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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Interannual variations of early winter Antarctic polar stratospheric cloud formation and nitric acid observed by CALIOP and MLS" *by* A. Lambert et al.

Anonymous Referee #1

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General comments

In this paper, Lambert et al. investigate in detail the start of the PSC season in the Antarctic stratosphere with the aim to explain the early formation of nitric acid trihydrate (NAT). For this, a decade of quasi-simultaneous observations of HNO3 (and H2O) by the MLS instrument on Aura and PSC measurements by the CALIPSO lidar are utilised. The authors argue that observed early loss of HNO3 from the gas-phase without co-incident detection of PSCs by CALIPSO is due to the formation of large NAT particles of low number density. These findings are corroborated by trajectory calculations indicating temperatures some Kelvin below the NAT equilibrium temperature as a prerequisite for the occurrence of such situations. This supports early NAT formation mechanisms

without the necessity for the existence of ice particles, thus backing previous observations in the Arctic wintertime stratosphere. Further, a comprehensive overview of ten years of MLS and CALIPSO early-Antarctic wintertime observations shows the relative strength of NAT versus STS formation in each year.

The paper is well written and conclusive. Especially I would like to point out the effort the authors have taken in the development of new methods of data presentation to support their argumentation. Thus, I strongly support its publication in ACP.

The only major points I would like the authors to consider are: (1) is the discussion of the IR limb-instruments' sensitivity of any help for the flow of the arguments or does it perhaps distract the reader from the important points? (2) I miss a more in-depth discussion on measurement restrictions, like the accuracy of MLS and the lack of observations in the central part of the vortex, on the conclusions drawn.

Specific comments

P4L30: 'scaled potential vorticity'

Please add a reference for sPV or a short explanation.

P5L20: 'with single-profile precisions of 4–15 et al., 2007) and 0.7 ppbv for HNO3 (Santee et al., 2007)'

Could you also give estimates on the accuracy of H2O and HNO3 from MLS in the wintertime high-latitude regions (systematic error estimation and/or comparison with independent observations)? How strong would errors in those gases affect your following argumentation?

P6L3: 'Typical lower stratospheric polar values for TICE and TNAT are 188 K and 195 K, respectively.'

These values depend on pressure, H2O and HNO3. Could you indicate the ranges?

P8L16: 'For a mid-infrared limb sounder operating in the window-region near 12 μm

we use an extinction threshold, kext = $4\times10-5$ km-1 based on measurements by the Improved Stratosphere and Mesosphere Sounder (Lambert et al., 1996).' Also e.g. P8L22,P10L29

Generally, I do not understand why IR limb sounders are included in the discussion while such measurements are not used at all in the scientific argumentation of the paper. Furthermore, such a threshold depends very strongly on instrumental details like e.g. spectral noise, field-of-view and radiometric calibration accuracy. I would not assume it justified to use here thresholds determined for an instrument (ISAMS) which has not been operated in parallel to CALIPSO and MLS but to skip MIPAS and HIRDLS. In addition, after the paper by Lambert et al, 1996, there have been findings regarding simulation of limb radiances and the importance of scattered mid-IR radiation (e.g. Höpfner, 2004) which do also impact the argumentation on sensitivity (by increasing the sensitivity for particles larger than 1 μm). In conclusion, I would suggest to either go into more detail regarding the IR limb-sounding thresholds (instrumental parameters, refractive indices used for STS and NAT, forward model including scattering...), or, perhaps better, skip this discussion and concentrate on LIDAR and MLS.

P9L2: 'The morphology of NAT particles is still an open question, as is the compactness of the particles (Molleker et al., 2014; Woiwode et al., 2014).'

One could also mention here a recent publication by Woiwode et al., 2016.

P15L13: 'However, further averaging of CALIOP backscatter (as discussed in Section 3) on 27 May (not shown) does indicate a considerably larger area of MIX1 class, and so it appears that we are dealing with the limit of the L2PSCMask detection range.'

Where does this extended area lie? Below (indicating sedimentation) or in the TTE region? Maybe a figure could be added as supplemental material

P15L20 and Fig. 9: 'can lead to a complete removal of the available ambient HNO3 from the gas phase.'

Could you comment on the large amount of negative HNO3 values. Are those compatible with the MLS precision or does it hint to some systematic error?

P17, chapter 6.

I would like to see some discussion on the centeredness of the polar vortex around the S-pole between the different years and whether this correlates somehow with the presented data – especially related also to the central part, which is missed by the observations.

P40, Fig. 10:

Could you show here also T-Tice (e.g. in the appendix)? Does this look significantly different from TTE?

Technical corrections

P3L10: 'predictibility' -> 'predictability'

P4L8: 'superceded' -> 'superseded'

P5L17: 'aproximately' -> 'approximately'

P11L16: 'correponding' -> 'corresponding'

P34, caption Fig. 6: 'amibient' -> 'ambient'

P39, x-axis labels: Please indicate if the major tick mark labels are the beginning or the middle of the month.

P40, Fig. 10: Please add information on the real date (in addition to 2009d132, ...)- at least in the caption for the first and last day.

References

Höpfner, M., Study on the impact of polar stratospheric clouds on high resolution mid-IR limb emission spectra, J. Quant. Spectrosc. Radiat. Transfer, Vol. 83, No. 1,

93-107, doi: 10.1016/S0022-4073(02)00299-6, 2004.

Woiwode W., M. Höpfner, L. Bi, M. C. Pitts, L. R. Poole, H. Oelhaf, S. Molleker, S. Borrmann, M. Klingebiel, G. Belyaev, A. Ebersoldt, S. Griessbach, J.-U. Grooß, T. Gulde, M. Krämer, G. Maucher, C. Piesch, C. Rolf, C. Sartorius, R. Spang, and J. Orphal, Spectroscopic evidence for large aspherical β-NAT particles involved in denitrification in the December 2011 Arctic stratosphere Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-146, accepted for ACP, 2016.

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