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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Thermodynamics of homogeneous nucleation of ice particles in the polar summer mesosphere" *by* A. Y. Zasetsky et al.

Anonymous Referee #2

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1 General Remarks

In the current manuscript the authors propose a new nucleation pathway for the formation of mesospheric ice particles known as noctilucent clouds (NLC) or polar mesospheric clouds (PMC). While the bulk of previous model studies of these particles has assumed heterogeneous nucleation on meteor smoke particles, the authors here speculate about homogeneous nucleation via a pathway starting at the formation of critical amorphous clusters via ice particles with a cubic crystalline structure and finally ending in ice particles with a hexagonal crystalline structure. This scenario is at least qualitatively attractive since the proposed step-wise nucleation scheme implies a reduced energy barrier as compared to classical nucleation theory and hence significantly en-

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hanced nucleation rates.

While the authors' proposal is principally very interesting, in its current form the manuscript is unfortunately largely superficial and also mixes theoretical estimates with data presentation from satellite observations which are either not of sufficient quality or simply presented out of context.

As such, I feel that major revisions are necessary to make this manuscript eligible for publication in Atmos. Chem. Phys. In particular, I recommend to remove the presentation of the satellite data and focus on an in-depth presentation of the theoretical concept of the nucleation mechanism. A publishable presentation of the latter needs much more detail than is presented here.

2 Specific Comments

1) In section 2, the authors attempt to estimate particle number densities from measurements with ACE-FTS assuming a fixed particle radius of 60 nm. Clearly, this procedure is not appropriate and subject to severe errors because of several reasons. First of all, the authors should note that it is well known from ground based and satellite observations that radii of NLC particles show a strong variation from say 20 nm to 120 nm (e.g., *Baumgarten et al.*, 2008; *Karlsson and Rapp*, 2006; *von Savigny and Burrows*, 2007). Hence, using a fixed radius of 60 nm could in the worst case result in an over-estimate (under-estimate) by a factor of 3 (2) and hence a misinterpretation of the particle number density by a factor of 27 (8) since the signal by ACE-FTS is proportional to ice volume. Secondly, any optical observation such as from ACE-FTS or Odin-OSIRIS or any other optical instrument can only provide information on the number density of the 'visible' part of the particle size distribution. Since optical measurements depend on the third (ACE-FTS) to the sixth power (any instrument depending on light scattering rather than absorption) of the particle radius it is obvious

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that any particle number density retrieval will result in a severe under-estimate of the true number density since the smallest particles in the size distribution will basically not make any contribution to the optical signal. For the issue of nucleation and subsequent ice microphysics, however, it is really the total ice particle number density which is important since all nucleated particles will compete for the available water vapor. The total number density of ice particles, however, can only be obtained making use of in-situ techniques which rely, e.g., on measuring charges carried by the ice particles (e.g., *Havnes et al.*, 1996; *Mitchell et al.*, 2001). In line with my arguments above, such measurements typically imply ice number densities of order 10^3-10^4 particles/cm^3 rather than the few tens to hundred/cm^3 implied in this manuscript.

2) I further note that using Odin/OSIRIS brightness measurements as a proxy for particle number density is impossible because brightness varies as number density times particle radius to the sixth power. Hence, minute changes of particle radius will result in dramatic changes of cloud brightness. Consequently, the apparent match of the two distributions shown in Figure 1 rather raise questions about the data sets. In any case, this comparison cannot demonstrate that the ACE-FTS observations are of high quality (which I don't doubt), but they show completely different things.

3) The authors further use ACE-FTS temperature and water vapor observations in order to estimate saturation ratios in the summer mesopause environment. The authors should note that even with the very optimistic error estimates of $\pm$8 K for temperature and $\pm$10 $\%$ for water vapor, they end up with an error of about 300 $\%$ for the resulting saturation ratio. More to that, for a fixed temperature, the data shown in Figure 5 imply a variability in S by more than 2 orders of magnitude. Do the authors interpret this as real variability in water vapor or are these rather statistical fluctuations of the data? In any case, the presentation of these data needs a much more in-depth discussion than is presented here.

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4) In section 4, the authors refer to their own unpublished work regarding molecular dynamics simulations of water clusters under mesospheric conditions. This is not acceptable. It is just these simulations which are critical for the contents of this manuscript. A detailed description of the simulations must be given here to give the reader the chance to judge about the basis of the presented results. This description should also contain a detailed discussion of the initial conditions of the simulations presented here. In the current version, the authors state that 'spherical liquid particles were then cut out and placed in a large simulation box'. As I understand it with the limited information which is supplied at this point, this seems to imply that the simulations start at a point where liquid drops have already formed. This is really confusing since I thought that the initial formation of such particles/droplets from the gas phase is just what is to be studied. In any case, the authors should add significantly more information to avoid any such confusion.

5) When discussing equation 1, recommend that the authors quantitatively compare their results to corresponding results from CNT. Also, I strongly urge the authors to add detailed calculations of nucleation rates and not just include relative order of magnitude comparisons which are difficult to follow. In the end, the important question is really whether the here proposed mechanism can quantitatively explain observed particle number densities or if it can only make a small contribution.

6) In their discussion of the new nucleation pathway, the authors repeatedly refer to laboratory data from Devlin and co-workers. While I am convinced that these are excellent lab-experiments, I wonder whether the findings from these papers can be applied to the conditions of particle formation at the polar summer mesopause. The ice clusters studied by Devlin et al. were created by an expansion of supersaturated air into a vacuum. Hence, ice nano-particles form on very short time scales (milliseconds)

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under extreme thermodynamic conditions with large super-saturations. In the polar mesosphere, on the other hand, we may rather assume that formation times are very different, presumably significantly longer than in the lab experiments. A corresponding discussion should be added to the text.

7) When discussing equations 1 and 4, the authors should also clearly point out that they are using macroscopic properties like surface tensions for the description of microscopic entities. A critical discussion about the validity of this concept would be helpful.

8) In section 6, the authors speculate about a cubic-hexagonal transition. They should note, however, that estimates by *Murphy* (2003) imply that the transition time is far to long to be of any relevance for mesospheric conditions. A corresponding short discussion should be added. In the same direction, the authors should also correct their statement that *Rapp and Thomas* (2006) were assuming an ice formation from saturated water vapor to crystalline hexagonal ice. Rather, these authors refer to Murphys's studies and only consider cubic ice.

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