Author response to Reviewer Comments for: “Denitrification by large NAT particles: the impact of reduced settling velocities and hints on particle characteristics”

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W. Woiwode et al.

We thank the two reviewers for a thorough review and concise comments that clearly will help to improve our manuscript. In the following, we address the comments of both reviewers. Page and line numbers refer to the discussion paper (*acpd-14-5893-2014.pdf*). We provide in each case the original referee comment (bold italic letters) followed by our response. Text that was added or modified in the revised manuscript is indicated in blue. For easy tracking of the modifications we furthermore provide the modified manuscript in the attachment (modified text in blue).

Summary of general issues

As a major point, both reviewers point out that alternative explanations for the observed apparent large NAT particles and denitrification were not ruled out. Specifically, the impact of temperature biases, temperature fluctuations, modified nucleation rate, alternative nucleation mechanisms (e.g. NAT on ice) and uncertainties in ambient H_2O and HNO_3 on the modelled particle sizes and denitrification should be investigated to rule out more simple explanations for the presented particle observations and observed HNO_3 redistribution.

We fully agree that the presented case study is limited and cannot serve as a proof for certain particle settling velocities or even the physical appearance of the particles (compare P5911/L18ff; P5912/L28ff). The comparison of our observations and simulations of HNO_3 redistribution is limited due to differences between the model world and the real atmosphere as seen by the instrument. Furthermore it is not clear how the presented particle observations have to be interpreted in the case of considerably aspheric particles.

However, we feel that the conclusions of our work have been misunderstood in some way. Motivated by the very large particle sizes observed, the obvious discrepancy between observed and modelled particle sizes and the characteristics of the available dataset we decided to go one step further and focus on particle settling velocities. Reduced settling velocities are a logical consequence of alternative particle properties that might solve this discrepancy. Therefore, reduced settling velocities are likely in our eyes.

The idea of our work was to investigate the sensitivity of this individual model parameter taking into account a state-of-the-art model parameterisation and compare the simulated HNO_3 redistribution to our observations. An extensive investigation of the influence towards the manifold other parameters taken into account in the simulation was however not our intention. The suggested sensitivity simulations with respect to different model assumptions would certainly be interesting, however it would still not provide a definitive proof for the properties of NAT particles in the real atmosphere. Some aspects of other parameters influencing HNO_3 redistribution were covered by Grooß et al (2014) and are subject of ongoing work in the RECONCILE framework and beyond.

To support the conclusion that the observed larges particles cannot be compact NAT spheres, we added the size distribution obtained from a sensitivity simulation with 1 K lower temperature.

We agree that the evaluation of our results needs improvement. Therefore we clarified the (speculative) statements as follows: Our work shows that reduced settling velocities significantly modify vertical HNO_3 redistribution and that settling velocity is an important parameter in simulations. Furthermore, taking into account the chosen state-of-the-art simulation, moderately reduced settling velocities result in best agreement between observed and simulated HNO_3 redistribution. Finally, taking into account the limited growing time of the observed particles and the results of the comparisons between simulated and observed HNO_3 redistribution we speculate that the apparent

large NAT particles observed on 25 January 2010 have been compact aspheric (columnar or better: elongated) NAT particles. Such particles could grow to large optically apparent dimensions in shorter time while having moderately reduced settling velocities that were compatible with our results. This is clearly a hypothesis. We think that the parameter "settling velocity of NAT particles" is important especially for simulations of Arctic vortices, where HNO_3 redistribution is often limited to frequent periods under supersaturated conditions.
In the following we address the specific comments and provide additional information suggested by the reviewers.

Response to Referee #1

This paper showed comparison of HNO3 measured and simulated vertical profiles inside the Arctic vortex. MIPAS measurements. It is well written and structured.
I have one major concern. The main conclusion is that the reduced sedimentation velocity of NAT particles in the CLAMS simulation may improve that agreement of gas phase HNO3 profile with airborne MIPAS-STR measurement.
The main conclusion depends on the results shown by Figure 7 (25.January) and Figure 8 (30. January). On 25 January, PSCs were observed. The HNO3 partitioning between gas phase and condensed phase (NAT and STS) depends critically on temperature. Therefore, one should focus more on 30. January, as the temperature was well above T-NAT and all HNO3 is in the gas phase.
We agree that one should focus especially on the observations on 30 January 2010. However, also the flight on 25 January 2010 contains valuable information, since significant amounts of HNO_3 have been released back into the gas-phase at lower altitudes already. As our focus is on settling velocities, it is interesting to see whether significant amounts of simulated particles have reached already altitudes where evaporation occurred or not.

Panel (a) of Figure 8 shows that the maximum HNO3 of CLaMS calculation lies around 400 K, while the MIPAS-STR data show maximal values around 420 K. The peak value of CLaMS is higher than MIPAS-STR.
The author made only sensitivity tests for sedimentation velocity of NAT particles. However, other factors may also influence the vertical redistribution of HNO3: for example temperature and NAT nucleation rate coefficient would like to see the results of the following sensitivity runs:
1) Increase or decrease the overall temperature by e.g. 1K;
2) Decrease the NAT nucleation rate by 50% and increase the rate by 100% and 200%;
3) Some combinations of 1) and 2).
In addition, in the present study, only NAT formation on dust particles is considered. However, in January 2010, synoptic ice PSCs were also observed (e.g. Engel et al, ACD 2013). NAT can also nucleate on ice particles forming PSC mix2 enhance (Engel 2013). It is totally unclear, how the NAT on ice effect the HNO3 redistribution. If the NAT number is too high, they may have less denitrification potential than fewer but larger NAT particle.
It would very useful, if the author could also implement NAT formation on ice. One can only make the conclusions after the sensitivity tests have been performed.
We agree that other factors are very important for the simulation of vertical redistribution of HNO_3. However, as indicated above, the aim of our work was not to investigate the influence of the manifold other parameters affecting the simulation. Based on a state-of-the-art setup, we wanted to address the specific parameter "settling velocity of NAT particles". Regarding the influence of other parameters on simulated HNO_3 redistribution we refer to the work of Grooß et al. (2014) and ongoing work in the

RECONCILE framework and beyond. We furthermore worked out the speculative statements more clearly and mentioned that other parameters also have important influence on the simulation. Therefore, following modifications were applied:

Abstract (P5895/L9-16): "Utilizing state-of-the-art simulations by the CLaMS and observations by the airborne Fourier transform infrared spectrometer MIPAS-STR we present a case study on the impact of reduced settling velocities of NAT particles on vertical HNO_3 redistribution. The results of our study show that reduced settling velocities of NAT particles significantly modify the simulated vertical HNO_3 redistribution and settling velocity is an important parameter in simulations. Our comparisons of simulated and observed HNO_3 redistribution show good agreement especially when moderately reduced settling velocities are considered. While simulated denitrification also depends critically on other parameters of the simulation not investigated here, we speculate that the large apparent NAT particles observed in situ on 25 January 2010 might have been compact aspheric (e.g. elongated) particles. Such particles could grow to largest maximum sizes in a short time while having only moderately reduced settling velocities that were best compatible with our results."

P5911/L22-26: "…results. Considering the discussed results we speculate that the large particles indicated by the in situ observations during the flight on 25 January 2010 were compact elongated NAT particles capable of fast growth to large maximum sizes and characterised by moderately reduced relative settling velocities (in the order of 70 %) compared to mass-equivalent compact spherical particles."

Conclusions (P5912/L11-5913/L7): "Using simulations by the CLaMS and observations of MIPAS-STR we studied the impact of reduced settling velocities of large NAT particles on vertical HNO_3 redistribution. The comparisons between measured and simulated vertical redistribution of HNO_3 show good agreement especially when the settling velocities of the simulated NAT particles are reduced by a moderate constant factor of 0.7 for the given CLaMS setup. In contrast, a factor of 0.3 results in a significant underestimation of the vertical HNO_3 redistribution by the simulation. Our work shows that reduced settling velocities significantly modify the simulated vertical HNO_3 redistribution. This aspect is important especially for simulations of Arctic vortices, where HNO_3 redistribution is often limited to frequent periods under supersaturated conditions.

Considering the limited growing time and the comparisons between observed and simulated gas-phase HNO_3 we speculate that the apparent large NAT particles observed on 25 January 2010 have been compact considerably aspheric (e.g. elongated) NAT particles. Such particles could grow to largest optically apparent sizes in a short time while having slightly reduced settling velocities compatible with the results of our limited case study. Mass equivalent spherical NAT particles with low mass density or compact disk-shaped NAT particles are less likely candidates, as considerably lower settling velocities were expected for the corresponding particles that could explain the maximum sizes observed in situ, resulting in increased discrepancies between simulated and observed HNO_3 redistribution.

We point out that the simulated HNO_3 redistribution also depends critically on other parameters affecting the simulation (e.g. temperature biases, temperature fluctuations that are not considered, alternative nucleation scenarios and uncertainties in the mixing ratios of HNO_3 and H_2O) which are not adressed by this work. The presented case study is not capable of determining the settling velocities or the physical appearance of NAT particles quantitatively. Our results however show that the settling velocity of NAT particles is an important parameter in simulations and offer a consistent explanation for the presented observations."

Minor points: 1) P1, line 23-25; The sentence "In situ observations by the particle probe FSSP-100 during the RECONCILE campaign indicate unexpected large potential NAT (nitric acid trihydrate) particles inside PSCs. " is misleading. The FSSP data show very large particles, possibly NAT, if the particles were spherical and compact. Rephrase: During the RECONCILE

campaign, apparent very large NAT (nitric acid trihydrate) particles were observed by : In situ observations by the particle probe FSSP-100 inside PSCs. I think that the word "apparent" is important, because this large size is the effect of the instrument.

We rephrased P5895/L4-6 as follows: "During the RECONCILE campaign, apparent very large NAT (nitric acid trihydrate) particles were observed in situ by the particle probe FSSP-100 inside PSCs."

2) P2, L4-6, "The results of our study support the hypothesis that denitrification is produced by significantly aspheric (i.e. columnar) compact NAT particles which are characterised by reduced settling velocities." One cannot exclude the possibility that the NAT particles are not compact and have a smaller density. Suggest: The results of our study indicate that the NAT particle may sediment with a reduced velocity than when they are spherical and compact indicating either the NAT particles are not compact or they are highly non spherical.

(P5895/L13-16) We agree that one cannot exclude that the particles are not compact and have a smaller density. Our limited comparison however would be best compatible with compact significantly aspheric NAT particles that would have large sizes and only moderately reduced settling velocities at the same time. We worked out the speculative character of this statement more clearly (see above: modifications in abstract and conclusions).

3) Add the corresponding size distribution obtained from CLaMs simulation for the same time and same location into Figure 1 would be great.

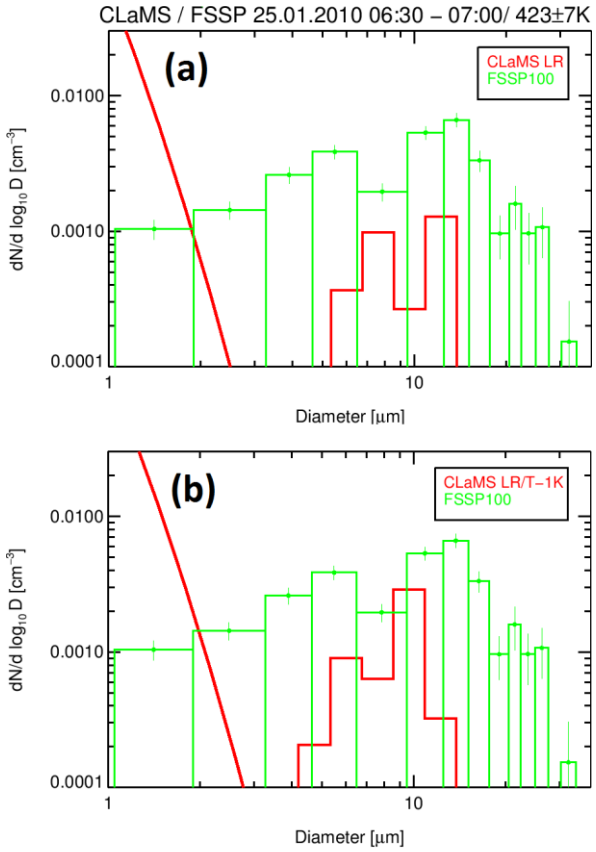

Fig. 1. FSSP-100 size distribution (sizes in diameter) derived for the flight on 25 January 2010 for the time interval 06:30 to 07:00UTC assuming spherical particles (flight altitude 18 km) together with the corresponding size distribution extracted from the CLaMS standard scenario (CLaMS HR). Also shown is the corresponding size distribution considering a temperature bias of -1 K in the simulation (CLaMS HR/T-1K).

Comment: The shown CLaMS size distributions correspond to sensitivity runs with lower resolution. An updated version of Figure 1 considering high resolution simulations (as other simulations discussed in this work) is in preparation.
After P5901/L3 we added:
"Figure 1 shows also the associated size distribution corresponding to the CLaMS standard scenario (see section 5) assuming compact spherical particles. The particle mode situated between about 5 and 12 µm and causing the simulated denitrification is by a factor of 1.5 to 13 lower compared to the observed number densities in this size range. The largest particles observed with sizes above 12 µm are not reproduced by the simulation. One possible (partial) explanation for the observed discrepancy might be that NAT particles have larger optically apparent sizes (diameter or length) compared to compact spherical particles due to non-compact morphology and/or aspheric shapes. The small particle mode below 2 µm corresponds to simulated STS droplets. The high number density mode of small NAT particles described by Grooß et al. (2005) not considered in the discussed simulations.
Also shown in Figure 1 is the corresponding size distribution obtained when a temperature bias of -1 K is considered in the simulation. While the shape of the NAT mode is modified, this setup also fails to reproduce the particles larger than 12 µm indicated by the observation."
The following sentence at P5901/L7-9 becomes obsolete: "For the geolocations … CLaMS domain."
P5901/L9ff was updated: "For 8 of the simulated particles with diameters between 6 to 12 µm and corresponding to the standard scenario backward trajectories were reconstructed and continued by airmass trajectories prior to the nucleation event. The obtained trajectories are presented in Figure 2…"

4) Figure 4. The figure caption is confusing: is the plotted quantity continuum extinction or continuum absorption coefficient? In the figure caption, "continuum extinction" is used and in the label, "continuum absorption".
In this context, extinction corresponds to the imaginary part of the complex refractive index of a material and is equivalent to absorption. For consistency, we adopted "absorption" in the entire revised manuscript.

Response to Referee #2

This paper presents a limited study of denitrification occurring in the 2009/2010 Arctic stratosphere, combining in situ particle measurements (FSSP-100 on the Geophysica), airborne FTIR (MIPAS-STR) and a 3-D simulation of chemistry with Lagrangian transport and sedimentation (CLaMS driven by ERA-Interim winds and temperatures.)
Particles detected by FSSP-100 have diameters up to 30 microns. It is claimed that the NAT particles cannot grow to these large sizes assuming the standard NAT mass density and compact near-spherical shapes within the actual time available for growth under super-saturated conditions (i.e. amount of time following nucleation the particles remain below TNAT). An attempt to model the particle growth with CLaMS shows that the sedimentation velocities may have to be reduced significantly to match the HNO3 gas phase vertical distributions. The reduced settling velocities are attributed to the formation of compact columnar particles with large aspect ratios ~8.
The main concern I have is that far too little has been presented to rule out more mundane explanations of the discrepancy between the observed FSSP particle sizes / MIPSA-STR HNO3 vertical distributions and the CLaMS modeled results. More detailed comments are given in the section below. Presumably more simulations runs would be required address these concerns, but these are essential to strengthen the conclusions of the paper and are clearly within the scope of this study.

As discussed above, we agree that our study cannot serve as a proof for certain settling velocities or physical appearance of NAT particles in the real atmosphere. Our work however clearly shows the importance of the parameter settling velocity. Considering the shown observations of extremely large particles and the temperature trajectories indicating limited time below T_{NAT} (~2 days) and thereby above T_{ICE}, non-spherical or non-compact NAT particles appear as a likely option and lower settling velocities would be a logical consequence. To support the conclusion that the observed larges particles cannot be compact NAT spheres, we added the size distribution obtained from a sensitivity simulation with 1 K lower temperature.
We agree that the speculative statements need to be worked out more clearly and modified abstract, conclusions and the discussion accordingly (see above).

/xxx/ ==> delete xxx
[xxx] ==> add xxx

P5895-L26: "composed of NAT" What about ICE coated NAT?
L5895/L26: "… NAT, ice-coated NAT and potentially…"

P5898-L22: Mie theory accounts for scattering from spherical particles only. Please give a reference for how scattering from "slightly aspheric" particles with random orientation can be approximated adequately by Mie theory.
We wanted to express that particle observations are often evaluated using Mie Theory for simplification and agree that this formulation is misleading. We replaced P5898/L23: "assuming … random orientation." by "assuming approximately spherical particles.".

P5898-L21-26: It's not clear if you used the advanced methods cited or how large the difference is in particle sizing between the MIE and more advanced methods. This seems important given that your conclusions are that the NAT particles have an aspect ratio of about 8 and are therefore not at all "slightly aspheric".
P5898/L24 "Advanced methods (not applied here) …"
P5898/L26: "…(Borrmann et al., 2000). In situ PSC observations during the RECONCILE period and in December 2011 and the FSSP observations are discussed by Molleker et al. (2014). For aspheric particles a broadening of the FSSP size distribution would be expected and the particle volume would be stronger affected by the oversized part. While for particles with aspect ratios of 2 already significant changes in the size distribution have to be taken into account (Borrmann et al., 2000), largest changes are expected for particles with considerably higher aspect ratios. This aspect is however not further exploited here."
We point out that we do not conclude that NAT particles have an aspect ratio of about 8 (P5911/L18ff; P5912/L28ff). We however discuss that our results would be compatible with such a scenario (compare P5911/L22; L5912/L20ff). In the revised manuscript we worked out more clearly the speculative statements.

P5899-L20: contributions [from aerosols] are typical
Done.

P5899-L25: high dynamic/s/[al] range
Done.

P5899-L29 and P5900-L3: I find the term "stray light" somewhat misleading. It would normally used to describe an instrument artefact such as unwanted reflections which could maybe have been reduced by a better optical design. Here you are talking about atmospheric radiation upwelling from the surface and troposphere and being scattered by clouds into the instrument

line of sight i.e. an external contamination of the requisite limb signal. In principle, this effect could be modeled. Since the unwanted tropospheric signature is not the fault of instrument engineering the effect should not be labeled as a stray light problem.
P5899/L28ff: "… indicating scattering of tropospheric radiation by PSC particles into the MIPAS-STR field-of-view at high limb views were not identified."

First you state that tropospheric CO2 and H2O signatures were not identified, but then state that as H2O was not retrieved that "stray light cannot be ruled out for this species". As shown in Hopfner (2004) this is viewed directly by examining the radiance spectrum for evidence of a tropospheric signature of the H2O lines (i.e. an absorption feature) providing particle sizes are in the range 1 to 7 microns.
We agree that clarification is necessary here. As discussed, CO_2-lines with sidelobes and inversed H_2O-lines were not identified. However, due to the strong tropospheric H_2O signature and the possibility that a weak tropospheric contribution potentially scattered into the field-of-view at high viewing angles might alter the weak H_2O signature observed while showing no obvious distortion of the line shape (such as sidelobes) we decided to not exploit the H_2O retrieval further. Another reason not mentioned in the manuscript was that only a rather low vertical resolution was feasible from the weak utilized H_2O signature due to cold temperatures and attenuation by the superimposed broadband continuum signature due to the PSC.
We modified P5899/L29-P5900/L3 as follows: "However, the H_2O retrieval was not exploited for this flight considering the weak intensity of the utilized H_2O signature under the conditions of this flight and the resulting high uncertainties and low vertical resolution of the retrieval results. Furthermore it was not clear whether potential weak contributions of scattered tropospheric radiation not obviously identified in the spectra might significantly alter the weak H_2O signature observed at higher viewing angles."

Overall, I think you need to make a better job of explaining how you extract information on aerosols, temperature and gases from the MIPAS-STR measurements. Additionally, no indication is given at all on the uncertainties in the retrieved HNO3 and temperature from MIPAS-STR.
We added after P5899/L12ff: "Temperature was retrieved utilizing the CO_2 signatures in the microwindows from 810.1 to 813.0 cm^{-1} and 955.6 to 958.5 cm^{-1}. Subsequently HNO_3 was retrieved using the signatures in the microwindow from 866.0 to 870.0 cm^{-1}. Retrieval parameters were the target parameter (temperature or volume mixing ratio), wavenumber-independent background continuum, spectral shift and O_3 as additional parameter in the temperature retrieval. Total combined 1σ-uncertainties were estimated considering uncertainties due to spectral noise, radiometric calibration, spectroscopic line data, line-of-sight knowledge, the adopted CO_2 profile (temperature retrieval) and retrieved temperature (HNO_3 retrieval). Details on the MIPAS-STR retrieval and its validation are discussed by Woiwode et al. (2012)."
We added at P5899/L24: "…logarithmically, resulting in low residuals between the observed and simulated spectra close to the noise level."
We furthermore show and discuss individual retrieved profiles together with estimated uncertainties and vertical resolution in the additional Figure 6 (see below).

P5900-L22: /probable/ [potential]
Done. ("potentially NAT-containing…")

P5900-L23: /sizes in/ diameter[s]
Done. We added furthermore the end of this sentence "if spherical particles are assumed." to clarify that spherical particles were assumed in data processing.

P5900-L27: The text apparently gives the differential number densities at the peaks of the two modes (i.e. the peak bin values corresponding to Fig 1). This is misleading because normally the total number density in each mode would be reported (e.g. Fahey et al (Fig 4 caption) give the integral of their large NAT mode as 2.3E-4 cm-3). You state that your FSSP large NAT mode is a factor of 5 larger than Fahey et al, so we have 5*2.3E-4 cm-3 => 0.00115 cm-3. You need to give the FSSP integrated mode densities so these can be compared directly. Also, try working out what would be the HNO3 content of both NAT modes. This is useful information.

Since the size bins of the shown FSSP-100 observations differ from the size bins given by Fahey et al., (2001), we prefer to give the differential number densities allowing for a direct comparison. The FSSP observations during RECONCILE are discussed in more detail by Molleker et al. (2014) and comparisons with the size distribution from Fahey et al. (2001) are presented. Considering that in our representation the size distribution from Fahey et al., (2001) peaks at 0.0014 cm^{-3} we find a factor of 5 with respect to the maximum of the shown size distribution. For clarification we added after P5901/L3: "…14.5 µm. In the shown representation indicating differential number densities the large NAT mode discussed by Fahey et al. (2001) peaks at about 0.0014 cm^{-3} (compare Molleker et al., 2014). Assuming spherical particles, the HNO_3 content of the complete size distribution is estimated to be equivalent to about 11.5 ppbv of gas phase HNO_3 considering an ambient temperature of 195 K and a pressure of 60 mBar. Thereby, 11 ppbv would correspond to the size bins higher than 9.5 µm. "

P5901-L2: mode is /by/ about [a] factor [of] 5

Done.

P5901-L19: "yielding compact spherical particles". The particle shape and form is not being simulated and is therefore not "yielded". Suggest changing this to "resulting in growth of the simulated spherical particles to maximum diameters..."

P5901/L19:" …resulting in growth of the simulated spherical particles to maximum diameters…"

P5902/L1-2:"…simulation results in growth of compact spherical NAT particles with maximum diameters lower…"

P5903-L1: A small temperature bias can have a considerable effect on particle growth. What would be the effect of a reasonable uncertainty in the CLaMS ERA-Interim temperatures on the eventual particle sizes? What about temperature fluctuations experienced along the trajectory? Errors in ambient HNO3/H2O? Could these act to reduce the discrepancy in the modeled and FSSP particle sizes?

Considering the compactness of the CLaMS particle backward trajectories shown in Figure 2 and the steady temperature increase we do not expect considerable changes in the modelled size distribution when a small temperature bias, temperature fluctuations or errors in the ambient HNO_3 and H_2O mixing ratios are considered. To support the conclusion that the observed larges particles cannot be compact NAT spheres, we added the size distribution obtained from a sensitivity simulation with 1 K lower temperature.

These effects should be ruled out before looking for more complex explanations. Did you compare the MIPASSTR and ERA-Interim temperatures? There is also the question of the nucleation mechanism. Have alternatives been considered? What about the NAT nucleation rate? These issues must be addressed adequately.

As discussed above, the focus of our work is on settling velocities and our aim was not to investigate the influence of the manifold other parameters affecting the simulation. Regarding the influence of other parameters on simulated HNO_3 redistribution we refer to the work of Grooß et al. (2014) and ongoing work in the RECONCILE framework and beyond. We worked out the speculative statements more clearly and mentioned that other parameters also have important influence on the simulation.

P5903-L6-7: /ppbv/ [ppmv]
Done.

P5903-L18-25: The measurements from MIPAS-STR should be explained in more detail, such as the uncertainties on the retrieved quantitities. Without this information the reader is unsure what significance to attribute to "excess HNO3" etc. Please quote some HNO3 values for the maxima and excess rather than leaving it to the reader to work these out from the plots. It will also help understand the comment below about P5904-L7.
P5903/L19"…17.0 km. In sections A and B maximum volume mixing ratios of HNO_3 of 8.5 ppbv are found compared minimum values below 6.5 ppbv around. The maxima in section B_1 to B_3 peak at about 10 ppbv above flight altitude and 11 ppbv at 16.5 km compared to minimum values around 7 ppbv in between. Another strong maximum is observed in section C peaking at 15.5 to 16 km altitude and shows enhanced HNO_3 mixing ratios of 10.5 ppbv compared to minimum values of less than 6 ppbv around."
We furthermore refined the discussion of Figure 6 (HNO_3 cross-section extracted from CLaMS) at P5906/L3f: "while CLaMS produces higher HNO_3 mixing ratios and additional local maxima above. The HNO_3 enhancement found in the MIPAS-STR results for the scans B_1 to B_3 around and above flight altitude is also reproduced well, while the maximum around 16 km is only weakly identified in the simulation."
The uncertainties of the MIPAS-STR retrieval results are addressed by an additional Figure (new Fig. 6; other Figure numbers updated accordingly) showing retrieved temperature and HNO_3 together with estimated errors and vertical resolution for the single limb scan at 08:32 UTC.
After P5903/L22 we added: "…approach T_{NAT}. Typical profiles retrieved from the MIPAS-STR observations, estimated uncertainties and vertical resolutions are shown in Figure 6. For the temperature retrieval (Fig. 6a), typical vertical resolutions in of 2 to 3 km were obtained, while the estimated 1σ-uncertainties are typically 0.7 to 0.8 K (slightly higher vertical resolutions were obtained in the other flight sections). For continuum absorption two distinct maxima are found peaking at 17.5 and 19 km, indicating two different PSC layers. The typical vertical resolution obtained is about 1km. It is pointed out that the alignment of the PSC layers in the horizontal direction along the line-of-sight is uncertain.
Also shown is the corresponding profile of calculated T_{NAT} considering the nominal values for HNO_3 (MIPAS-STR) and H_2O (FLASH-A smoothed) together with the same profiles considering biases of +10% for the volume mixing ratios of these gases. The retrieved temperature is equal to T_{NAT} at about 16 km altitude and supersaturated conditions are found above. The continuum absorption maximum associated to the lower PSC layer peaks 1.5 km above, indicating that supersaturated conditions are also present at regions with weak continuum absorption (i.e. cloud-free). The retrieved temperatures above 16 km are below T_{NAT} by up to about 2.5 K. The shifts in T_{NAT} due to enhanced HNO_3 and H_2O are comparable to or below the uncertainties of the retrieved temperatures.
Figure 6b shows the retrieved profiles of HNO_3 with typical 1σ-uncertainties of 10 % and a high vertical resolution of about 1 km above 15 km altitude. The corresponding HNO_3 profile from the CLaMS standard scenario is shown for comparison together with simulated passive NO_y* (i.e. considering no condensation and vertical redistribution of HNO_3). The comparison shows that the maxima observed by MIPAS-STR peaking at about 16 and 18 km are reproduced well, reminding that vertical fine structures in the order of 1 km and localised profiles are considered. The simulated peak values are each by about 2 ppbv higher than the retrieved values and the lower maximum is significantly shifted towards lower altitudes by 0.5 to 1.0 km. Considerably higher simulated HNO_3 mixing ratios compared to the observation are found above 18 km. While even fine structures are reproduced well to by the simulation, the observed discrepancies of the individual profiles are primarily attributed to the complex PSC scenery observed."P5903/L23-25 was modified according to: "Based on the assumption that the observed HNO_3 maxima had just evolved or were still developing, the results shown in Figures 5 and 6 strongly support denitrification by particles composed of NAT."

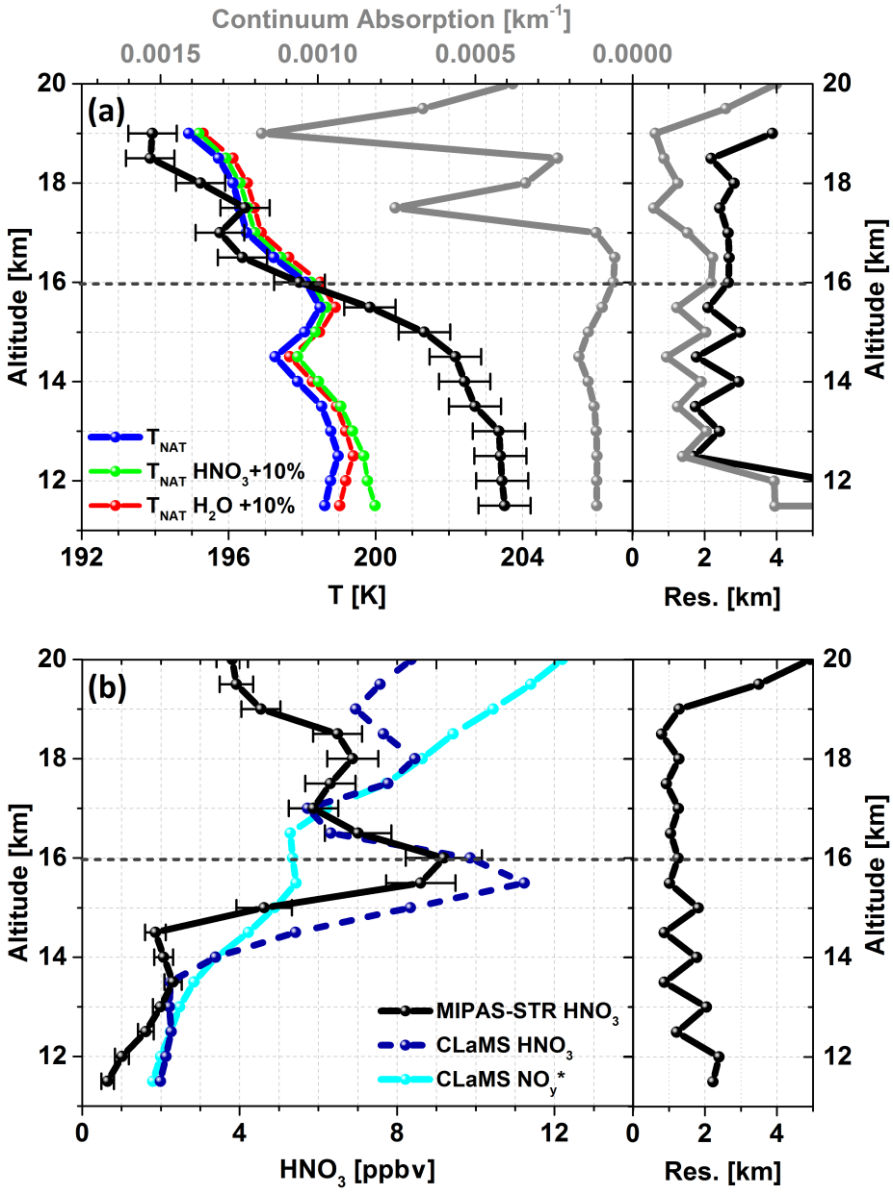

Fig. 6: Upper panel (a): Temperature (T) and wave-number independent background continuum absorption (compare Fig. 4) retrieved from MIPAS-STR for the limb scan at 08:32 UTC. Corresponding profiles of T_{NAT} were calculated considering retrieved temperature and HNO_3 in combination with the smoothed H_2O profile from FLASH-A (alternative profiles considering increased HNO_3 and H_2O also indicated). Lower panel (b): Corresponding profiles of HNO_3 retrieved from MIPAS-STR and extracted from CLaMS together with simulated passive NOy*. Res. Corresponds to vertical resolution of the retrieved profiles and error bars to estimated total 1σ-uncertainties of the retrieval results. Grey dotted lines indicate altitude with T equal to T_{NAT}.

P5903-L13: Here you are invoking effects of retrieval uncertainties and horizontal gradients without giving any indication of their magnitudes.

P5903/L14-16: "….direction. For example, the PSC layer might be located in a colder region along viewing direction, while warmer temperatures in other sections along viewing direction might lead to a warmer net temperature retrieved. Furthermore… of H_2O (compare Khaykin et al., 2013) and retrieved HNO_3. The sensitivity of calculated T_{NAT} towards variations in the mixing ratios of these gases is shown in Figure 6 and can serve as an estimate for the impact of horizontal gradients."

P5903-L13: Uncertanties in HNO3 also affect the calculated TNAT.
See new Figure 6.

P5904-L6-7: the [continiuum] retrieval: You need to explain why the continuum is not sensitive to large NAT/low number density. Although I think your interpretation is incorrect if the amount of HNO3 in the NAT particles is significant (say around 1 ppbv or more?). I would expect this amount of HNO3 uptake into large NAT particles to generate detectable mid-ir emission.
We wanted to express that the continuum retrieval would be not/hardly be sensitive to very few large NAT particles (i.e. less than indicated by the in situ observation shown in Figure 1) falling out of the PSC above (where the in situ observation was performed and the optically dense PSC was detected by MIPAS-STR, compare Fig. 1 and Fig. 4). This is clarified in the modifications given below.

P5904-L8-10: "large NAT particles falling ... dense PSC cloud ... increased opaqueness". The wording here would appear to contradict your earlier assertion that the mid-ir is insensitive to large NAT/low number density particles. To what do you attribute the cause of the increased opaqueness? Are you talking about the large NAT falling out of a mixed phase STS/NAT cloud or from a population of large/small NAT? Is it cold enought to form STS? It is not clear from your description.
We wanted to express that the observed scenery would be compatible with a dense PSC composed of NAT and STS (considering that the existence temperature of STS is only about 2 K lower than T_{NAT}) present around flight altitude and above, resulting in enhanced continuum absorption in the MIPAS-STR observations and the size distribution detected by the FSSP-100 (Figure 1). In contrast, only very few large particles falling out of the optically dense PSC layers would result in weak or insignificant continuum absorption in the MIPAS-STR observations below flight altitude.
Figures 4 and 1 indicate that the flight path in section B and the discussed particle observations were situated at the lower edge of the PSC. The large particles observed sizes would be expected to sediment several hundreds of meters per day (compare Pruppacher and Klett (1997); Fahey et al. (2001)) and therefore could sediment to lower layers under supersaturated conditions. We furthermore mention that the FSSP-100 started detecting potential NAT particles around 17 km altitude (i.e. below the PSC detected by MIPAS-STR)
We replaced P5904/L4-10 by: “However, considering the in situ observations of large potential NAT particles with sizes of tens of µm (Fig. 1) at the lower edge of the PSC (Fig. 4, section B) and potentially capable of settling several hundreds of meters per day (compare Pruppacher and Klett, 1997; Fahey et al., 2001), the shown scenario would be compatible with very few large NAT particles falling out of a dense PSC around flight altitude and above (e.g. mixed phase PSC containing STS and NAT) and evaporating at altitudes with temperatures above T_{NAT}. This is furthermore supported by the fact that the FSSP-100 started detecting potential NAT particles already around 17 km altitude during the ascent phase and below the PSC detected by MIPAS-STR in this region (compare Figure 4, sections A and B).”

P5906-L16-18: Essentially you are saying that trying to match up individual vertical profile comparisons is not possible, but never mind because over the vortex the ensemble profile is more reliable?
P5906/L16-18: “While quality of the agreement between the MIPAS-STR observations and CLaMS simulations varies for individual profiles and subsections, the corresponding ensemble profiles for entire flights cover significant parts of the polar vortex allow for more meaningful comparisons.”

P5910-L7: give the dimensions [of] potential
Done.

Technical modifications

We adopted consistently “Figure” in the running text and “Fig.” for text in brackets and in figure captions.

P5898/L3 and P5917/L3: Reference Woiwode (2014) was corrected to Woiwode (2013)

The reference Molleker et al. (2014) was updated.

At P5912/L9 we omitted “on 25 January 2010” since the date of the flight is clear from the previous sentence and from the context.