**Atmospheric
Chemistry
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Discussions**

Interactive comment on "Testing our understanding of Arctic denitrification using MIPAS-E satellite measurements in winter 2002/3" *by* S. Davies et al.

Anonymous Referee #1

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Review of the manuscript submitted to ACPD "Testing our understanding of Arctic denitrification using MIPAS-E satellite measurements in winter 2002/3", by S. Davies, G. Mann, K. Carslaw, M. Chipperfield, J. Remedios, A. Waterfall, G. Allen, R. Spang, and G. C. Toon

The paper presents a comparison between HNO_3 distributions produced by the DLAPSE/SLIMCAT model and HNO_3 fields observed by MIPAS/Envisat during December 2002 to January 2003. Denitrification in MIPAS data is estimated at HNO_3 - NO_y* where NO_y* is determined from a correlation with N_2O, while in the model the

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quantity to compare with is (HNO3 - passive NOy). The evolution of the Arctic denitrification during December 2002/January 2003 as produced by 2 different model runs is compared to the "denitrification" measured by MIPAS, and the timing and spatial distribution of denitrification is analyzed. One MarkIV profile measured on 16 December 2002 contributes another data point to compare with, providing the opportunity to compare NOy-NOy* instead of HNO3-NOy*. The authors conclude that the DLAPSE model is broadly able to capture timing of onset and spatial distribution of the observed denitrification.

General comments:

Provided that two assumptions are valid, namely: 1) HNO3 is a measure for NOy. 2) The air masses observed are PSC free, i.e. NOy is not reversibly sequestered in PSC particles.

the model-data comparison has been done carefully and provides a solid piece of work. However, I have some doubt on the two assumptions given above. For this reason, I did not go into each detail with respect to specific comments, as long as the authors have not dispelled my reservation. Provided that the authors can convincingly argue against my comments, I recommend publication of the paper.

1) The authors do not demonstrate sufficiently well in their paper that HNO3 indeed is a measure of NOy. In a previous paper (Davies et al., ACPD, 2005) they state: "...but we did not couple the denitrification model to the full chemical evolution of the Arctic winter vortex. In this study, we include that coupling because NOy partitioning is important when comparing with observations of HNO3 and NOy species." I.e. the authors are aware that sequestration/denitrification has impact on the NOy partitioning. Mengistu Tsidu et al. (2005) have demonstrated that the NOy partitioning and the HNO3/NOy ratio is not constant, neither in time nor in space, over some days of a polar winter. This is in clear contradiction to the assumption here that HNO3 is a measure of NOy. Comparing model (HNO3-passiveNOy) to measured (HNO3-NOy*) alone does

not help since there is no proof that the NOy partitioning as modeled and measured, respectively, are sufficiently consistent.

Some arguments have been given that most of NOy should be composed of HNO3 (Section 2.4, first para). However, to my opinion the authors must demonstrate more thoroughly and quantify, by reference to previous publications or other literature, or by comparison of their model NOy partitioning to observational data, to what degree HNO3 indeed is a measure for NOy; i.e. they must show either that NOy partitioning is captured correctly by their model (on basis of observational NOy data), or that the HNO3/NOy ratio is sufficiently constant over time and space. Further, why has NO2 also available from MIPAS not been used to further constrain the total NOy?

2) The authors define denitrification as "irreversible loss of total reactive nitrogen (...) from an airmass by the gravitational sedimentation of NOy-containing particles." This can be derived from reduction of gas-phase NOy (or HNO3, if validity of item 1 is demonstrated) if PSCs are no longer present. In presence of PSCs reduction of gas-phase NOy (HNO3) could also be due to reversible uptake in PSC particles without sedimentation.

In order to exclude PSC-affected observations, the authors restrict the MIPAS observations used in the study to those with a cloud index < 2.0 (see for example, page 11002, line 16, or figure 2). However, Spang et al. (ACP, 2005) state (page 681, end of first/beginning of second column): "In MIPAS operational processing a conservative threshold of 1.8 has been implemented which guarantees the detection of strong cloud events over relatively broad height range of 12 - 40 km. The clouds identified with this threshold value are those which are most likely to affect the retrieval of trace gases from MIPAS data. For the detection of PSCs we extended the threshold value to 4 but limited the height region to 14 to 30 km. At this threshold level, optically thinner cloud events are detectable at levels which are still in excess of the noise level;...".

According to this reference, PSC detection is not safe with a cloud index of < 2, and

PSCs can occur up to a CI of 4.0. This is in agreement with our own experience. As a consequence, I assume that many of the MIPAS observations used in this intercomparison are not for PSC-free conditions. The additional selection criteria, TNAT-2K, also allows for NAT formation. Further, even if a PSC was not in the MIPAS' field-of-view, it could have been close-by, also affecting the concentration of NOy in the air mass sampled by MIPAS. The authors explicitly state that the MIPAS observations were taken during a period with considerable PSC occurance. I conclude that sequestering of HNO3 (NOy) in PSCs cannot be excluded for the data set from MIPAS. For this reason, validation of a denitrification scheme in a model is, to my opinion, not possible on basis of this data set.

In particular, since R. Spang is co-author of this paper, I am surprised to see such contradicting application of the cloud index introduced by him.

Specific comments: page 11002, line 13: Validation of HNO3 and N2O has been published by Oelhaf et al., 2004, and Camy-Peyret et al., 2004, respectively. These papers should be cited here. For exact references, see below.

page 11002, line 15: there are two references "Spang et al., 2005"; please specify which one is meant here.

page 11005, line 5: The Popp et al. correlation for NOy vs. N2O obviously has been applied to HNO3 to determine NOy*. Has HNO3 been corrected by 1/0.9, as estimated from the model runs, to give measured NOy before calculating NOy*?

Fig.1, caption (c): It is not clear to me what negative denitrification means; is this re-nitrification? In this case I would expect the altitude distribution vice versa than shown.

Technical corrections: page 11015, line 24: remove doubled "of distribution"

page 11007, last line: it must read CI $<$ 2.0 (instead of CI $>$ 2.0)

References: Camy-Peyret, C., G. Dufour, S. Payan, H. Oelhaf, G. Wetzel, G. Stiller, Th. Blumenstock, C.E. Blom, T. Gulde, N. Glatthor, A. Engel, M. Pirre, V. Catoire, G.

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Oelhaf, H., T. Blumenstock, M. De Mazière, S. Mikuteit, C. Vigouroux, S. Wood, G. Bianchini, R. Baumann, C. Blom, U. Cortesi, G.Y. Liu , H. Schlager, C. Camy-Peyret, V. Catoire, M. Pirre, K. Strong, G. Wetzel, Validation of MIPAS-ENVISAT version 4.61 HNO3 operational data by stratospheric balloon, aircraft and ground-based measurements, Proc. ACVE-2 meeting, 3-7 May 2004, Frascati, Italy, 2004.

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