

Interactive comment on “Denitrification and polar stratospheric cloud formation during the Arctic winter 2009/2010” by F. Khosrawi et al.

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We thank reviewer 1 for the constructive, helpful criticism. We followed the suggestions of reviewer 1 and revised the manuscript. The discussion on many major aspects of this paper has been significantly improved by taking into account Aura/MLS measurements and WRF simulations. In the frame of the revision two figures have been added.

Major concerns:

The discussion of many major aspects of the paper is superficial

We improved our discussion according to the comments given below.

It is not clear why trajectory calculations have been included at all since (1) only few

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examples have been shown at all and (2) the main analysis from which conclusions are drawn (i.e. where temperatures below T_{ice} appeared in relation to CALIPSO and ODIN measurements) could be based on T-analysis data alone.

The intention of this study is to investigate denitrification together with PSC formation. If one only applies T-analyses one only knows when and where temperatures below T_{ice} occurred but not when and where an air parcel passed these areas and if PSC have been formed or not in these areas and especially under which conditions these PSCs have been formed. The physical state of PSCs in the stratosphere is strongly controlled by the temperature history of the air mass as has been shown in earlier studies by e.g. Tabazadeh et al. (1995), Larsen et al. (1997) and Toon et al. (2000). It is correct that we only show few examples of the trajectories and simulations. Though not shown explicitly these trajectories/simulations results are unambiguous for this study and are described and applied at several places in the paper. For example Fig. 8 (now Fig. 9) would have not been possible to be created without the trajectory analyses. To refer more to the trajectory analyses and model simulations we included text parts as given in the answers to the detailed comments.

The mechanism of denitrification through the formation of NAT ‘mother clouds’ by mountain waves (Fueglistaler et al., ACP, 2002; Dhaniyala et al., GRL, 2002; Mann et al., JGR, 2005) is not mentioned at all. However, this process might be rather important in winter 2009/10 since in the first half of January there has been major mountain wave activity with formation of high-number density NAT-clouds as described by Pitts et al., ACP, 2011. It is necessary that this mechanism is discussed in relation to possible denitrification caused by the synoptic sub- T_{ice} temperatures in the second half of January.

We agree that this mechanism is an important one for denitrification and should be mentioned as well. We included the following text in the introduction: *In connection with mountain waves it was suggested that NAT clouds could form on mountain wave ice particles and that these NAT clouds could serve as “mother clouds” producing a small number of large NAT particles that could sediment out and cause denitrification (Fueglistaler et al.,*

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2002, Dhanilaya et al. (2002), Mann et al. (2005). For the removal of HNO_3 by NAT clouds that were formed by mountain waves in the first half of January this mechanism could be indeed the responsible one. In fact, very low HNO_3 values were observed by Odin/SMR on 5 January around Greenland shortly after mountain wave PSCs were formed. We included the following sentence in section 4.4. and the conclusion: *In contrast, for the denitrification that was observed between 9–15 January sedimenting NAT particles that formed on mountain wave ice PSCs as it was discussed by Fueglistaler et al. (2002), Dhaniyala et al. (2002) and Mann et al. (2005) could be a possible mechanism.* However, another removal of HNO_3 occurred during the second half of January due to the synoptic cooling and subsequent ice cloud formation over the Arctic sea. During that time period denitrification was observed with accompanying dehydration that indicates that nitric acid containing ice particles have sedimented. Thus, ice formation on NAT and sedimentation of these particles could be a possible process for the observed denitrification during that time period.

Detailed comments:

p.11383, l. 12: *In an introduction on denitrification in the Arctic, the papers by e.g. Fueglistaler et al., ACP, 2002; Dhaniyala et al., GRL, 2002; Mann et al., JGR, 2005 on the denitrification caused by large NAT particles which originate from high number density NAT clouds ('mother clouds') formed by nucleation on mountain wave ice particles should be mentioned.*

We agree that this process together with the according references should be mentioned in the introduction. We included the following text: *In connection with mountain waves it was suggested that NAT clouds could form on mountain wave ice particles and that these NAT clouds could serve as "mother clouds" producing a low number of large NAT particles that could sediment out and cause denitrification (Fueglistaler et al., 2002, Dhanilaya et al., 2002, Mann et al., 2005).*

p.11385, l. 15: *Could you give the information on the horizontal and vertical resolution of the CALIPSO PSC observations which are, due to averaging, not identical to the original*

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single shot measurements?

The data are averaged to a vertical resolution of 180 m and a horizontal resolution of 5 km. The following sentence has been included: *For the PSC analyses, the CALIPSO profile data are averaged to a resolution of 180-m vertical and 5-km horizontal.*

p.11386, l. 16: *closing bracket missing*

We inserted the missing bracket.

p.11388, l. 3: *'a five-day period ending on 19 January' Pitts et al., 2011 states that the period with synoptic temperatures below T_{ice} is 15-21 Jan. (At least on 20 Jan there was still a large area below T_{ice} according to ECMWF temperature analysis.)*

Thanks for pointing this out. It should indeed read 21 January as in the Pitts et al. (2011) paper.

p.11389, l. 10, 19: *The translation of potential temperatures to absolute altitudes is not correct for a typical Arctic winter atmosphere: 450K \approx 18 km (instead of 19 km), 575 K \approx 23 km (instead of 27 km), 465 K \approx 19 km (instead of 20 km), 585 K \approx 23 km (instead of 28 km)* We applied the Taylor series expansion ($z = 0.0635 * \Theta - 9.22$) for converting potential temperature to altitude. However, we agree that Crutzen and Freie's rule of thumb ($z = \Theta / 25$) is probably giving the more accurate conversion for the typical Arctic winter atmosphere. The altitudes have been changed accordingly.

p.11389, l. 15-19: *'The denitrification observed in January 2010 was also the strongest denitrification observed in the entire Odin measurement time period' and Fig. 3: This statement is not obvious and the discussion of the Figure is much too short. What are about other winters with similar low HNO_3 values as 2009/10? E.g. 2007/08 and 2004/05 at lower altitudes and 2006/07 at higher levels? Further, from these plots the other winters seem to have similar low values in e.g. February which does not support the statement that denitrification in 09/10 has been much stronger than in other winters. (There are lowest values at several levels in*

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mid-Jan 2010, but these are not signals of pure denitrification, but also contain the reversible uptake from the gas phase into particles). The effect of dynamics should be further discussed. Correlation plots of HNO₃ with a dynamical tracer to prove and possibly quantify the extent of denitrification in 2009/10 might help.

To discuss the figure more and also to point out the low HNO₃ values during the other cold winters we included the following text: *Though also very low HNO₃ mixing ratios are found during the cold winters 2004/2005, 2006/2007 and 2008/2009, these low values are not found over all potential temperature levels between 465 and 585 K as is the case for the winter 2009/2010. Further, from deriving the same time series as shown in Fig. 2 for the other cold winters we still do not see as low HNO₃ values as in the winter 2009/2010 at the end of the winter.*

Why are the MLS values of gas-phase HNO₃ during the winter months 1991-98 higher than those of Odin/SMR between 2001 and 2010? Has the Arctic stratosphere during all winters 1991-98 been warmer or the vortex weaker than in 2001-2010?

The difference between Odin/SMR and UARS/MLS are caused by systematic differences of the two instruments as has been discussed by Urban et al. (2009). Systematic differences are e.g. that the measurements of Odin/SMR and UARS/MLS are performed with different vertical resolutions (1.5-2 km for Odin/SMR and 6 km for MLS) as well as for slightly different altitude ranges, with Odin/SMR reaching higher up and UARS/MLS lower down in the stratosphere. Generally, it was found that Odin/SMR has systematically slightly lower values (2-3 ppbv) than UARS/MLS. We included the following text to explain this: *The UARS/MLS climatology shows generally somewhat higher HNO₃ mixing ratios than the Odin/SMR observations in the Northern Hemisphere high latitudes. This is due to systematic differences between these two instruments as discussed by Urban et al. (2009). Odin/SMR measures with a much higher vertical resolution than UARS/MLS (1.5-2 km compared to 6 km). Further, the altitude ranges of measurements by these two instruments are slightly different.*

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p.11393, l. 1: *Why are these trajectories selected here? One should discuss all (or at least a major part) to get an impression on similarities/differences between the ground-based PSC observations.*

We chose the trajectories that were started on 23 January since this was one of the days where the most pronounced PSCs were measured with the lidar and where also a PSC at the end and the begin of the box model simulations was found. The lidar measurements are described in section 3.3. Comparison of all our box model simulations show good agreement with the ground-based lidar measurements. We included the following text in section 4.1: *Generally, the air masses were 6-days backward in time originating from Greenland, northern Scandinavia or Northern Russia and were then transported over the Arctic sea, passing via the North-pole and Greenland or Canada to Kiruna. Further below the following sentence has been included: We chose this date since the lidar measurements were most pronounced during this time period and we will discuss the corresponding box model simulations in more detail in section 4.2. As well as at the end of the section: PSC formation in the area of Kiruna and in the area north of Scandinavia was found in almost all box model simulations that were performed along trajectories that were started from Esrangle (17–24 January) and in the majority of box model simulations along the back trajectories that were started from IRF Kiruna between 22–24 January. To discuss our box model simulations of PSC formation in the area of Kiruna and in the area north of Scandinavia and to compare these simulations with the ground-based and space-borne measurements, we chose the back trajectory that was started on 23 January from Esrangle at 19:00 UT at 22 km (blue line in Figure 5). Additionally, the following text has been included in section 4.2: The PSCs that were simulated in the area of Kiruna were compared with the lidar measurements at Esrangle and IRF Kiruna, respectively. A good agreement was found for all back trajectories. The PSCs were most pronounced both in the lidar measurements and box model simulations between the 22 and 24 January.*

p.11393, l. 4: *'was originating'* This is very arbitrary since it depends on the time of the backward trajectories.

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We agree that the origin of the air mass is dependent on the time the trajectory has been calculated backward. The changed the sentence as follows: *The air mass at 19 km was originating 6-days backward in time from Russia, while the air mass at 22 km was originating 6-days backward in time from the sea north of Scandinavia, between Svalbard and Novaya Zemlya and the air mass at 25 km originated 6-days backward in time from Greenland.*

p.11393, l. 7: ‘was transported furthest’ *Is this really the case? Have you determined the absolute length?*

We meant that the trajectories have different geographical origin and that this one was coming from the geographical location furthest away. We changed the sentence as follows: *From Fig. 4 (now Fig. 5) it can be seen that the air mass at 25 km was transported from the geographically furthest location during the 6-days compared to the air masses at 19 and 22 km.*

p.11395, l. 25: *Why are the temperatures on the (restricted number of) trajectories used for such kind of analysis? I do not see that the information that CALIPSO has observed ice along the trajectories is used to analyse more closely the ground-based lidar observations for which the trajectories have been calculated. In this entire paragraph the analysis could be performed based on temperature analysis fields alone.*

The analysis could not have been performed on the basis of temperature analyses alone. To understand how the PSC that were measured with the ground-based lidar have formed one needs to know the temperature history of the air mass (see answer above and according references). We need the combination of box model simulations and ground-based and space-borne measurements to understand PSC formation and by which PSC particles the denitrification was caused.

p.11396, l. 3: ‘temperatures below T_{ice} were caused by waves on 2 January’. *Could you discuss here the resolution of the model you’ve used for the trajectories and how well mountain waves can be resolved by the model?*

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HYSPLIT trajectories were calculated based on GDAS analyses. The horizontal resolution is $1^\circ \times 1^\circ$ which is not sufficient for resolving mountain waves. However, from the analyses by Pitts et al. (2011) and Dörnbrack et al. (2011) as well as WRF simulations that were performed in the frame of this study for the 2009/2010 winter we know that mountain waves were generated over Greenland. We changed the sentence in section 3.4 as follows to give the information on the resolution of the meteorological analyses used in the HYSPLIT model: *Analyses are provided four times a day (00:00, 06:00, 12:00 and 18:00 UTC) with a horizontal resolution of $1^\circ \times 1^\circ$ on 23 pressure levels (1000 to 20 hPa). The following sentence was included on p11396 to explain why we know that the occurrence of T_{ice} was due to mountain waves: *Simulations with the WRF model and the studies by Pitts et al. (2011) and Doernbrack et al. (2011) showed mountain wave activity with subsequent ice PSC formation, respectively, around Greenland in the beginning of January.**

p.11396, l.12: ‘Further, the PSC formation north of Scandinavia agrees spatially and locally quite well with the area where denitrification was observed by Odin/SMR’ *This is not right: Odin/SMR observes only the missing HNO_3 in the gas phase: this might be due to uptake into the PSC and due to denitrification. It cannot be decided how strong the denitrification has been when particles are still present.*

We agree. In that sentence we replaced “denitrification” by “ HNO_3 removal”. The following sentence has been changed as follows: *Thus, from this coincidence we suggest that ice formation on NAT particles could have occurred during that particular winter and that subsequent sedimentation of these particles could have caused the denitrification that was observed between 15-21 January.*

p.11396, l.13: ‘Thus, from this coincidence we suggest that ice formation on NAT particles with subsequent sedimentation of these particles caused the denitrification as observed by Odin/SMR’. *This argumentation is not convincing: strong HNO_3 removal has also been observed by ODIN on 9 and 13 Jan (see Fig. 1). As can be seen from Fig. 2, lowest vortex mean HNO_3 values are reached before mid-January, i.e. before the period of synoptic sub-ice*

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temperatures starting on 15 Jan. Further, even on 15 Jan, lowest HNO_3 values are also visible in the region NE-of Novaya Zemlya where no temperatures below T_{ice} are visible in ECMWF analysis maps.

This is correct. HNO_3 -removal as well as denitrification/re-nitrification is observed on 9 January as well as between 13-15 January in Odin/SMR and Aura/MLS observations, respectively. Thus, before temperatures cooled synoptically down. This text part has been revised as given in the answer to the previous comment. We discuss now renitrification by taking into account Aura/MLS observations. We added to section 3.2 showing the temporal evolution of the Aura/MLS and Odin/SMR HNO_3 observations as function of pressure. The following paragraph has been included to discuss the Aura/MLS and Odin/SMR HNO_3 observations: *If permanent removal of PSC particles and thus denitrification occurs, HNO_3 is vertically redistributed. Thus, the air below the denitrified layer is renitrified (Kondo et al. 2000, Irie et al. 2001, Dibb et al. 2006). Renitrification cannot be observed by Odin/SMR since measurements do not reach to sufficiently low altitudes where renitrification usually is observed. To overcome this we use Aura/MLS measurements. Aura/MLS and Odin/SMR measurements are very similar as has been shown in recent validation studies (e.g. Santee et al. 2007). However, geographical and temporal sampling as well as the altitude resolution are different and some differences can therefore be expected. The MLS measurements of HNO_3 versus equivalent latitude for the 5, 9, 13, 15 17 and 19 January show that very low HNO_3 values are found between 22-26 km (not shown). The accompanying renitrification that can be seen during these days suggests that the air mass has been denitrified. Figure 3 shows the temporal evolution of temperature from ECWMF, HNO_3 from Odin/SMR and Aura/MLS as well as N_2O from Odin/SMR at high equivalent latitudes (70-90°N) as function of pressure. Potential temperature levels are given as grey lines. Temperatures below 195 K occurred in the altitude region between 480 and 700 K from mid-December to end of January. The Odin/SMR observations show that from the beginning of January onwards gas phase HNO_3 is removed. After a minimum in the first half of January gas-phase HNO_3 remains low until the end of January. Higher values are observed in the beginning of February and throughout March between 500 and 600 K. In the overlapping pressure range, a similar picture*

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is provided by Aura/MLS. The maximum values of HNO_3 in December are slightly higher in Aura/MLS while the HNO_3 maxima in March are slightly lower. In the Aura/MLS data distinct HNO_3 minima are visible between 480 and 600 K, one in the beginning of January, one in mid of January and a weaker one in the end of January. Below these distinct minima, distinct maxima are found indicating redistribution of HNO_3 . A long tongue of gas phase HNO_3 is observed between 420 and 480 K which is slowly transported downward during the first half of January and persists until mid February. The downward transport is due to subsidence of air in the polar vortex as also indicated by the time versus pressure cross section of the inert tracer N_2O measured by Odin/SMR.

p.11396, 1.24: 'showed that the air masses were dehydrated during 16 to 19 January' A personal communication is rather weak. Is there a paper or could the measurements be discussed here a bit in more depth? What was exactly measured by the balloon? Only gas-phase H_2O or gas-phase+particulate? What is the accuracy of these measurements? What does ODIN/SMR measurements tell about dehydration?

We agree that a personal communication is rather weak. However, at the time of submission we had not more than this. Sergey Khaykin is working on a publication of his results concerning dehydration in the Arctic winter 2009/2010. We included now the reference to his manuscript that is in preparation and will be submitted soon. Since there is no more than a manuscript in preparation we added some more details on his study as suggested. The text reads now: *During the second half of January 2010 a series of balloon soundings was conducted from FMI (Finish Meteorological Institute) Arctic Research Center in Sodankylä (350 km East from Kiruna). The high-resolution in-situ measurements of the stratospheric gas-phase water vapour were performed by two different hygrometers: FLASH-B optical fluorescent hygrometer (Yushkov et al., 1998) and CFH frost point hygrometer (Vömel et al., 2000), showing close agreement between the data. The water vapour measurements were accompanied with aerosol in-situ observations by the COBALD backscatter sonde flown on the same balloon. The obtained profiles revealed clear evidences of an irreversible dehydration with ice particles sedimenting and evaporating hundreds of*

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meters below the initial level. The signatures of dehydration and rehydration in the water vapour vertical profiles were observed within the 18–24 km range between 17 and 29 January. Further trajectory analysis performed using HYSPLIT on ECMWF reanalysis suggests that the dehydration occurred primarily between 16 and 19 January and the dehydrated air masses were originating from the area North-East of Scandinavia where the coldest temperatures were observed. Trajectory analysis performed using HYSPLIT trajectory model run on ECMWF reanalysis suggests that the dehydrated air masses were originating from the area north of Scandinavia where the coldest temperatures were observed (Khaykin2011). Concerning the Odin/SMR of dehydration: measurements by Odin/SMR reach not to sufficiently low altitudes so that rehydration could be observed. However dehydration/rehydration can be and was observed by Aura/MLS during that time period.

References:

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