Author response to Editor Decision and Reviewer Report for: "Denitrification by large NAT particles: the impact of reduced settling velocities and hints on particle characteristics"

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Dear Editor,

Dear Referee #2,

we thank you very much for the thorough revision and your suggestions for improving our manuscript. We followed the recommendation of Referee #2 and performed more simulations in order to strengthen the conclusions of our work. We furthermore tried to elaborate our working hypothesis more clearly: The simulated maximum particle masses might be close to the reality, while their maximum sizes might be larger than those of compact spheres, supporting the assumption of reduced settling velocities. Moreover, we worked out more clearly that the focus of our study is on the impact of reduced NAT particle settling velocities on vertical HNO_3-redistribution rather than on the physical appearance of the particles.

As suggested by the Referees, we performed further sensitivity simulations considering temperature biases and modified nucleation rates. While the implementation of the alternative nucleation mechanisms like NAT nucleation on ice and vice versa is out of the scope of our work, we briefly discuss potential effects of these processes in the context of this study.

In the revised manuscript we provide additional figures including the CLaMS size distributions corresponding to all sensitivity simulations, the retrieved vertical cross-section of temperature (MIPAS-STR) associated to the flight on 25 January 2010 and the MIPAS-STR observation geolocations and meteorological conditions for the flight on 30 January 2010. We furthermore included the retrieved vertical cross-section of HNO_3 for the flight on 30 January 2010 and the vertical cross-sections of HNO_3 corresponding to all simulations and for both flights. Additionally, we compare the ensemble profiles from all simulations with the MIPAS-STR observations.

We hope that the revised discussion will help showing that (i) ice-containing particles were unlikely to be present during the observations on 25 January 2010, (ii) the large particles observed were unlikely to be spherical and compact particles at the same time and (iii) that the impact of reduced settling velocities on HNO_3-redistribution is significant.

In the revised manuscript we used an updated CLaMS setup which considers the JPL 2011 recommendation for reaction rate coefficients and further minor updates of the model. Therefore, the model results included in the revised manuscript slightly deviate from the corresponding results in the prior version of the manuscript, while the overall conclusions remain unchanged.

We hope that the revised version of our manuscript will meet the acceptance of the Editor and the Referees and we are open for further improvements. Below, we provide specific responses to the points addressed by the Editor and Referee #2 (responses in bold letters). We furthermore provide the revised manuscript in the attachment. Substantial changes compared to the previous version of the manuscript are highlighted in blue, while minor changes (e.g. modified wording / updated discussion) are not marked.

With best regards,

Wolfgang Woiwode on behalf of the coauthors

Response to Editor Report:

Dear authors,
please find enclosed the report from referee 2. Unfortunately, the referee was not satisfied with the revisions of the manuscript. The major criticism from the referee is that the simulated HNO_3 distribution depends critically on the other parameters which are still not properly addressed in your work and that this forces you to "speculate" about underlying causes of the discrepancy between observations and model. The referee also requests, as already done in his previous report, more investigations, e.g model simulations. Further, referee 2 states that "as the work stands almost nothing else has been taken into consideration in the simulations and therefore that only one "consistent explanation" (for which there no actual physical evidence) has been explored."
In my opinion the criticism by the referee is justifiable. However, I do not think that you necessarily need to perform further model simulations, you rather should discuss more the possible influence of the other factors and state more clearly that you have one theory here that is tested. Some more detailed comments will follow below:
Following the recommendation of the referees, we performed additional simulations considering temperature biases of +/- 1K and the nucleation rates multiplied by factors of 0.5 and 2.0. We agree that these simulations give valuable information on the relative consequences of changes in these parameters and hope that the additional simulations help improving our manuscript. We furthermore briefly discuss the potential influence of NAT nucleation on ice and vice versa and potential temperature fluctuations along the trajectories on the results of our study.

Abstract:
How does the reduction of the settling velocity agree with reality? Later you explain it, but in the abstract the explanation is missing and would be worth to be included here as well. Other questions that came to my mind when reading the abstract was: Are the calculated settling velocities wrong so that one needs to reduce them? Isn't what you are doing just another way to "tune" the model to the desired results? Doesn't the discrepancy just mean that the size distribution of the the NAT particles derived from the model is incorrect due to the processes that have not been considered as stated by referee 2?
We find that in the context of our study moderately reduced settling velocities slightly improve the agreement of simulated and observed HNO_3-redistribution, while strongly reduced settling velocities are not compatible with our results. The combination of observations and simulations for the flight on 25 January 2010 shows that the observed particles were unlikely to be spherical and compact NAT particles at the same time and that ice-particles are an unlikely option. Non-compact or aspheric particles offer an explanation for the large observed sizes, and in both cases reduced settling velocities have to be expected. Therefore, we think that reduced NAT particle settling velocities might have the potential to better resemble the settling properties of real NAT particles.

P4, L21-L25:
Do I get it right that you use the reduced settling velocities to overcome the problem that in the model spherical particles are assumed though the particles in reality can be aspherical?

We simulated reduced settling velocities of NAT particles to investigate how NAT particles with alternative characteristics would influence the simulated HNO_3-redistribution. While reduced settling velocities clearly have to be expected for non-compact and aspheric particles, conclusions from our study on the actual physical appearance of the particles remain speculative.

P7, L23:
In the model one of the latest parameterizations for NAT nucleation is considered. However, in the model no ice formation is considered and thus you cannot consider formation of NAT on ice or vice versa ice on NAT which also could lead to the large particles measured by the FSSP. I do not expect you to include ice formation in the model to calculate other possible formation mechanisms, but you at least should discuss these processes and the possible influence they could have on the results of your study. Further, other studies discussing PSC formation during that winter (Khosrawi et al., 2011; Pitts et al., 2011) show that ice was frequently observed during mid of January and that a warming occurred around 24 January, so that the probability for ice on 25 January may have been low.
We included a short discussion of the potential effects from these alternative nucleation processes in Section 2. Our study however does not support the presence of ice particles during the flight on 25 January 2010 due to too warm temperatures (compare particle backward trajectories in Figure 2 and retrieved vertical cross-section of temperature in new Figure 4). We furthermore added the suggested references along with Dörnbrack et al. (2012) to support the low probability for ice during this flight.

P8, L27:
I am quite skeptical that the difference in the settling velocity of spherical and aspherical particles can cause such a large difference between measurements and observations.
We worked out more clearly that we suggest that the simulated maximum sizes in terms of particle masses might be close to the reality and that the discrepancy between the observed and simulated size distribution might be due the physical appearance of the particles. If the sizes corresponding to the simulated particle masses were expressed in terms of diameters of spheres with lower mass densities or the maximum extensions of considerably aspheric particles, similar maximum sizes compared to the sizes inferred from the observations could be obtained. Therefore, the largest particle sizes indicated by the observations might correspond to the masses of the largest simulated particles. We hope that this aspect is worked out more clearly in the revised abstract, discussion and conclusions.

P11, L21:
The denitrified air masses above have not been measured by MIPAS-STR and this should be clearly stated. That denitrification/renitrification occurred during that time period is shown e.g. in Khosrawi et al. (2011) in Figure 4 using measurements from MLS. It would be useful to refer to this figure to confirm the measurements by MIPAS-STR.
Done.

P17, L19:
What are the growth rates of spherical/aspherical particles? Which of these particles grow faster. Can NAT particles really grow to such large sizes as measured by the FSSP? These have been larger

than the ones by Fahey et al and such sizes have not been measured before. How do you know that these particles are real and that these are really NAT and not ice particles?
In our study we did not modify the particle growth rate. We assume that the overall model parameterization is close to the reality and focus on the parameter settling velocity. We think that the sizes of the largest particles observed on 25 January 2010 might be reconciled with the simulations, if the observed particles had similar masses like the simulated particles but larger maximum extensions for the reasons mentioned above. Accordingly, we do state that a compact spherical 30 µm NAT particle might arise if the parameter settling velocity is adjusted. We rather suggest that a simulated particle with a mass equivalent to a compact spherical 10 µm particle might have maximum extensions of about 30 µm (height or diameter) if it was non-compact or considerably aspheric. And if the particles should be non-compact or aspheric, they are expected to have reduced settling velocities compared to compact spheres. While our study shows that reduced settling velocities are compatible with our observations, we however can only speculate on the actual physical appearance of the particles.

Figure 1 and model/observation comparison:
What does the FSSP measure NAT particles or all type of PSC particles? How can you be sure that the large particles measured by the FSSP are NAT particles? What are the uncertainties of the FSSP? There is a large discrepancy between the size distributions and this discrepancy cannot be solely explained by the differences in the settling velocities of spherical/aspherical particles. Other processes must play a role.
Regarding the uncertainties of the FSSP observations and their sensitivity towards different particle types we refer to Molleker et al. (2014). For the evaluation of the FSSP-100 measurements the refractive index of NAT was adopted in order to provide realistic sizes of NAT particles. A composition of NAT is supported (i) by the temperatures along the CLaMS particle trajectories well above T_{ICE} and (ii) by the retrieved temperatures of MIPAS-STR indicating temperatures below T_{NAT} but above T_{ICE} for the flight on 25 January 2010. While STS particles are not expected to be capable of growing to the large observed sizes, other particles (e.g. NAD) cannot be ruled out. However, there is only little evidence in the literature for other particles than the discussed cases capable of growing to such large sizes.
While the scattering properties of moderately aspheric particles can deviate strongly from spherical particles (Bormann et al., 2000), a quantitative assessment of the uncertainties of the FSSP observations in context of strongly aspheric particles is not known to us.
We suggest that the largest simulated and the largest observed particles might have approximately the same masses, while the large sizes observed are a consequence of non-compactness or aspheric shape. As mentioned above, we however focus on the consequences of reduced settling velocities as a potential consequence of alternative particle properties, while conclusions on the physical appearance of the particles remain speculative.

Figure 7.
This figure should also be included for the CLaMS simulations with reduced settling velocity.
Done.

Figure 8:
From this figure I would say everything except a settling velocity of 30% looks like a good agreement between measurements and model simulations. However, your conclusion is the opposite.
We worked out more clearly in the conclusion that the results considering relative settling velocities between 50% to 100% result in good agreement. For the flight on 30 January 2010 the shape and scattering of the observed overall HNO_3-redistribution in the ensemble profile is however best reproduced by the 70%-scenario. This aspect resulted more clearly in the updated CLaMS simulations considered in the revised manuscript.

Response to referee #2:

As the authors themselves state, the simulated HNO_3 distribution depends critically on the other parameters which are still not addressed in the present work. Therefore, I do not see that the revised manuscript has addressed the major concern raised in the previous review. This forces the authors to "speculate" about underlying causes of the discrepancy between observations and model. With more investigation (as previously outlined) the conclusion leading to acceptance of reduced NAT sedimentation velocities as a viable explanation could be made.

The authors also state that "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". However, the main point is that as the work stands almost nothing else has been taken into consideration in the simulations and therefore that only one "consistent explanation" (for which there no actual physical evidence) has been explored.

We thank the Referee for pointing out the additional value of more alternative simulation scenarios. Following the suggestion by the Referee, we performed additional simulations considering temperature biases of +/-1 K and the NAT nucleation rates multiplied by factors of 0.5 and 2.0. We agree that the results provide valuable information on the relative effects of the other parameters being changed and hope that the additional simulations and the revised discussion will help improving our manuscript.

Further Comments:

- **We now consistently use the term "renitrification" instead of "nitrification" to indicate HNO_3-enhancements attributed to evaporation of NAT particles. To our best knowledge both terms are used in the literature. However, we think that "renitrification" is commonly accepted and preferred.**
- **In Section 4 we added a personal communication by P. Achtert (MISU, Stockholm University, Sweden), saying that PSCs classified as NAT were observed on 24 January 2010 above Esrange (Kiruna, Sweden) around 18 km altitude.**
- **We subdivided Section 5 into three subsections for more clarity.**
- **In Figures 2 to 6, 8 and 10 we replaced "Woiwode, 2013" by "Woiwode PhD thesis, 2013" for clarification.**