Referee Report on « Particle shapes and infrared extinction spectra of nitric acid dihydrate crystals: Optical constants of the β-NAD modification" authored by R Wagner, A.D. James, V.L. Frankland, O. Möhler, B.J. Murray, J.M. Plane, H. Saathoff, R. Weigel and M. Schnaiter submitted to Atmospheric Physics and Chemistry as manuscript acp-2023-28

This paper deals with the formation and characterization of a thermodynamically stable polymorph of nitric acid dihydrate using a multidiagnostic approach within the well-known large coolable aerosol chamber (AIDA) equipped with various detection techniques including high-resolution 2D imaging of submicron and supermicron-sized ice particles in the presence of HNO_3. This work is convincing, well-done with many quality control benchmarks and informative. The report is well organized, easy to follow, well written and represent a good compromise between sufficient detail and flow of presentation. The new aspect of this work, apart from the surprising discovery of fast growth conditions of β-NAD by heterogeneous nucleation in the presence of proxies of meteoric smoke aerosol particles, resides in the detailed analysis of the scattering properties of resulting solid α- and β-NAD aerosol particles resulting in a marked crystal shape of the two polymorphs. Time will tell whether or not the shapes of elongated needles (α-NAD) or compact spheroids (β-NAD) will explain the atmospheric lifetime or residence time of these aerosol particles in future real observations of given strata within the atmosphere in case β-NAD particles occur at all in nature. I propose the publication of this work in acp once my sparse comments will have triggered a suitable response by the authors.

In what follows I will submit the following comments/questions and remarks concerning the submitted manuscript in the hope to provide a line along which the authors may make changes to the manuscript:

- The displayed FTIR absorption spectra in Figures 5, 6, 8, 9, 10 and 11 are important spectral observables representing key elements in support of α- and β-NAD. However, in comparison to recent spectra, for instance by Iannarelli and Rossi (2015) recorded at nominally identical spectral resolution of 4 cm^{-1} the present spectra show relatively few details in comparison. Have the present FTIR spectra been smoothed in order to suppress small albeit potentially important details? I am aware that recording conditions (T and/or growth conditions, particle size distribution functions, scattering properties, etc.) may lead to minor frequency shifts and small differences in spectral appearance, but what are the reasons for the apparent lack of spectral details in the FTIR spectra? In cases where several FTIR spectra are shown in a stacked manner (for instance in Figure 5 or 6) it is unclear what the displacement of every superimposed spectrum (spectra b to e) is in terms of optical depth or absorption compared to the lowest. I assume that the labelled scale only applies to the lowest displayed absorption spectrum.
- Compared to the referenced precursor studies the present work clearly starts out with liquid droplets of HNO_3/H_2O aerosol, and it is this starting condition that enables the unambiguous observation of β-NAD when meteoric smoke proxies (Illite, $MgFeSiO_4$) are used as seed crystals, otherwise α-NAD is observed for homogeneous freezing without ever ending up as stable β-NAD. This case corresponds to an immersion freezing event triggered by specific (solid) seed aerosol. On the other hand, we and others have exclusively observed the formation of α-NAD in case gas-phase HNO_3 is deposited on a macroscopic ice surface without ever observing the conversion from α- to β-NAD. In this case the mechanism might be a case of condensation freezing on PSC II particles that are less prevalent compared to PSC Ia and Ib clouds. Even though the authors state this fact at the end of the article this difference seems to be important enough to alert the reader early on in the report. In

addition, the bifurcation between NAT and NAD depends on the partial pressure (activity) of HNO_3 at constant partial pressure of H_2O vapor (Iannarelli and Rossi, 2015): by doubling the flow rate (concentration) of HNO_3 we obtain NAD at the expense of α-NAT that converts to β-NAT with increasing temperature. It thus very well may be that NAD (including β-NAD) may never be accessed depending on the freezing mechanism. Relevant experiments on ice aerosol substrates may be performed in the AIDA chamber in order to test several growth mechanisms, even if the partial pressure of H_2O may be increase somewhat to artificially produce PSC II cloud particles.

- My guess is that both OPC and SID-3 instruments were exposed to low temperatures. Did the authors encounter any temperature problems or other anomalies (calibration) in the vicinity of the measurement ports (inlet or optics)?
- On line 350 what is the definition of the saturation ratio S_{NAD}? Ratio of the activities of HNO_3 to H_2O water vapor? Please include the expression into the manuscript by mentioning why this was a calculated and not a measured parameter.
- What is the significance of the magenta window in the right and left hand panels of Figure 4: is it to highlight the change in time duration for reaching the peak of rh and the concomitant onset of aerosol particle nucleation?
- The linear depolarization results discussed in lines 491-497 are hard to rationalize in terms of differences of the shapes for α- and β-NAD. What is the physical reason for the three times larger depolarization ratio of β-NAD vs. α-NAD despite the more needle-like shape of the latter? This is somewhat counter-intuitive despite the invoked reference of Zhakarova and Mishchenko (2000) and may have far-reaching consequences for the interpretation of LIDAR atmospheric backscattering signals.