Answer to Anonymous Referee #2

Thank you for your positive evaluation of our article and the helpful comments. Below we address your individual comments and describe the associated changes (in bold) that have been made in the revised manuscript version.

Main comments:

This is an overall well written and structured publication on the measurements of particle shapes, infrared extinction spectra of nitric acid dehydrates (NAD) and optical constants of the metastable form β-NAD in the AIDA aerosol and cloud chamber. The paper is well structured and the main results on the optical constants for β-NAD are especially important for the remote sensing measurements and the characterisation and retrievals of particle size distributions of polar stratospheric clouds (PSCs).

The study is well suited for Atmospheric Chemistry and Physics, but due to its large number of details in laboratory measurements it would even better fit – to my mind - to the sister Journal Atmospheric Measurement Techniques.

We can understand this train of thought, but would still like to promote a publication in Atmospheric Chemistry and Physics (ACP). One of our earlier articles on the optical constants of ammonium nitrate was indeed transferred to Atmospheric Measurement Techniques (AMT) (https://amt.copernicus.org/articles/14/1977/2021/). This decision was understandable because that article focused on the procedure for determining the optical constants, while the underlying experiments on the formation of ammonium nitrate had already been described in another article. In our opinion, this is clearly different in the present work. Here we do not only describe the retrieval procedure, but present a set of novel laboratory experiments on the immersion freezing of HNO_3/H_2O solution droplets that led to a surprising result, namely the rapid growth of the β-NAD phase. We also provide a detailed literature review of previous studies on NAD/NAT nucleation and an outlook on future experiments planned in our laboratory. From our point of view, the article therefore goes beyond the scope of AMT, which deals with the development, comparison and validation of measurement instruments, and ACP is a much better fit.

The only caveat with the presented study is the open question why the authors have taken so much effort to measure and characterise a PSC particle type in the laboratory so far not measured in the real atmosphere. The authors have presented a discussion addressing this critical question at the end of the manuscript, where they very nicely outline how the 'next' measurements on NAT particles with the AIDA chamber should look like. Hopefully, these kind of measurements will be successful and are coming soon. It might be helpful for the reader to include one summarizing sentence in the abstract and/or introduction section to highlight the importance of NAD lab measurements even though NAT are so far the only measured nitric acid hydrates particles in the atmosphere.

This is a good suggestion. In the introduction we addressed the importance of metastable states with reference to Ostwald's step rule and the potentially lower nucleation barrier of NAD compared to NAT, but we failed to do so in the abstract. We propose to add a summarising statement at the end of the abstract as follows:

“While direct evidence for the existence of metastable NAD in the polar stratosphere is still lacking, our experiments add to the wealth of previous laboratory studies that have identified various conditions for the rapid growth of metastable compositions. In the atmosphere, these could be intermediate states that transform into thermodynamically stable NAT on longer time scales in aged PSCs.”

Minor comments:

At some place in the manuscript the authors should state that the PSD parameter (e.g. effective radius and or bimodality) are also an important factor for the exact location of the spectral features PSC particles (e.g. Kalicinsky et al. (2021) showed changes in the 820 cm-1 NAT feature in direction to a step function for observations under NH polar vortex conditions with radiative transport calculations without taken aspherical particle into account).

Thank you for pointing out this study, which of course deserves to be mentioned in the introduction. We added the information in line 91:

“Note that the particle size distribution parameters of β-NAT can also change the spectral features in the range from 810 to 820 cm^{-1}, even if the simulations are limited to spherical particles (Kalicinsky et al., 2021). Increasing the median diameter transforms the 820 cm^{-1} peak into a shifted peak and further into a step-like signature (Kalicinsky et al., 2021). But only highly aspherical particle shapes could also explain the unexpectedly large optical diameters (in some cases larger than 20 μm) of HNO_3-containing particles detected with aircraft-borne optical in situ instruments (forward scattering and optical array imaging probes) (Molleker et al., 2014).”

L396ff: Here the authors present quite a number of parameters and numbers, maybe a table summarizing the details would be feasible.

Yes, we have inserted a reference to a new table in line 367 in which we have summarised the start parameters of the homogeneous and the heterogeneous nucleation experiment shown in Fig. 4:

“Relevant start parameters of the two nucleation experiments are summarised in Table 1.”

Table 1: Start parameters of the homogeneous α-NAD and the heterogeneous β-NAD nucleation experiment shown in Fig. 4. AIDA temperature (*T*), pressure (*p*), and number concentration of added illite particles (N_{illite}) were directly measured. Mass concentration (m_{NA}) and composition ($wt\%_{NA}$) of the HNO_3/H_2O solution droplets were retrieved from the FTIR spectra. The log-normal parameters of the size distribution of the HNO_3/H_2O solution droplets (N_{NA}, σ_{NA}, CMD_{NA}) were constrained using the welas OPC measurements.

Exp. type	T (K)	p (hPa)	N_{NA} (cm^{-3})	σ_{NA}	CMD_{NA} (μm)	m_{NA} (mg/m^3)	$wt\%_{NA}$	N_{illite} (cm^{-3})
Hom (α-NAD)	190.7	994	25000	1.2	0.5	2.7	50	–
Het (β-NAD)	190.3	1012	20000	1.2	0.5	2.2	53	100

L634: Are not Höpfner et al. (2002) and Spang and Remedios (2003) the first publications regarding the NAT identification at 820 cm^{-1} and should be listed here first? Same in the introduction L41, where only Spang and Remedios is mentioned but Höpfner et al. is missing.

Thanks for pointing this out. We will add Höpfner et al. (2002) to line 41 and both Höpfner et al. (2002) and Spang and Remedios (2003) to line 634.

Technical details: To my mind the 800-900 cm^{-1} wavenumber range is not very well presented in many figures (6,8,9). It is too tiny, the details are not visible. I have seen this in many publications and is caused by the presentation of a typically huge wave number range. This specific region is of special interest for the remote sensing community due to the characteristic features for nitric acid trihydrate (NAT) at 820 cm^{-1} and the shift of this position and changes in the form of the feature due to various reasons (Woiwode et al. 2016, 2019, Kalicinsky et al. 2021). Maybe the separation in two wavelength region and/or an additional zoom, like in Figure 5 (still very small), for the Fig. 6,8, and 9 would help to show some more details here.

This is a good suggestion. With the spectra for α-NAD shown in Fig. 6, we deliberately wanted to focus on the range of the nitrate doublet between 1500 and 1100 cm^{-1}, where the shape-dependent variations are most obvious. However, for the new spectral measurements and retrievals of β-NAD, we agree that it is extremely useful to better represent the wavenumber range between 900 and 800 cm^{-1}. Since the best-fitted spectra shown in Fig. 9 make direct use of the optical constants shown in Fig. 8, it seems redundant to add a zoom to both figures. Therefore, we have concentrated on Fig. 9 and added separate panels for the range from 900 to 800 cm^{-1} for all three case studies.

New Fig. 9:

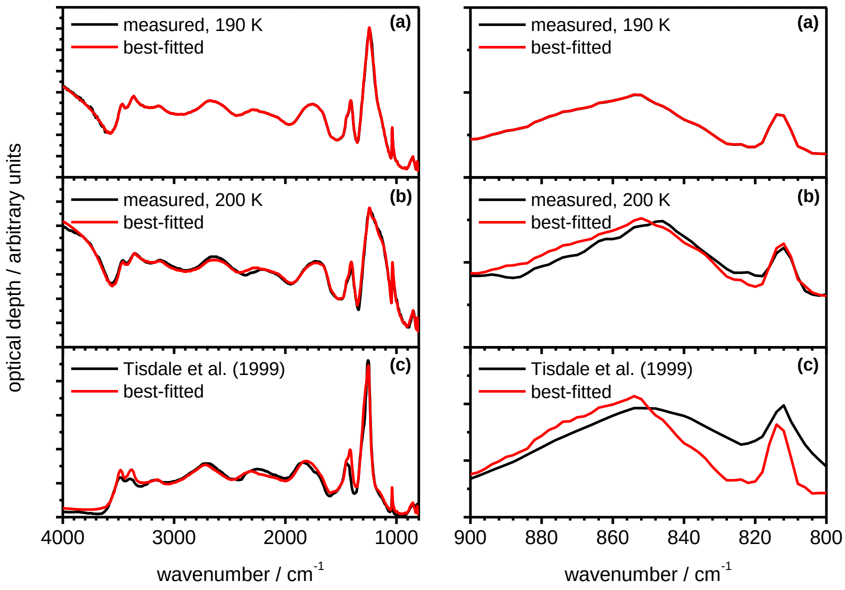

Fig. 6+7: The SID-3 measurements/images are very dark and the diffraction patterns are not very well to see. Can you brighten the images?

Yes, we will try to brighten the images a bit and also pay special attention to the quality of these figures during the production process.