Review of: Di Liberto et al., Lagrangian analysis of microphysical and chemical processes in the Antarctic stratosphere: a case study.

We thank the referees for the careful review of the manuscript that has allowed us to improve its content.

The new version of the manuscript now has figure 4 presenting an additional CALIOP match and figure 5 with reported the evolution of HNO_3, as detailed below. Figure 6 has now reported the O_3 range of variability observed by the balloon- borne ozonometer in the 400K ± 10K range, as red (first balloon observations) and blue (second balloon observations) areas. Figure 3 has been re-edited to enlarge the fonts. Figure 1 now present OPC data at higher spatial resolution. Below our responses to their comments, with revision of the text highlighted. We also attach a provisional version of the revised manuscript, for a broader picture of the corrections made.

Response to Interactive comments of Referee #1

(#1;1):The authors should emphasize the advances in PSC box modeling with respect to previous studies. This is the first time that I see all PSC components represented in model simulations with prescribed particle size distributions. I suggest to add an outlook on the potential of the box model with respect to the comparison to groundbased and spaceborne lidar measurements as well as regarding the understanding of PSC evolution in the polar vortex.

We have added the following lines to the end of Section 2.5 (32637,26): "Using a box model such as ZOMM for PSC studies allows a detailed microphysical calculation of cloud processes, which cannot be achieved with global models due to limited computing resources. Case studies without wind shear, as often observed in the polar vortex, offer the possibility to run ZOMM as a column version and, by doing this, to simulate also the vertical redistribution of particles. "

(#1;2):Satellite instruments: Can you please add the horizontal resolution of the MLS and MIPAS observations.

We have added the following lines to (32635,6) : " MLS ClO profiles have a vertical resolution of 3 km in the altitude range of interest for our study, and v2.2 data are reported with a precision of 0.1 ppbv in the range 100-3 hPa. H_2O profiles have a vertical resolution of about 2-3.5 km in the range 316-0.22 hPa, a precision of 6-8 % and accuracy of 4-6%. In the region and altitude of interest, MLS horizontal resolutions in along-track direction are reported to be of the order of 200-300 km for most targets (including H_2O), 400-500 km for ClO, cross-track resolution is approx. 3 km, and the separation between adjacent retrieved profiles along the measurement track is 1.5° great circle angle (approx. 165 km). "

And to (32635,23): " MIPAS along track sampling step is about 410 km. The width of the Field Of View at tangent point in the cross-track direction is about 30 km. The smearing of information around the tangent point given by the width of the single scan horizontal averaging kernels is 200-300 km, for most species (Raspollini et al. , 2012). However, the MIPAS products used here are

obtained by a 2-D (along track) analysis code, and its horizontal resolution is limited by the 410 km sampling step adopted in the retrievals. "

(#1;3):Trajectories: Trajectories are shown between 350 and 460 K. However, the PSC was observed between 330 and 420K. Why the difference?

The altitudes below 14 km (below approx 360 K) are only marginally impacted by the PSC, which has a very low BR there. We concentrated our attention to the more intense part of the PSC, and above that to provide a full picture of the stratospheric transport around the theta level we have then chosen for further analysis (i.e. close to the PSC top, at 400 K, where the airmass returned close to McMurdo)

(#1;4): Microphysical and optical model: page 32637, line 19-20: Can you please add studies that have used the model under those conditions.

We have revised the text indicated, as follows: "So far , ZOMM has always been initiated at typical background conditions for the winter polar stratosphere (Hoyle et al. 2013, Engel et al. 2013). The study by Engel et al. (2014) was the first study, which used the microphysical model as a column model for PSC simulations. For the present study, we developed the model further and introduced the possibility to prescribe a particle size distribution."

(#1;5):The modeled PSC (Figure 5) only reflects the second observed layer from the lidar measurements (14 to 17km). What is the reason for that?

As reported above, we concentrated our study in the altitude region where a second passage over McMurdo occurred ten days after the first observation, i.e. close to 400 K.

(#1;6):Further, the backscatter ratio and the aerosol depolarization ratio are higher in the model compared to the lidar observations. What could be the reason for that?

Modelled Aerosol BR comes from T-matrix calculations of aerosol volume backscatter coefficient, which are done under the assumption of aspherical particles with aspect ratios of 0.9 (diameter-to-length ratio). The refractive index is 1.48 for NAT, as chosen in several earlier studies (e.g., Carslaw et al., 1998a; Luo et al.,2003a; Fueglistaler et al., 2003).

Modelled BR is around 4 at 375K (between 3 and 4 in the observations), drops to 2 at 385K (compared to 1.8 in the observations) and has a second maximum of 3 at 400K (between 2.5 and 3 in the observations,) the comparison between the modeled and observed values show a small (less than 20%) overestimation of the modeled aerosol backscatter ratio. To our knowledge, the order of magnitude of such mismatches is what is to be expected when performing optical computations on observed particle size distribution (see, as instance, Cairo et al., 2011). For what concerns the aerosol depolarization , the modeled depolarization stays at values between 0.4 -0.6 throughout all the PSC history, thus demonstrating the NAT nature of the PSC cloud during its lifetime. In the initial stage of the modeling, its value hovers around 0.6 throughout the PSC extension, which is 30% higher than the observed values of 0.40-0.45. The discrepancy between the modeled and observed depolarization may stem from the assumptions on particle shape and aspect ratio: as instance, T-matrix simulations (Liu et al., 2001) show that aerosol depolarization may vary as much as 50% depending on the aspect ratio and shape of the modeled particle. Increasing asphericity

would result in lower values of aerosol depolarization. However, the arbitrariness in the choice of such parameters in the optical computations does not hamper the main finding of the model exercise, that the cloud remained of NAT type throughout its lifetime. Moreover, the choice made of average values of aspect ratio and shapes, assures that the particle area density is not significantly affected by their uncertainty.

(#1;7):The bimodal lognormal distribution is only fitted to the PSC observation between 14.5 and 16 km. Why not over the entire altitude region of the observed second PSC layer between 14 and 17 km?

In fact, the usual processing of OPC data provides mono and/or bimodal fittings wherever particles are present and counted with sufficient statistics. In the sentence reported in our paper, we meant that in the region between 14.5 and 15 km (15, not 16! This typo has now been corrected in the revised version of the manuscript) this processing produced an average estimate of 22 ppb HNO_3 in condensed phase. Please see also our response to (#3;1).

(#1;8):Chemical simulations: results are only shown for 400 K. However, the double overpass of the air-mass was between 380 and 420 K. Are the simulations robust for the entire altitude region?

The model simulations represent the altitude region 380-420 K in a qualitative way (they correspond to an "intermediate" situation), but not in a strict quantitative way. To be more specific, if the initial ClO_x concentration varied monotonously with altitude, we might conclude that the amount of O_3 depletion, attributable to the ClO_x present at the beginning of the trajectory, at 400 K, is "intermediate" between that at 380 K and 420 K. However we do not have ClO_x with a sufficient vertical resolution to check whether ClO_x really varies monotonously with altitude. In fact, a similar statement on monotonicity is not true for the O_3 mixing ratio: it has a minimum near 400K and is larger at both 380K and 420 K. Moreover and more important, for what concerns the effects of heterogeneous chemistry along the trajectory, it must be stressed that the comparison of runs with and without heterogeneous chemistry depends critically on the PSC present along the corresponding trajectory. As we investigate an altitude at the upper edge of the PSC, a change in altitude significantly changes the PSCs along the trajectory (higher altitude: less or even no PSC; lower altitude: more PSC). Thus the results at a different single altitude may differ from those at 400K.

(#1;9):Figure 5, upper panel: altitude axis is partially covered.

Figure 5 has been re-edited.

Response to Interactive comments of Referee #2

(#2;1):On p32643 l23 the authors note that limb measurements average over large horizontal distances which makes analyzing processes in small portions of air difficult. This also applies to the initialization of the trajectories. It might be helpful to look at daily maps of MLS or CTMs to assess the homogeneity of the vortex in the vicinity of the trajectory initialization. Especially, the uncertainty in ClOx is of interest in modeling ozone loss. Quantifying the uncertainty here will add value to the authors calculation of ozone loss.

We have added the following lines to (32638,11) " inspection of MLS maps for H_2O, HNO_3, ClO, O_3, HCl, N_2O available at 56 hPa level at http://mls.jpl.nasa.gov/plots/mls/mls_plot_locator.php for 10 September 2010, shows relatively low meridional gradients of the species of interest over McMurdo, thus giving confidence on the low spatial variability of the values used to initialize our model calculations. "

For what regards the impact of the ClO uncertainty, please refer to our answer to (#2,9)

(#2;2):In this context, please add the in-situ measured ozone mixing ratios to Figures 6 and 8.

Done.

(#2;3):From Figure 2 it appears that on 20 September ozone at 400K is at about 100 ppbv. Is the calculated ozone loss along the trajectory in agreement with this?

Figure 2 shows at 400K that ozone changes from 400 ppbv to 100 ppbv in 10 days, which provides an ozone loss of approx. 30 ppbv/day. This estimation, which remains fairly constant in a 5K range from the 400K level, is in fair agreement with the modeled ozone loss of 35 ppbv/day reported in the Conclusions paragraph (line (32647,23)).

(#2;4):Please discuss how good the model reproduces the observed ozone loss.

The remark outlined in answer to (#2,3) above has been introduced in the text. We have added to (32647,23) the following lines.

"…35 ppb day^{-1} , in fair agreement with an ozone loss of approx. 30 ppbv/day that can be deduced from the balloon data. We should note that the value of the modeled ozone loss rate is within the ranges reported in the literature (Schofield et al., 2015) and the difference between its value whether heterogeneous chemistry on PSC particles has been considered, or not, is beyond both its reported atmospheric variability, and the sensitivity of our measurements."

(#2;5): The authors have observations of all relevant species and can add value to the discussion concerning the parameters determining the ratio of ClO/Cl2O2. Are the recommended values for the Cl_2O_2 recombination constant and photolysis frequency in agreement with ozone loss observations (for details see von Hobe et al. (2007), doi: 10.5194/acp-7-3055-2007)?

The model simulations, using reaction rate constants from JPL 2011 (including the Cl_2O_2 recombination constant $K_{ClO+ClO}$ and Cl_2O_2 photolysis frequency J_{Cl2O2}), are consistent with the O_3 loss observed (see Fig. 6a). However, the information that we have is not sufficient to draw conclusions about individual reaction rate constants: We know two numbers that are relevant for the O_3 loss by the Cl_2O_2 cycle: the ClO mixing ratio at (near) the beginning of the trajectory and the O_3 loss along the trajectory. In an ideal case (no errors at all) it would be possible to draw conclusions on two "unknown" numbers, e.g. $K_{ClO+ClO}$ and a scaling factor for J_{Cl2O2} . However, the errors associated with the observations (measurement errors, spatial averaging by satellite sensors, distance between the measurement and the corresponding trajectory point) are much too large to allow such an inversion. Significantly more data and the application of statistical methods would be needed.

(#2;6):The authors should discuss the uncertainty of their trajectories. From Figure 3 it appears that after 10 days trajectories which had been started around the same altitude (400K, cyan) spread some hundred kilometers. The authors could start several trajectories around the initial balloon profile and monitor their spread after ten days.

A trajectory cluster analysis has been performed, using NCEP/NCAR reanalysis. Isentropic forward trajectories have been launched, each member of the trajectory ensemble calculated by offsetting the starting lat-lon position by 1° and the vertical position by 400 m upward and downward (corresponding approx. to 5 K). The trajectory cluster remained geographically confined, passing over the McMurdo longitude with a latitudinal spread of approx. plus or minus 5°, roughly centered over McMurdo. The overpassing occurred on average after 9.5 days, with a time spread of approx. 0.5 days. We have further tested the dependency on the particular trajectory model by using three different trajectory models, computed from meteorological fields from different analysis centers: Goddard Space Flight Center (gsfc), NOAA (Hysplit) and European Centre for Medium-Range Weather Forecasts (ECMWF) using the ERA Interim meterological database. The differences between the three different models of trajectories are very small and mainly due to different airmass speeds .

(#2;7):Is the 400K isentrope representative for the 380-420K range where the air mass returned over McMurdo after 10 days? From Figure 5 it appears that 400K is at the upper edge of the PSC and the authors note on p32645 l9 that sedimentation prevents NAT particle existence after two days. At 380K NAT appears to be present until the PSC evaporates, how does this influence heterogeneous chemistry?

As noted by the reviewer, the 400K isentrope is peculiar in the sense that it is at the upper edge of the PSC, hence is the region where sedimentation is more likely to produce an effect on heterogeneous chemistry, as the particles removed are not replaced by particles settling down from above. This is why the models were run at 400K only. The modeled study on the effect of sedimentation has been carried out by running twice the chemistry model at 400K, fed with particle surface areas coming from the PSC microphysical model both with and without sedimentation taken into account. We believe this approach is more effective than studying the evolution at 380K.

(#2;8):Add information about H_2O and ClO MLS products to the discussion.

Please refer to answer to remark (#1;2)

(#2;9): MLS v2.2 has a known bias in ClO (Santee et al. (2008), doi:10.1029/2007JD008762), has this been accounted for in the initialization?

The initialization is based on a ClO amount derived from v2.2 MLS data. At the time of writing the manuscript, we did not take into account the bias present in ClO, studied in detail by Santee et al. (2008). In their work, they assessed a substantial (~0.1–0.4 ppbv) negative bias that may be present in the v2.2 ClO values at retrieval levels below (i.e., pressures larger than) 22 hPa.

The ClO product has been significantly improved in v3.3; in particular, it is reported (Livesey et al., 2013) that such negative bias has been largely mitigated, primarily through retrieval of CH_3Cl (a new MLS product in v3.3). In particular, it is reported that virtually no bias remains at 32 and 46 hPa, and only small biases may still be found at 68 hPa. We checked that the difference between the

initial ClO values derived from v2.2 and v3.3 are not higher than 10% for that particular day, location and pressure levels (68 and 46 hPa we used to interpolate the value at 400K, see answer to (#2,10)). Same results of the v2.2 and v3.3 comparison are found for some other profiles chosen at random around 10 September 2008, finding differences below 10%.

We performed a sensitivity analysis assuming that ClO and Cl_2O_2 were in chemical equilibrium at the time of the measurement (ClO+ClO+M->Cl_2O_2+M, Cl_2O_2+hv->ClO+ClO), and increasing the initial amount of ClOx taking into account a negative bias of 10% of the MLS value with respect to the true value. Such increase of initial ClO would have some visible consequence for the O_3 loss, after 8 days of model running, as depicted in figure A1.

We estimated that such bias would not produce qualitatively significant consequences (a 10% of available ClO would produce an additional 2.5 ppbv/day), and we decided to keep the initial value of ClO as derived from v2.2.

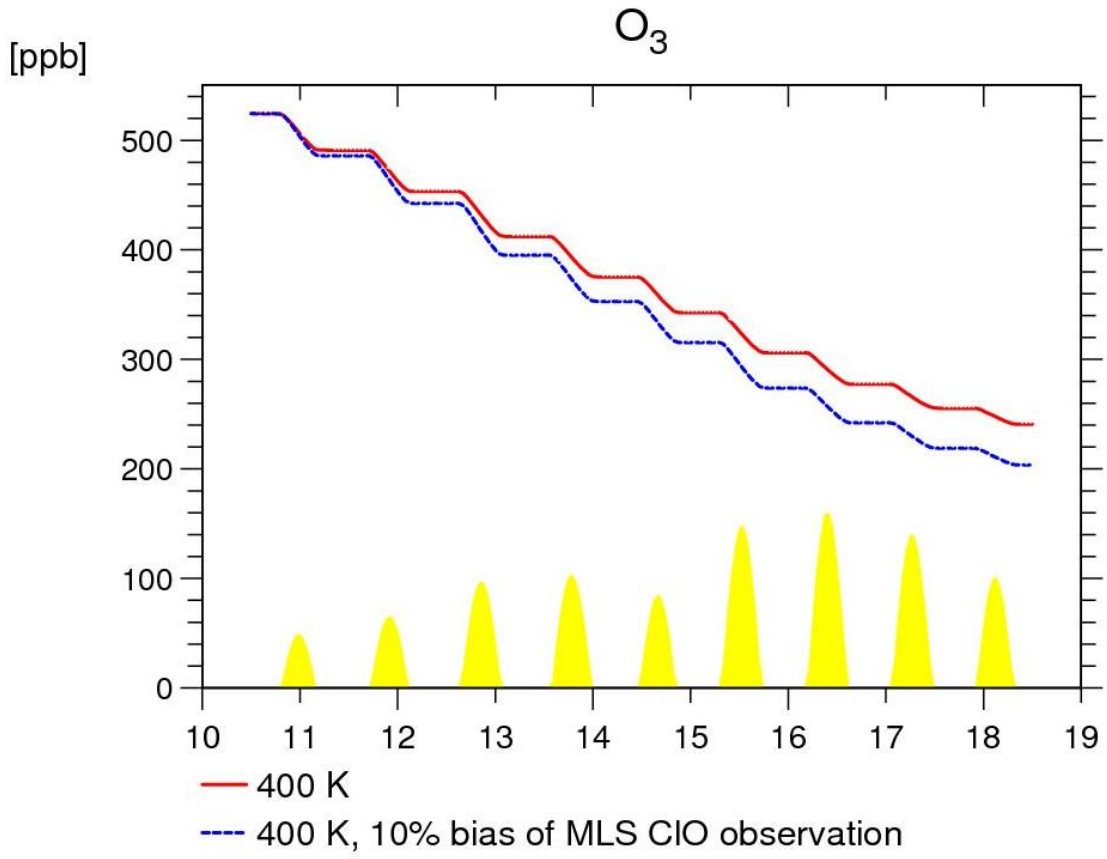

Figure A1: Red line, evolution of O_3 with the amount of ClO prescribed according to MLS values. Blue line, evolution of O_3 with a 10% increase on initial ClO values. The model was run with heterogeneous chemistry and sedimentation taken into account.

(#2;10): Also, has the MLS averaging kernel been taken into consideration?

We do acknowledge the fact that, due to a relatively coarse vertical resolution, satellite data we use to initialize model runs at a given altitude, or compare with the model results, do not correspond exactly to a given altitude, but represent a weighted average of mixing ratios at different altitudes

(which may be expressed by the averaging kernel). However, we think this is the best we can do, because a correction or an elimination of the effect of the coarse resolution of these data is not possible.

If the referee comment has to be intended in the sense that, to compare model results to MLS data, we should process our results with the MLS averaging kernel in order to avoid the presence of fine structures in our results which cannot be resolved by MLS, then the answer would be that we do not see any reason for doing this in our study since we compare to MLS, only results from one single trajectory. To use the averaging kernel, we should have worked with many trajectories at different altitudes.

A final note is due to explain how we have used the MLS, available on pressure levels, in our simulation on a constant theta level. We have interpolated the MLS data x1 and x2, available from the web database at pressure level P1 and P2, to retrieve the data x at the pressure level p corresponding to the 400K theta level, by means of the following formula:

(ln P - ln P1) / (ln P2 - ln P1) = (x-x1)/(x2-x1)

A similar approach is suggested in Ridolfi et al. (2010). Correspondence between pressure P and the 400 K theta level were retrieved from the pressure and temperature data of the 10 September McMurdo balloon sounding.

(#2;11):Add a Figure comparing ZOMM results (backscatter and depolarization) to intersecting CALIPSO overpasses (red circles in Figure 4). How does ZOMM perform in reproducing the PSC several days after the initialization?

We broadened our CALIOP match criteria and considered CALIOP profiles during night time and matching the +/-1.0° latitude, +/- 2.5° longitude, +/- 12 h time difference criteria, with respect to the traced airmass location. By doing this, we were able to add another reasonable comparison on 11 September 2008. ZOMM backscatter results seems to overestimate the CALIOP profiles as seen in Fig. A2. This might be for the relatively large matching criteria, or for the same reason as we do overestimate the condensed HNO_3 as described in *(*#3;1*)*. However, the simulated location and altitude range of the PSC is in good agreement with the observations.

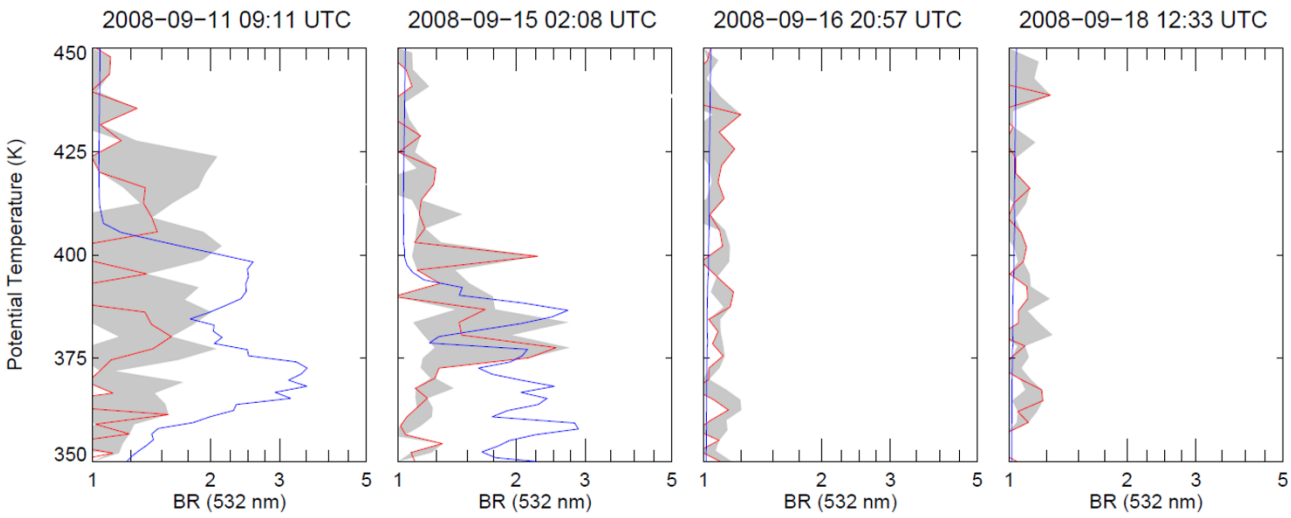

Figure A2. Comparison between ZOMM and CALIOP. Simulated ZOMM backscatter ratios are shown in blue calculated for 532 nm. The gray shaded area comprises maximum and minimum backscatter values from CALIOP within a range of +/- 50 km around the CALIOP profile in red, which is closest to the 400 K trajectory.

Figure 3 in the manuscript was updated to include the 11 September 2008 match, and the text revised accordingly.

(#2;12):How does sedimentation of NAT particles change the abundance of HNO_3 within the PSC? Please add a panel showing total HNO_3 to Figure 5 so the layers of denitrification and re-nitrification become visible. The vertical extent of the PSC alone is rather inaccurate to quantify the extent of particle sedimentation and evaporation.

The following panel (Figure A3) , has been added to fig. 5 in the manuscript, as requested.

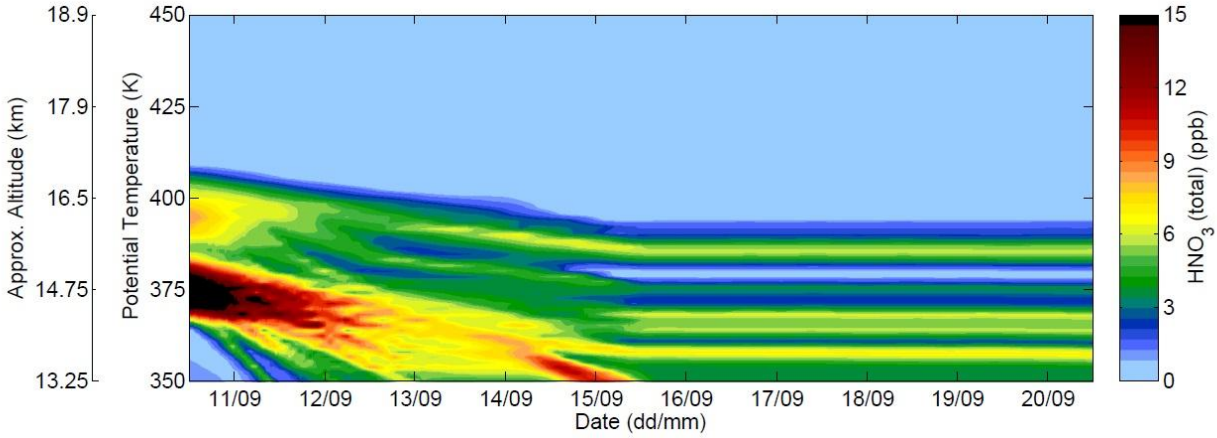

Figure A3: Evolution of total HNO_3.

MLS data do provide evidence of a large vertical redistribution of the HNO_3 in the gas phase after the evaporation of the PSC.

Figure A4 shows MLS HNO_3 profiles taken at the geographical location of the 400K trajectory, on September 10 and on September 15 along the 400K. Even taking into account the coarse vertical resolution of the data, a descent of the HNO_3 peak can be clearly discerned. The order of magnitude of the HNO_3 amount matches with the results of the simulation.

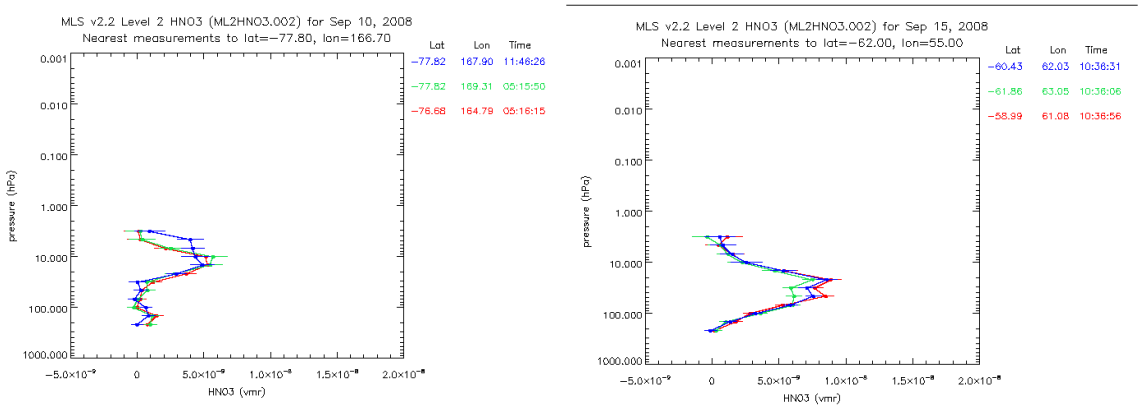

Figure A4: MLS profiles on 10 September (left panel) and 15 September (right panel), at locations where the 400K airmass was.

(32642,3) and following have been modifies as: "Figure 5 shows the modelled evolution of the PSC in terms of Backscatter Ratio (upper panel), Aerosol Depolarization (middle panel) and total HNO_3 (lower panel) during the 10-20 September period.

The persistence of the PSC for almost a week is evident. The cloud remains of NAT type for 6 days after the first McMurdo balloon sounding, and totally evaporates three days before the second McMurdo sounding, as a warming caused its disappearance before 16 September. This warming in the second part of the trajectory coincides with an increasing distance of the airmass from the vortex centre. A vertical redistribution of the cloud is also evident, caused by the modelled particle sedimentation. The cloud vertical extent changes from 410-360 K (approx. 17-14 km in geometrical altitude) to 390-350 K (approx. 16.5-13.5 km) in 6 days. A large vertical redistribution of the gas phase HNO_3 can be seen after the evaporation of the PSC. This occurrence has been confirmed by MLS HNO_3 data on locations along the 400K trajectory."

(#2;13):So far it has only been shown that ZOMM agrees with observations that after 10 days the PSC has evaporated. To conclude that the evolution and type have been reproduced an intermediate comparison with CALIPSO should be shown.

Please refers to what written in response to question (#2,11)

(#2;14):How does the modeled ozone depletion rate compare to the measured O_3 change between both sondes and how does this rate compare to previous studies like MATCH campaigns?

As quoted in response to question (#2;3), figure 2 shows at 400K that ozone changes from 400 ppbv to 100 ppbv in 10 days, which provides an ozone loss of approx. 30 ppbv/day. This estimate remains fairly constant in a plus/minus 5K range from the 400K level, and is in fair agreement with the modeled ozone loss of 35 ppbv/day reported in the Conclusions paragraph (line (32647,23)).

In contrast to the Arctic (von der Gathen et al., 1995; Rex et al., 1998, 2002), Antarctic ozone loss rates by means of Match campaigns have been relatively less studied. A recent article producing loss rates from ozone observed in situ over long periods of time by specially designed ultraviolet photometers flown on long-duration balloons launched in the first half of September 2010 as part of the Concordiasi campaign (Rabier et al., 2013) out of McMurdo Station (Schofield et al., 2015) reports a vortex average ozone loss rate of 74 +-70 ppbv/day over the 410-430 K range, our values are within such variability. The Schofield et al. (2015) reference has now been cited in the text (see our answer to comment (#2;4)).

(#2;15):Figure 6: Describe how the error bars on the satellite observations were determined

Errors bars associated to MIPAS satellite data are the retrieval errors due to measurement noise on the used single values. For MLS data they are derived from the precision reported in the data files (see for description Livesey N. J., 2011) propagated to the 400 K interpolation level (see our answer to (#2,10),

Response to Interactive comments of Referee #3

(#3;1):My major concerns is the estimate of condensed nitric acid… …the result would be more convincing if appropriate values of condensed NAT were used.

The sentence in (32641,25), which has raised the perplexity of the referee, is incorrectly expressed. In fact the number reported there refers to values encountered between 14.5 and 15 km. In the microphysical simulation at 400K, which we analyzed in further detail afterward, the initial value of total HNO_3 is around 7 ppb, and the gas phase HNO_3 is 1.7 ppb. In fig. A5, the evolution of total (left) and gas phase (right) HNO_3 at 400K is shown, for the case when denitrification is acting (red line) or not (blue line).

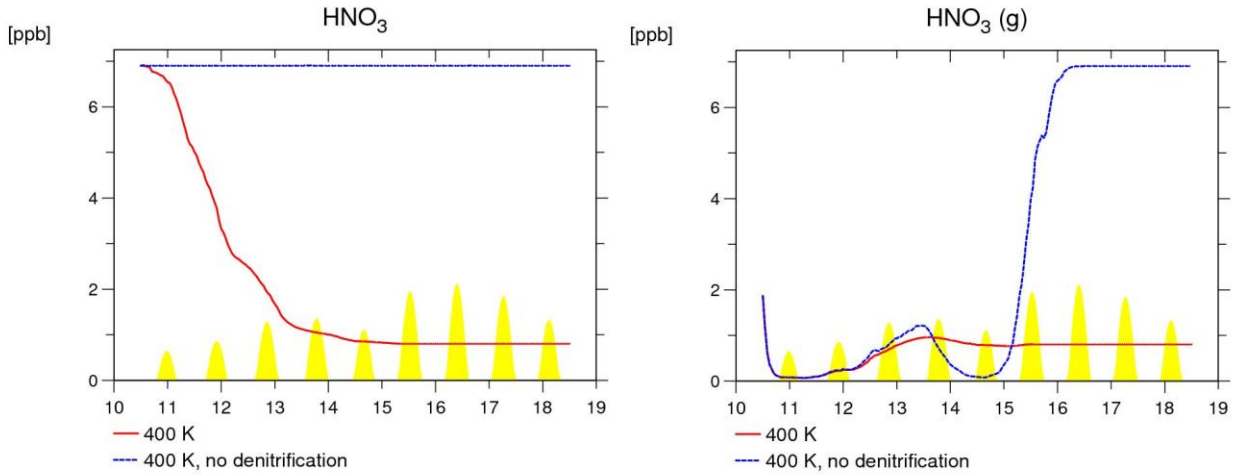

Fig. A5: Total (left) and gas phase (right) HNO_3 evolution at 400 K, from the microphysical model with (red line) or without (blue line) sedimentation

Much higher values of total HNO_3 were reached close to 370 K, where such enrichment can probably partly be due to sedimentation, occurred in the previous days. However, as suggested in the revised text, the value of 22 ppbv HNO_3 in the NAT particles, met in some region of the cloud, computed assuming spherical particles in the largest lognormal mode, may be an overestimation, based on the HNO_3 expected to be available, but not based on the particle measurements. The particle measurements were looked at closely and there seems to be no reason to dismiss the estimates of particle volume from them. The large particle concentrations are significant (0.1 cm^{-3}) and the particles are large 2-4 µm. Thus the measurement uncertainties are not large. Both lognormal fits and simple histogram summations lead to similar estimates of particle volume, which, given the assumptions above lead to the large estimates of condensed HNO3. The paper of Molleker et al. (2013) confronts a similar problem, reporting Arctic HNO_3 mixing ratio in condensed phase up to 17 ppbv ±40 % (observations on January 2010) and up to 25–35 ppbv ±40 % (11 December 2011), derived from a particle diameter estimation based on the hypothesis of spherical particles combined with an assumption of a pure NAT composition. Although large HNO_3 containing particles were consistently observed by a condensed phase NO_y instrument operated at the same time of the optical particle counter, nevertheless in general the particle volumes seemed to overestimate the HNO_3 content in the condensed phase. Two possible explanations were put forward, namely (i) non-compact or non-spherical NAT particles, and/or (ii) NAT coating on

preexisting ice particles. In our case, hypothesis (ii) cannot be ruled out due to the previous thermal history of the airmass at 400K which would have allowed ice formation in the week before our observation. We must acknowledge the fact that NAT particles may have larger apparent sizes compared to compact spherical particles, e.g. due to non-compact morphology or aspheric shape or ice presence . In any case, either NAT coated, or high aspect ratio, or spongy particles, would lead to a reduction of our estimation of condensed HNO_3 that would be basically unpredictable, leaving us with only the resource of educated guesses, as the one (4-5 ppbv in condensed phase) correctly suggested by the referee. It is worth noting that all these hypothesis put forward to reconcile particle dimension with HNO_3 content, would impact particle settling speeds as well, but unfortunately in opposite directions, again leaving us in uncertainty. That is why we decided to stay with the assumption of compact, spherical NAT particles but acknowledge here the fact that our assumption leads to HNO_3 in condensed phase which may be an upper limit (we delete the sentence "considered realistic" in (32641,26) and to discuss what may be the consequences of reducing the condensed HNO_3, in our results.

Lines (32641,18) –(32641,26) are rewritten as:

"With these assumptions estimates of HNO_3 content in the particles larger than ~ 0.3 μm ranged from 20-30 ppbv from 370-385 K (14.5-15.5 km), to near 10 ppbv from 385 – 405 K (15.5-16.8 km). While these values are higher than expected, they result from the OPC measurements of significant concentrations (~0.1 cm-3) of particles in the 2-3 μm range, so are not easily dismissed. A possible overestimation of the HNO_3 content in the condensed phase from particle size measurements was reported in some recent airborne measurements from the RECONCILE campaign (von Hobe et al., 2013) and possible reasons for that are extensively discussed in Molleker et al. (2014). The microphysical run was initialized with an estimate of the initial value of total HNO_3 of 6.9 ppb, of which 5.2 ppb in condensed phase, considered as a realistic upper limit for the amount of HNO_3 in the particles. "

(#3;2):On what scales do you expect these processes to occur and how will this smearing effect impact your analyses?

The problem of smearing out temperatures and winds from meteorological analysis, and trace gas concentrations from satellite fields, is present and unavoidable every time box models are used in process studies and compared with those datasets. In our work, subgrid temperature fluctuations are superimposed to synoptic scale temperature histories for the microphysical simulations. For what concerns small scale fluctuations in trace gas fields, they exists as documented in many stratospheric aircraft in situ measurements. As instance the recent Rolf et al., (2015) paper shows 50% ozone fluctuations over linear distances of few hundreds of km, in Antarctica at 16 km altitude (flight on 13 September 2012). Inspecting the data set in our possession indicates that satellite trace gases maps do not show strong and large gradients over McMurdo during the time of our in situ observations, and that the Eulerian observation of the large PSC event over McMurdo during 10 September 2008 show a significant stability of the cloud in terms of optical properties and spatial distribution during the course of the balloon measurements. This information has been introduced in the revised text.

(#3;3):Why haven't you compared the CALIPSO data with the model calculations in a more quantitative fashion?

Please refer to our answer to (#2;11)

(#3;4):P.1, L.27-29: M L.27-29: Minor point- the use of 'conversely' here seems inappropriate.

This has now been deleted.

(#3;5):P.2, L.58: You state here that Antarctic PSCs are predominantly type NAT? (…) Are your 'type NAT' PSCs actually liquid/NAT mixture PSCs.

We reformulated the sentence in (32631,16-17) as follow: "STS-NAT mixtures are the predominant composition classes of PSC observed throughout the stratosphere from mid-June until mid-September in Antarctica, with NAT particles prevailing at altitudes above 15 km. (Pitts et al., 2009). Over McMurdo, PSC of prevalently NAT type appear in early June…"

(#3;6):P.3, Section 2.3: You don't mention the horizontal resolution (cross-track and alongtrack) of the MLS or MIPAS instruments, which can be hundreds of kilometers. I think this information is relevant to the study and should be mentioned here.

Please refer to answer to remark (#1;2)

(#3;7):P.3, Section 2.3: The coincidence criteria for MLS and MIPAS is about +/- 200 km and for CALIPSO is about +/- 100 km. Given the spatial variability of the gas species, PSCs, etc., how do changing these criteria impact the comparisons?

We have tried larger criteria for the gas phase, but doubling the acceptance radius increases the number of coincidences with the only benefit to add more scattered point. For what concerns the gas phase, 200 km seemed a good compromise between data statistics, average length of gas phase gradients and the distance from McMurdo of the airmass sampled the second time. PSC spatial variability can occur on much smaller spatial scales, so we had chosen to narrow CALIOP coincidence criteria. However, in the revised version of the manuscript, the coincidence criteria for CALIOP have been widened to match the MLS and MIPAS, the text of the revised manuscript has changed accordingly (please refer to our answer to (#2,11).

(#3;8):P.3, L.232: CALIPSO and Aura are in the same orbit, but Aura was repositioned in early 2008 and now CALIPSO and MLS make measurements within about 30 s of one another. MLS profiles are separated by about 165 km along the orbit, while CALIPSO samples on a much finer horizontal resolution of ~1 km in the lower stratosphere. See Lambert et al., Atmos. Chem. Phys., 12, 2899–2931, 2012 for more details.

This information, and the suggested reference, has been inserted in the text:

(32636,9-11) "The CALIPSO and Aura satellites are part of the A-train track (Stephens et al., 2002). Since early 2008 CALIOP and MLS make measurements within about 30 s of one another. MLS profiles are separated by about 165 km along the orbit, while CALIPSO samples on a much finer horizontal resolution of approx. 1 km in the lower stratosphere (Lambert et al., 2012)."

(#3;9):P.4, Section 2.5: I'm somewhat familiar with the standard ZOMM, but not the column version. Can you describe in a little more detail how sedimentation is handled? Does it assume that the entire column of air is moving at the same velocity and in the same direction (i.e. no horizontal or vertical shear)? Are these reasonable assumptions in this case? I could imagine near the vortex edge this would not be the case. How do these assumptions potentially impact the results?

The approach of using a column model is indeed only possible in situations without horizontal wind strain or vertical wind shear. Changes in wind direction or velocity with changing altitude would otherwise lead to errors in the location of the sedimentation events. However, at 400 K, the trajectories stayed sufficiently close to meeting the criteria of a homogeneous wind field. In the figure A4, hereby reported, a quality criteria was introduced to check whether the trajectories were diverging. The presented horizontal displacement is calculated over a vertical distance of 500 m. As can be discerned from the figure, the assumption of a homogeneous wind field was reasonably good throughout the lifetime of the PSC, at the altitudes of interest. After 15 September and below 350 K, the assumption of a homogeneous wind field is not fulfilled.

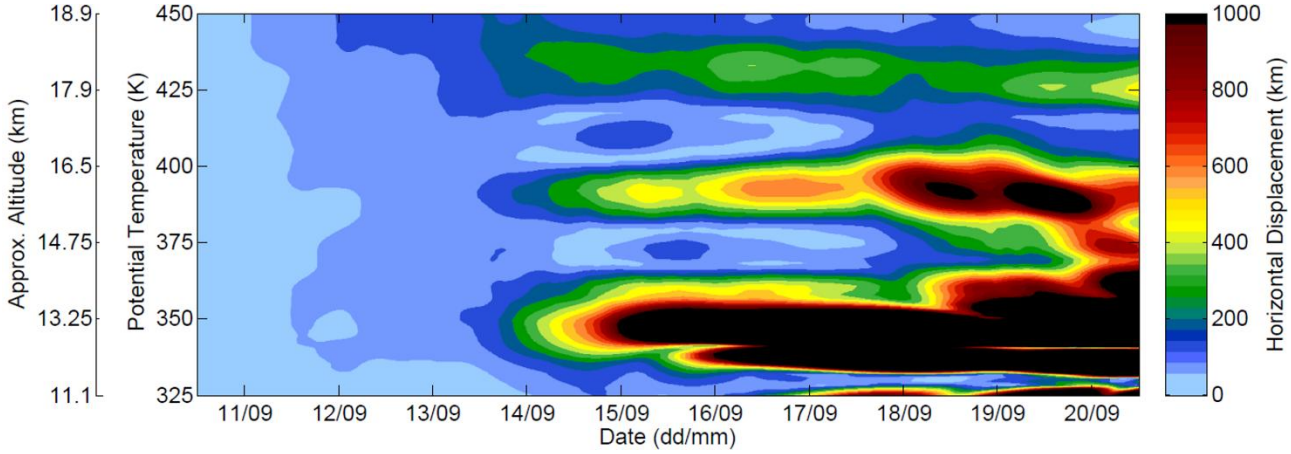

Figure A6: Horizontal displacement between air mass trajectories originating from McMurdo and run 10 days forward. The horizontal displacement is calculated considering an altitude range of 500 m.

(#3;10):P.4, L. 303-305: Are these MLS measurements assumed to be representative of the total (gas phase + condensed phase) abundance? How is uptake on PSCs estimated?

MLS measurements presented here are concerned with gas phase only.

(#3;11):Can the coarse resolution of MLS accurately capture the scales of the processes of interest?

Probably not if PSC occurs in highly localized regions whose spatial extensions is smaller than the horizontal resolution of the limb sounder, as PSC processing of air may create gradients of species within the spatial resolution of the sensors. However, as instance, Santee et al. (2002) studied the redistribution of HNO_3 in the Arctic due to PSC particle sedimentation by following the PSC evolution from formation through dissipation with a Lagrangian approach, and concluded that every time the models displayed large changes in either HNO_3 or aerosol extinction, they were reflected in the MLS data.

In our study, of course, there can be a degree of inaccuracy of the trace species observed by the satellite, due to their inherent spatial average, and we indicated that in the text as a possible explanation for the coarse comparison between model results and data, The extent of this inaccuracy cannot be known a priori.

(#3;12):P.4, L. 325-326: How far did the balloon fly downwind from McMurdo during the four hour flight? How much variability was there in the lidar measurements over the four hour period? This would provide some insight into how homogeneous the PSC field .

The PSC observation occurred in approx. 70 minutes out of the 4 hour balloon flight duration, and the balloon flew approximately 50 km. During that time window, the optical parameters of the PSC observed by the lidar over McMurdo remained quite stable and consistent. This information has now been added in the text:

(32638,24-32639,2) "A PSC was observed for 70 minutes in the initial part of the flight, during ascent, over a horizontal distance of approx. 50 km. The left panel of Fig. 1 depicts the lidar backscatter (solid line) and aerosol depolarization (dotted line) ratios vs. altitude, averaged over the OPC observation time, while the right panel shows the particle size distribution measurements obtained by the OPC."

(#3;13):P.4, L.335-340: Is it likely that there were also STS droplets present in this 'NAT' cloud? What number concentration of STS is present?

As reported in (32642,15-16), we have assumed that the smallest size bins of the measured size distribution (particles < 0.3 μm radius) consisted of liquid STS droplets. Their number concentrations can be discerned by inspection of Figure 1.

(#3;14):P.4., L.358: Same comment as earlier- do you think 300 km is sufficiently close for an overpass to assume the balloon and lidar are sampling the matched air mass?

The study has been done under that assumption. 300 km is a value that lies at the lower range of variability of match radii used in previous studies.

(#3;15):P.5, L.367-370: Can you speculate on what caused the very different ozone values in the uppermost part of the soundings? Is this a chemical or dynamical effect? Does this imply that the air in the column may be exposed to horizontal or vertical shear and therefore the ZOMM column model would not be representative?

The air sampled on 20 September 2008, in the uppermost part of the sounding, had a different origin with respect to the 390-410K layer. On 10 September, this latter layer was above McMurdo, while the former was displaced more than a thousand km northward. Moreover, from 10 Sept onward, the air above 410K never experienced temperatures low enough to allow PSC formation. As the two soundings sampled different airmasses, on the 10 and 20 September, we could not speculate about ozone loss rates based on such observations as they are certainly to be ascribed to both chemical and dynamical effects.

We do not share the view that, since the air in the column over McMurdo on 10 September was exposed to different horizontal winds, therefore the ZOMM sedimentation could not be used. We

concentrated our study on the top part of the PSC observed on 10 September. There, sedimentation acted to remove particles from that level, but not to deliver additional particles settling from above, as there was no PSC above. Therefore at that particular level the possible wind shear do not impact the microphysical model results and how they have been used in the chemistry simulation.

(#3;16):P.5, L.376-378: Are these ozone losses observed in the past also representative of the ten-day time scale of this study?

The time scale of our study is of the same order as those considered in previous Match campaigns. Please refer also to response to comment (#2;14) for a comparison between ozone loss rates.

(#3;17):P.5, L.400: Why are there not more MLS values coincident with CALIPSO in Fig. 3? Am I misinterpreting the figure?

We used all MLS data available on its database, which we acknowledge are in fact less than MLS passages over the trajectory. The reason for this lack of data in the database is unknown to us.

(#3;18):P.5, L.404-422: It seems like you could find good coincidences with CALIPSO on almost every day-or at least find a representative CALIPSO orbit. Are there CALIPSO orbits available on 10-15 September, but not shown? The CALIPSO data as currently used does not add much to the paper- other than show that there was a PSC in the area.

Please refer to what expressed in response to (#2,11).

(#3;19):P.5, Section 3.2: See general comment above about estimates of condensed HNO_3. I don't believe 20 ppb is reasonable.

Please refer to what commented on (#3;1).

(#3;20):P.5, L.460-464: How reasonable is the descent of the cloud from 17-14 km to 16.5- 13.5 in 6 days? What size particles and fall speeds would this correspond to? Is this consistent with the size distribution observed with the OPC? Does CALIPSO observe a similar vertical descent of the PSC? Is there any signature of renitrification in MLS data at the lower altitudes after the PSC evaporated?

Fall speeds (Luo et al., 2003b) depend upon particle densities and dimensions (but also on shape, not taken into account) which are calculated by the microphysical model initiated by size distributions from OPC measurements. Hence there is an internal consistency between sizes and fall speeds. To present here a back of the envelope calculation, 100 m/day (approx. 10^{-3} m/s))would correspond to spherical particles of approx. 1 μm diameter, so there is a good order of magnitude match with the observed and modeled particles. It is worth noting here that aspherical, or spongy, or NAT coated ice particles would have a different but basically unpredictable, fall speed (although recent studies (Woiwode et al., 2014), suggest a 30% reduction of NAT rocks fall speeds, to match with sedimentation observations carried out during the recent RECONCILE campaign). As noted in the first version of the manuscript, we did not have useful CALIOP matches when the PSC was present, as the closest match we presented is on the 15 September, when the PSC was vanishing. More relaxed matching criteria allowed us to use a match on 11 September 2008, discussed in the

answer to (#2,11). For what concerns signs of renitrification upon PSC evaporation, please refer to what exposed in answer to (#2,12).

(#3;21):P.6, L.512-519: This is true for all of the comparisons and should be discussed much earlier (e.g. Section 2.3). Can you elaborate more on how this may affect your analyses- does it limit your conclusions?

Sasano et al. (2000) and Terao et al. (2002) have used the Match approach in the Arctic based on ILAS satellite measurements, to compute ozone loss rates, and a similar Match study in Antarctica, based on POAM satellite data, has been reported by Hoppel et al. (2005). Match radii higher than the one used in our study (respectively 400 km for ILAS, and of 1000 km for POAM respectively) were used, and the problems due the relatively low vertical resolution and a large sampling air mass volume of the satellite sensors, leading imperfections in the matches (i.e., the re-sampled air mass is not identical, and the satellite data report a spatial average of the targeted specie) were reduced by computing ozone loss rates, based on a statistical analysis of many matches rather than a single pair. Obviously this approach cannot be pursued in our case, and the problem of satellite data averaged over their spatial resolution can only be reported. We have added at the end of paragraph 2.3, as suggested by the referee, the following lines:

"Although satellite data lacks the resolution and precision of in situ measurements, they have been used in the past for Lagrangian studies of the polar stratosphere (Santee et al., 2002, Sasano et al., 2000, Terao et al. ,2002, Hoppel et al.,2005, Riviere, 2003), relying on averaging observations and trajectory matches over long periods. As we are interested in a single case study, this standard approach cannot be used, and our comparison between modeled result and satellite observation will suffer from the inaccuracy due to the spatial averaging inherent to the satellite observations. The extent of this inaccuracy cannot be known a priori, but should be kept in mind when discussing our results."

(#3;22):P.8, L.719-732: How dependent are your conclusions on the time scales and spatial scales of the case you've analyzed? Do you think the impact of denitrification on ozone depletion remains small over the course of the spring?

We have been trying to make clear that our conclusions are limited to this single case study, and the suggested message it drives should be that it may not be true that denitrification is ALWAYS important. Our study would suggest that there may be conditions under which this is not the case. Our study does not imply that the impact of denitrification is small or negligible over the course of the spring. This is now more explicitly stated in the revised text.

The new text: (32647,28-32648,4) "Although no general conclusion can be drawn from a single case study, our findings supports the view that there may be cases when additional surface area provided by solid PSC particles does not increase appreciably the chlorine activation, since in cold conditions the activation could originate from heterogeneous chemistry on surfaces basically provided by a background aerosol distribution."

(#3;23):Figure 3: Much too small! I had to enlarge this significantly to even see the MLS and CALIPSO measurement locations and trajectories. This needs to be much larger.

Done.

(#3;24):Technical P.6, L.546: "Groo" should be "Grooß." P.6, L.568: "Groo" should be "Grooß."

This has been corrected.

(#3;25):P.7, L.596-598: Citation is missing.

The citation at the end of paragraph 3.3.1 is correctly present in the on-line version of our ACPD: Arnone et al., (2012).

(#3;26):P.8, L.706: 'as will be discussed later' - is this really discussed 'later'?

We have changed the sentence in (32648,16) to:

"Moreover, although the effects of denitrification on HCl and $ClONO_2$ are opposite, so they cancel to some extent, the increase of $ClONO_2$ (not-denitrified vs. denitrified scenario: approx. 0.6 ppb by the end of the time interval) is larger than the decrease of HCl (approx. 0.2 ppb), so that in total more ClO_x is converted to reservoir species in the not-denitrified case."

We have added the following references to the manuscript:

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