEX study. We keep the comparison to CRYSTAL-FACE since it is done for the untreated Ka parameterization. We also note that if the parameterization for treated Ka was used to compare to the CRYSTAL-FACE study, we would not get good agreement between (lab) predicted vs. observed IN numbers and this is reported on page **31, lines 20-24**.

Based on the modification of Section 5 (atmospheric implications) we have now adjusted the last two sentences of the abstract in the revised manuscript to read ‘From our results, we present parameterizations in terms of $n_s(T)$ that can be used in models to predict ice nuclei concentrations based on available aerosol surface area’ (**page 2 line 25-27 revised manuscript**).

Comparisons to previous work: this is attempted in two places (Much of Section 3 and Section 4.5). The text isn't very clear how well the quantitative agreement is. Generally, comparing the active fraction as a function of freezing temperature gives (relatively)poor agreement when comparing across different studies, CFDCs, and other IN techniques. A specific example are comparisons between the ATD results from Welti et al., Sullivan et al. and Niedermeier et al. It is something we need to accept to be the case and quantify better. Personally, I don't think it is a big problem for this study as the changes are relative to the same instrument/technique.

We agree, it is difficult to quantify given the varying experimental and aerosol parameters. We have now added specific temperatures and RHs to the comparisons to facilitate more clarity for the comparisons in section 3 and 4.5

It would be helpful though if the comparison to previous results centered around a summary graph. Also the comparisons in the different sections should be consolidated and can be presented more concisely. Since the authors derive active site densities, they can account for size effects (or polydispersion), assuming that density only depends on surface area and that these properties do not systematically change with particle size.

We agree with Referee 2 in principle, but because of the comment above, we know quantitative comparisons on a plot would not yield agreement and be hard to achieve because of the said reasons. A summary graph will just highlight this further which will result in necessitating the same description that is currently the text. The summary graph would also add length to the paper. In addition, a relatively recent paper by Hoose and Möhler (2012) shows this very fact.

We agree with Referee 2, we have now consolidated all the comparisons for the ageing work in Section 4.5 of the revised manuscript. We perform short comparisons of the un-aged aerosol in section 3, since this is not the focus of the work and Ka and ATD ice nucleation has been a subject of numerous IN studies.

I also concur with the excellent points made by the first referee.

We refer Referee 2 to the response to comments from Referee 1

References

Hoose, C., and Möhler, O.: Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments, *Atmos. Chem. Phys.*, 12, 9817-9854, doi:10.5194/acp-12-9817-2012, 2012.

Salam, A., Lesins, G., and Lohmann, U.: Laboratory study of heterogeneous ice nucleation in deposition mode of montmorillonite mineral dust particles aged with ammonia, sulfur dioxide, and ozone at polluted atmospheric concentrations, *Air Qual., Atmos. & Health*, 1, doi: 10.1007/s11869-11008-10019-11866, 2008.