

Anonymous Referee #1, 02 Feb 2025

This is an excellent paper. I have some items the authors may wish to consider, but there are no glaring errors or omissions.

We thank referee 1 for the very positive feedback. We will address all comments in the following. The referee comments are in black, our response is given in blue color. Changes in the manuscript text will be highlighted.

The authors cite books in several places. I'll use Seinfeld and Pandis as an example. That book is over 700 pages, and covers a wide variety of topics. Please, at least cite a chapter in the book. For example, in line 38 when you cite Seinfeld and Pandis for deliquescence, point to the paragraph or section in the book that is relevant for this.

We followed the reviewer's suggestion and added book chapters and pages accordingly:

For the general citations about deliquescence, the following changes have been made:

lines 28, 35, 38-39: Khvorostyanov and Curry (2014) → Khvorostyanov and Curry (2014, Chapter 11, pp. 547-575),

lines 35, 38-39: Seinfeld and Pandis (2006) → Seinfeld and Pandis (2006, Chapter 10.2, pp. 449-461),

lines 38-39: Hellmuth et al. (2013) → Hellmuth et al. (2013, Chapter 12, pp. 317-347)

For the temperature dependence of the deliquescence RH, the following changes have been made:

lines 29, 53, 54, 224: Seinfeld and Pandis (2006) → Seinfeld and Pandis (2006, Chapter 10.2.2, pp. 453-454)

lines 29, 54: Khvorostyanov and Curry (2014) → Khvorostyanov and Curry (2014, Chapter 11.4.3, pp. 562-563)

For the theoretical description of deliquescence, the following changes have been made:

lines 284, 336, 337: Khvorostyanov and Curry (2014) → Khvorostyanov and Curry (2014, Chapter 11.3, pp. 553-558)

line 336: Hellmuth et al. (2013) → Hellmuth et al. (2013, Chapter 12.2, pp. 319-334)

Line 336: Lamb and Verlinde (2011) → Lamb and Verlinde (2011, Chapter 7.1, pp. 290-295)

The section on particle generation, size selection, and pre-conditioning is comprehensive, but I think there needs to be some mention of doubly charged particles, because a DMA is used for size selection. Is the size used such that you don't have to worry about larger particles being present in the sample? Maybe having larger particles in the sample is not so important because you are considering deliquescence, not activation? A sentence explaining how the possibility of larger, doubly charged particles affect the results of the paper would be appreciated.

Due to the use of the DMA, multiply charged particles can be present. There are two reasons why we chose particles with a mobility diameter of 400nm. First of all, these particles can be clearly detected by the welas 2300 sensor. Secondly, the amount of doubly charged particles is very low. Based on the particle size distribution resulting from the used atomizer, the amount of doubly charged particles is less than 1%. Actually, looking at Fig. 3 in the manuscript, it can be seen that there is a very small second mode for the deliquesced particles at about $D_p = 1.3\mu\text{m}$. Calculating the corresponding dry, solid NaCl particle size by means of the Köhler equation ($RH_{\text{mean}} = 72.5\%$), yields a mass equivalent diameter of about 680 nm. This agrees well with the mass equivalent diameter of the doubly charged

particles, being about 650 nm. In conclusion, we argue that larger, doubly charged particles do not affect our results.

The following sentence has been added to the text: “For the experiments, we selected a mobility diameter of $D_{p_mob} = 400$ nm in order to be able to detect dry solid particles optically by means of a Promo 2000 with welas 2300 aerosol spectrometer (Palas GmbH, Karlsruhe, Germany) inside LACIS-T, **as well as to minimize the amount of larger, doubly charged particles being present. As it turns out, their proportion to the total number of selected particles is less than 1%, i.e., doubly charged particles do not affect our results.**”

Line 187: I know this is picky... “About 78.000...” That’s five significant figures. I recommend “78”.

We made a mistake here. It should read seventy-eight thousand particles. So, the dot as a separator is wrong. It was changed accordingly to 78000.

I was particularly interested in the discussion of the time for deliquescence in the appendix. I think deliquescence is a nucleated phase transition, as noted by the authors when they cite Khvorostyanov and Curry. See also Lu et al (2008) and Cantrell et al (2002). I am not convinced that the authors are seeing evidence for nucleation in their experiments though. I would expect salt particles resulting from efflorescence of an atomized solution to be defect rich, which would lower the nucleation barrier to a value that I doubt you would detect it in these experiments.

The referee raised a valuable point. We agree that we are not able to observe/resolve the nucleation process itself in our lab experiments. However, this is also not the focus of our study. For us, it is important whether the NaCl particles are solid or deliquesced, and that we are able to distinguish between both phase states.

The model study was performed to test whether or not we are sensitive enough to observe the effects of turbulent RH fluctuations on the number of deliquesced particles. With the – probably too simplified – assumption of a constant time scale of deliquescence, we found a qualitative agreement between lab observations and model results. A Quantitative agreement could not be achieved. A possible explanation for this missing agreement could be the too simple treatment of deliquescence in the model. However, testing the applicability of different deliquescence models is not in the focus of our paper, but could be the aim of future studies. This is already mentioned in the original manuscript. However, we added a sentence at the beginning of Appendix B:

“The model study was performed to test whether or not we are sensitive enough to observe the effects of turbulent RH fluctuations on the fraction of deliquesced particles.”;

as well as a sentence at the end of Appendix B: **“However, it should be noted that we were not able to observe / resolve the nucleation process itself in our laboratory experiments.”**

References:

Cantrell, W., McCrory, C. and Ewing, G.E., 2002. Nucleated deliquescence of salt. The Journal of chemical physics, 116(5), pp.2116-2120.

Lu, P.D., He, T. and Zhang, Y.H., 2008. Relative humidity anneal effect on hygroscopicity of aerosol particles studied by rapid-scan FTIR-ATR spectroscopy. Geophysical research letters, 35(20).