

# ***Interactive comment on “Widespread polar stratospheric ice clouds in the 2015/2016 Arctic winter – Implications for ice nucleation” by Christiane Voigt et al.***

## **Anonymous Referee #2**

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The manuscript by Christiane Voigt and co-authors is based on a unique data set on polar stratospheric clouds measured with the WALES lidar instrument onboard the High Altitude and Long Range Research Aircraft HALO. The measurements were performed during the winter 2015/16 with exceptionally low temperatures in the Arctic stratosphere and the formation of a widespread polar stratospheric cloud layer with clearly distinct areas of mostly supercooled ternary solution (STS), nitric acid hydrate or ice particles. The highly resolved WALES measurements along the aircraft flight path on several days were combined with lidar measurements from space obtained with the CALIOP instrument onboard the CALIPSO satellite. The information on the PSC types were obtained from a combined analysis of the lidar backscatter ratio and the depolarisation

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at 532 nm.

This is an excellent data set which provides new and important insight into the structure of PSC clouds and allows for conclusions on PSC microphysical processes which still are still uncertain or unknown in many respects. The manuscript includes new results on an important topic and therefore is appropriate for publication in ACP. However, I agree to reviewer 1 that the microphysical part is very weak and that the manuscript needs major revision before acceptance to ACP. My major concerns and questions are the following:

The authors argue at the beginning of section 6.2 that there are “two branches in the ice regime linked directly to STS or NAT/mix2 regimes respectively as shown in Figure 4. . .”. Later in the discussion of Figure 5 it is shown that the latter is also spatially located to the NAT/mix2 regime. This is a very nice result, but the spatial collocation alone does not necessarily point to the role of the solid nitric acid hydrates for the nucleation of ice observed in these areas. Also the trajectory analysis shown in Fig. 7 is not really conclusive in this direction because the temperature history of trajectories 1 to 4 is almost identical after the temperature dropped below the ice frost point temperature  $T_{ice}$ . The only difference is that cases 1-3 reached temperatures above  $T_{NAT}$  where a pure STS cloud is formed because NAT particles disappeared at these conditions, whereas for case 4 the temperature stayed below  $T_{NAT}$ . But now one may ask what happens to the STS particles when the temperature of an STS regime drops below  $T_{NAT}$  and nitric acid hydrates start to nucleate? Will all STS particles nucleate? This will probably not be the case even if homogeneous nucleation occurs. The authors argue that NAT is formed by heterogeneous nucleation on meteor smoke particles. This should be an even more a selective process, means most of the STS particles remain in the supercooled liquid state, either as STS or later on, depending on how fast the nitric acid particles grow in expense of the nitric acid content of the STS particle reservoir, may convert into mostly binary sulphuric acid solution particles. But what about their role in forming ice after the temperature drops below  $T_{ice}$ ? They may also

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be the source for the ice observe in these locations.

In general, the manuscript lacks of more solid microphysical arguments and a balanced discussion of microphysical processes. For instance, it is not just the temperature difference to the frost point temperature that drives or quantifies the nucleation and crystallization of ice. Many laboratory studies formulate homogeneous and heterogeneous ice nucleation as a function of the ice supersaturation or saturation ratio. In the discussion of ice nucleation modes, the temperature difference to  $T_{ice}$  should also be expressed as an ice saturation ratio. Also the supersaturation thresholds for heterogeneous and homogeneous ice and NAT nucleation should be discussed if available and reference to respective laboratory work should be given. Sentences like in line 32 of page 7 (“As soon as temperatures decrease below  $T_{ice}$  . . . ice PSCs are present . . .”) are misleading because also heterogeneous ice formation usually requires some supersaturated conditions to occur at significant rates.

The manuscript is not very clear in defining and using terms like “STS regime”, “NAT/mix2 regime”, “NAT regime”. For instance, is there a “NAT regime” in PSCs? I recommend to stay with the term “NAT/mix2” throughout the manuscript, or is there independent evidence that the PCS discussed in this manuscript included pure NAT clouds?

And what about the question of NAT vs. NAD? It is mentioned in the manuscript, but how does the temperature in the PSC layers compare to  $T_{NAD}$ ? I recommend to also include  $T_{NAD}$  in Figures 1 and 7 and to keep the discussion neutral to the formation of the actual hydrate form, unless there is clear evidence from your data or the analysis of e.g. the temperature history that it can only be NAT that nucleated in the STS clouds. If there is no clear direct evidence from the current observations that NAT was present in the PSC layer, clear statements in the existence of a “NAT layer” should be avoided. It should be stated more clearly in the manuscript what really was observed and what was assumed or expected, or just evident on the basis of other studies.

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Is there any independent microphysical or instrumental explanation for the need to shift the  $1/R532$  value in order to separate the NAT/Mix2 from the Ice regime? In the current manuscript this shift appears to be more motivated from empirical arguments, just to make things fit better. On page 8, lines 19-24, the authors state: “Without the latter change, a substantial part of the branch connecting STS and fully developed ice clouds would have been counted as NAT/Mix2 instead of ice ...”. This is not really clear to me. What do you mean with a “fully developed ice cloud” and why can it not be a mixed ice/NAT/Mix2 cloud? Thermodynamically, ice nitric acid hydrate particles can well coexist.

The introduction could be shortened for the heterogeneous chemistry part and extended for a more thorough introduction to the PCS classification from lidar data which is more relevant to this manuscript. How is “depolarisation” defined here? And how does it depend on particle size, shape, and mixtures? What explains the higher depolarisation ratio of the upper so-called “NAT ice” branch in Figure 4? And what do you mean with “particle depolarisation”? It may be a common term, but I think it is the light scattered at the particles that is depolarized.

In revising the manuscript, I also recommend to check the text for the proper use of English grammar and punctuation. Also please consider the proper definition and use of terms, e.g. explain in the abstract what kind of “histogram” you are referring to, replace “frost point Tice” with “frost point temperature Tice”, replace “warm/cold temperature” with “high/low” temperature, etc.

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Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-1044>, 2017.

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