Atmospheric
Chemistry
and Physics
Discussions

ACPD

Interactive
comment

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

Christiane Voigt et al.

christiane.voigt@dlr.de

Received and published: 5 July 2018

Reply to Anonymous Referee #2

We thank referee 2 for the positive evaluation of the "uniqueness of the dataset on polar stratospheric clouds", their quality and their scientific importance. We agree that the data provide "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."

We thank referee 2 for insightful comments, which helped to improve the manuscript.

[Printer-friendly version](#)

[Discussion paper](#)

In the following we abbreviate and enumerate the referee's comments and the replies (comment 1: C1; Reply 1: R1)

Main comments:

C1: 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.

R1: We thank the referee for this comment. We agree that the spatial collocation of NAT Mix2 and high-depol ice alone does not necessarily point to the role of solid nitric acid hydrates. Therefore, we now added a Lagrangian match study to the manuscript, using domain filling trajectory calculations starting in the ice PSC matched with all CALIPSO PSC curtain plots detected up to 5 days prior to the 22 January 2016 PSC. Using a statistical approach we investigate whether PSCs were present on the match points before ice, - and if so, which PSC type has been detected by CALIPSO on the match points. The result is convincing: NAT (and only NAT, no other PSC type) has been measured on the match points for the upper ice branch, now called high-depol ice mode, prior to high-depol ice. On the contrary, STS or no PSC has predominantly been measured on the match points for the lower ice branch, now named low-depol ice mode. The comprehensive Lagrangian study therefore adds new observational evidence for ice nucleation on NAT Mix2 for the high-depol ice mode. In addition, it supports ice nucleation in STS for the low-depol ice mode. We added a section and a new figure of PSC composition prior to ice to the manuscript. We excuse the lack of detail that the referee is missing in the previous version of the manuscript.

C2: The second comment is on the evolution of STS with decreasing temperatures par-

C2

allel to the existence of 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. ...But what about their role in forming ice after the temperature drops below Tice?

R2: Binary sulfuric acid/water (BS) droplets grow by condensation upon cooling, initially mainly by uptake of H2O from the gas phase and then upon further cooling to temperatures 3 to 4 K < T_NAT by uptake of H2O and HNO3, forming ternary solution particles STS. Thereby the STS equilibrium temperature T_STS is approximately T_NAT-3.5 K (Carslaw et al., J Phys Chem, 1995). If NAT already exists, then due to the presence of NAT which determines the HNO3 and H2O partial pressures, T_STS decreases and the liquid droplets remain binary down to lower temperatures and grow more slowly. At temperatures a few K above T_ice, STS and NAT can coexist, that is why NAT is called NAT Mix2 to indicate the potential coexistence of STS (or BS) and NAT. If NAT nucleates in the presence of STS, the STS particles will release HNO3 and H2O to the gas phase and will decrease in size. Gasphase HNO3 and H2O condense on NAT due to the lower HNO3 and H2O partial pressures of NAT compared to STS. Depending on their size, due to the Kelvin effect the binary solution particle might be too small to get activated into sizes that allow for ice nucleation at the ice frost point T_ice and further cooling is required to enable their growth. So the meteorological conditions, temperature, H2O, HNO3 (and H2SO4) mixing ratios and the time scales for ice / NAT nucleation determine equilibrium or non-equilibrium conditions and respective PSC particle compositions.

Regarding ice formation in the high-depol ice regime on 22 January 2016, at the time of ice nucleation the STS droplet population with particle depolarization ratios < 0.03 would have to jump into the high-depol ice mode to particle depolarization ratios > 0.3, which is less obvious than ice nucleation on NAT. In the latter case the particle depolarization ratio changes only slightly.

C3: 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 Tice 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.

R3: We thank the referee for this comment and now show the temperature different to T_ice along the trajectories and include the ice saturation ratio S_ice in the discussion of ice formation pathways in order to facilitate the comparison to laboratory measurements. Further include the supersaturation thresholds for heterogeneous and homogeneous ice and NAT nucleation in the manuscript, discuss them and include the respective references from the laboratory (e.g. hom ice nucleation: Murphy and Koop, Q. J. R. Meteorol. Soc., 2005; heterogeneous ice nucleation: Hoose and Möhler, Atmos. Chem. Phys., 2012; homogeneous and heterogeneous NAT nucleation: Knopf et al., Atmos. Chem. Phys., 2002; Knopf, J PhysChem, 2006; Hoyle et al., Atmos. Chem. Phys., 2013; and references therein). We note that small scale temperature fluctuations add a significant uncertainty to the to the large scale saturation ratios derived from the meteorological model.

C4: Sentences like in line 32 of page 7 ("As soon as temperatures decrease below Tice ice PSCs are present." are misleading because also heterogeneous ice formation usually requires some supersaturated conditions to occur at significant rates.

R4: We excuse that inadequate wording and take this sentence out in the revised version of the manuscript.

C5: 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?

R5: We are sorry for this inconsistency pointed out by the referee and now use the PSC nomenclature defined and presented in Figure 4 more consistently throughout the manuscript.

C6: And what about the question of NAT vs. NAD?

R6: As mentioned in the manuscript, the formation of NAD from binary nitric acid water solutions has been observed in the laboratory (e.g. Knopf et al., ACP, 2002; Wagner et al., J Phys Chem, 2005; Stetzer et al., ACP, 2006; Möhler et al., ACP, 2006), but homogeneous nucleation rates are too low to explain denitrification (Knopf et al., ACP, 2002). In addition, pseudo-heterogeneous nucleation rates of NAD and NAT (Knopf, J PhysChem, 2006) are too low to explain observed nitric acid hydrate PSC number densities. Observational evidence for the presence of NAD in the atmosphere is missing so far, e.g. Höpfner et al. (2006) found no spectroscopic evidence for the presence of NAD from MIPAS observations of PSCs over Antarctica. The Lidar measurements cannot distinguish between NAT and NAD, additional information e.g. from infrared spectroscopy is required.

We now discuss the laboratory studies in more detail in sight of the atmospheric measurements. However, for reasons of consistency with the lidar classification of NAT Mix1 and NAT Mix2, we prefer to use this nomenclature consistently throughout the manuscript as suggested by the reviewer in the previous comment.

C7: 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?

R7: The $1/R_{ice}$ value of 0.3 is used to include the major fraction of ice with low particle depolarization ratios (low-depol ice) into the ice mode (linked to STS). This is evident in Figure 4. We further give now additional evidence for this threshold by showing the ice, NAT Mix2, NAT Mix1 and STS occurrence versus temperature to T_ice in a new

figure. We also show the T_ice contour in Figure 5. We discuss the choice of the 1/Rice threshold for the WALES observations in more detail in Section 5.2 and in the supplementary material S1. We note that "The analysis of CALIPSO measurements throughout the winter 2015/16 with changing HNO3 and H2O concentrations requires a variable 1/Rice threshold as used by Pitts et al. (2018)."

C8: 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?

R8: We renamed the NAT ice branch to "high-depol ice mode". Further we now give extended information on the particle depolarization in the instrumental section 2.1. The particle depolarisation ratio is sensitive to the particle shape and size. Spherical particles do not depolarize and particles much smaller than the wavelength of the laser light also show unmeasurable low values. But in general there is no simple relation between depolarization and particle shape, size or composition, see for example Reichardt et al. (2002) for a more detailed discussion of this topic. Nevertheless, cloud regions which show distinct depolarisation ratios point to a significantly different shape or size distribution.

The higher depolarization of the high-depol ice mode could be explained by differences in the shape of the size distribution of the high-depol and the low-depol ice mode. Ice nucleation on NAT might lead to fewer and larger ice crystals and larger asphericity of the ice particle nucleated on solid NAT compared to the ice crystals nucleated in STS.

C9: Check for proper use of English and use of clear definitions.

R9: We checked for the proper use of English and explain the type of histogram in the abstract. We further use high and low temperatures and the frost point temperature T_ice.

[ACPD](#)

Interactive
comment

Printer-friendly version

Discussion paper