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Interactive Comment

Interactive comment on "Spectroscopic evidence for β-NAT, STS, and ice in MIPAS infrared limb emission measurements of polar stratospheric clouds" *by* M. Höpfner et al.

M. Höpfner et al.

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We would like to thank referee 3 for encouraging us to use more quantitative arguments to describe the fit quality between our measurements and simulations.

1. *... The authors have used a very complete and time-consuming method to assign the composition of the PSCs observed in three specific instances. Then, a simpler, more tractable method is presented for evaluating the entire winter. What I do not see, however, is any connection between the two. Was the simpler method tested against the more rigorous method? How did it fare? How sensitive is it? Would the inclusion of other bands allow for a more constrained but still efficient*

process.

The logical structure in the paper regarding the two applied methods is the following:

(a) We have applied the time-consuming spectrally broadband simulations to the three example cases where Lidar observed a uniform cloud type over its measurement range. This provided us with information, which of the published refractive indices agree best with the different cloud types.

(b) The resulting choice of refractive indices of step (a) was applied to perform synthetic observations over a wide range of possible PSC number densities and radii in order to characterize the fast colour ratio method.

(c) The fast method is finally compared with available MIPAS/Lidar coincident measurements to test its applicability to real data.

The colour ratio method relies on the 820 cm^{-1} signature, which is related to NAT (c.f. spectral broadband calculations in chapter 3 and Figure 4). Thus, we believe that connection between the two methods is reliable. We have, as the referee suggested below, highlighted the three examples in the colour-ratio figure 9 of the revised paper.

In this paper we intend to investigate more quantitatively the fast method which has been used in previous publications and for other instruments. The development of new methods by including other bands is beyond the scope of this paper but will be tackled in future.

2. *Returning for a moment to the spectroscopic analyses of Section 3: The assignment of the best composition for the three examples shown in Figs 4-6 looks reasonable in a qualitative way, but can it be quantified? can you tell me what to look for specifically? Is it just the least variation over the whole window that defines best or are there more focused spectral regions you used when tuning*

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the radii, and judging goodness of fit? Did you look at residuals between the fits and the observations? how much better were the chosen identities than the ones not chosen?

Following the referee's suggestion to make the comparison more quantitative we have added tables with root-mean-square differences between measurements and simulations for all cases in the revised manuscript . The discussion in the text has been extended accordingly.

3. *When looking at Fig 4, I do see that the overall qualitative fit is best with the Biermann beta-NAT optical constants. But the significant difference between Biermann and Toon makes me wonder if the Richwine data isn't at least as good as those two, suggesting that alpha-NAT might also be a possibility.*

   From the new Tables, one can see that the α-NAT simulations based on Richwine indices fit better than those by Toon in the low wavenumber channel. However, in the high-wavenumber region those by Toon result in better agreement. We agree with the referee and don't claim any longer evidence for β-NAT, but for NAT. We have skipped β it in the title and adjusted the abstract and the conclusions accordingly.

4. *(Thank you for including the dashed, no-PSC spectrum. It's crucial for separating the important parts of the spectrum from the irrelevant parts - but it's hidden under the legend! Please change the scale, or move the legend, or something.)*

   We have changed the plots accordingly.

5. *On the same note, on p. 10698, line 11-12, you suggest that the Biermann[mol] indices might be more appropriate, yet in Fig 4, right hand side, the [coa] plot looks like a better fit. How do you weight the two in deciding what is better, especially since the results plotted for the Richwine alpha-NAT indices look very similar to the [mol] results.*

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From the spectroscopic comparison (also by investigating the new tables) we agree with the referee: Biermann[coa] β-NAT clearly results in the best fits. However, comparing the retrieved altitude parameters with Lidar the results based on the Biermann[mol] data seem to agree better. These observations are discussed against each other in the conclusions.

6. *Section 4 presents a simpler, quicker method for assaying the whole winter, but I found it difficult to really understand what you are doing. Is the whole process based solely on the 820 cm-1 band? Why were 792 and 832 cm-1 also chosen? Did they provide the best discrimination among the possible compositions? In looking at the laboratory data in Fig 1, it seems that including points near 1400 and 1450 cm-1 would be very useful for discriminating between NAT and NAD.*

   The 820 cm^{-1} band is used for identification of NAT. The other two wavenumber regions are used for general cloud detection at a nearly trace-gas free location (832 cm^{-1}) and as a reference point with medium optical depth to reduce the temperature sensitivity of emission measurements (792 cm^{-1}). This is explained in more detail in the revised paper.

   However, we would like to stress that the method has not been invented in this paper but has been described and used in several publications previously. By our investigations we tried to give the method a better basis by (1) simulating for the fist time the 820 cm^{-1} signature well and identifying it as a NAT feature (which previously has only indirectly been deduced by correlations with temperature) and (2) quantification of its limits in terms of particle size and volume density.

   We have done first tests to include further bands in the colour-ratio method, but this did not lead to any improvement. The reasons are that other bands are, compared to the 820 cm^{-1} signature, (1) more influenced by trace gas emissions, (2) not as spectrally sharp and prominent in the observation, and (3) more influenced by scattering (see answer to comments by referee 1).

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7. *Figure 9 is confusing and frustrating in several ways. Please describe it more thoroughly. Is everything plotted in Fig 9 theoretical, i.e., calculated from indices and radii and number densities, but not from MIPAS observations? What is the solid black line? How are the boundaries of the four regions determined? How did you test the placement of the boundaries; that is, what is the uncertainty in the assignments near the boundaries? Where would your three cases from Figs 4-6 fall on this plot? How many lidar/MIPAS coincidences do you have? Could they be plotted like Fig 9 to show how clustered or not the observations are?*

   The original figure caption lead to some misunderstanding. There were only simulations shown in the plot. We have revised the figure caption and discussion in the manuscript for clarification:

   “We have analysed this empirically derived colour-ratio method in a quantitative way by radiative transfer simulations. As a basis for these calculations we applied those refractive indices which resulted in the best agreement between MIPAS spectra and simulations in the broadband retrieval tests performed for the three typical Lidar cases as discussed in section 3.3: [Biermann(1998)][coa] for NAT, [Biermann et al.(2000)] for STS, and [Toon et al.(1994)] for ice.

   We have used PSC volume densities for a variety of temperature profiles covering the range of variability in the Antarctic stratosphere, derived via equilibrium calculations and assuming different supersaturations. Further, various median particle radii were applied. The results are shown in Figure 9.

   The plot is subdivided into regions of colour-ratios clearly related to one PSC composition and such which do not allow an unambiguous assignment. Region R1 contains only points from spectra calculated for β-NAT (red symbols). The solid black line (NAT detection line) separates the colour-ratios unambiguously related to NAT from the ambiguous ones. It was constructed as follows: a regression function of the type $1/(a + b \times CI + c \times CI^2)$ has been fitted to the STS and ice radiance ratio distributions (open blue and green symbols in Fig. **??**).

This curve has been scaled by factor constant for all CI such that all points for NAT simulations with particles $\leq 2\,\mu$m radius were still above the resulting curve $(1.13/(0.164 + 0.884 \times CI - 0.065 \times CI^2))$. For larger particles, the 820 cm^{-1} signature flattens and, thus, it cannot be separated from STS or ice any more.

The R3 region, below the NAT detection line and left of the CI = 1.3 reference line, is dominated by simulated ice data points, and the R2 and R4 regions, below the NAT detection curve and right of the CI = 1.3 reference line contain simulated data points for all types of PSCs: STS, ice and large NAT particles with radii $>3\,\mu$m. Region R2 contains less than 5% simulated ice data points and the R3 region less than 5% STS and no NAT. Thus, a measurement falling into the R1 region can be assigned to NAT, a measurement falling into R3 can be ice or STS, and R2 and R3 measurements do not allow any clear assignment. In case of optically thick spectra, STS can be ruled out also for R3 measured data which implies unambiguous identification of ice.

The three test cases used for the detailed broadband simulations well represent the different colour-ratios of PSCs (solid bullets in Fig. 9."

We have also included a new figure (Fig. 11 in the revised manuscript) where we show the MIPAS-Lidar coincidences in a colour-ratio scatterplot as the referee suggested. The following discussion has been added:

“Figure 11 shows the same coincident data between MIPAS and Lidar as in Fig. 9, but as scatterplot of colour-ratios as derived from measured MIPAS spectra like the simulations in Fig. 9. The data points are colour-coded on basis of the types derived from the coincident Lidar observations (Type 1a: red, Type 1b: green, Type 2: blue). Though there are not many Lidar observations of ice and STS compared to NAT, the plot is in general agreement with the simulations (Fig. 9): ice is mainly located in R3, STS in R2 and NAT in R1. Additionally only two ice-cases fall in R1 and the separation curve for R1 also follows rather well the ice and STS points. While most of the Lidar Type 1a PSCs lie in R1 some also fall

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in other regions, even in R3, which is not indicated by our simulations in Fig. 9. These cases have been discussed in context of Fig. 10 and are attributed to non-perfect co-incidences in sounded air-masses by MIPAS and Lidar, especially in the vicinity of large horizontal inhomogeneities."

8. *Other comments: pp. 10694-95: I don't understand why you used ranges of water and nitric acid mixing ratios to calculate the STS composition when you state in the next paragraph that you determined the abundances of the trace gases directly from the MIPAS data. Please clarify.*

   The problem here is that the equilibrium calculations should be made on basis of the total HNO_3 and H_2O abundances (i.e. gas phase + sequestered). MIPAS, however, can only measure the gas-phases. Thus, for the June cases (Type 1b and Type 2), we applied mean vortex gas-phase MIPAS measurements from May when no PSCs were present. Here we assume that neither strong denitrification nor strong subsidence had taken place between mid-May and mid-June. This seems to be roughly the case since the STS volumes fit quite well. However, for our Type 1a example in August, there is no such reference MIPAS observation of the total HNO_3 or H_2O inside the vortex since there are always PSCs present. The reason to show the MIPAS derived HNO_3 of the same limb-scan was to make clear that there must have been denitrification above about 20 km, since there are only small amounts of gas phase HNO_3 and additionally, only little volume of PSC. Thus, the reasoning for the Type 1a case can only be qualitative here. We have tried to state this more clearly in the revised version.

9. *I, too, was quite confused by your switching back and forth between volume and number density when discussing Fig 7. Please clarify and remove any unnecessary switching. Could the entire argument (and figure) be presented in terms of volume?*

   Thank you for this suggestion. In the new version we have skipped the number

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density in this chapter and the Figure and hope to have improved readability.

10. *Figure 8: I would recommend switching the dashed and solid lidar lines. It is easier to compare three solid lines than to ignore one solid line while comparing two solids and a dashed line.*

    OK, done.

11. *Minor comments*

    We agree with all minor comments and have corrected these in the text. Especially, also in response to comments by referee 2, we have revised the paragraph on volume density comparisons.

References

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