Interactive comment on “Total depletion of ozone reached in the 2010–2011 Arctic winter as observed by MIPAS/ENVISAT using a 2-D tomographic approach” by E. Arnone et al.

Anonymous Referee #3

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Overall comment:

This manuscript presents results for the Arctic winter of 2010/2011 based on 2D tomographic retrievals of ENVISAT/MIPAS data and ECMWF analyses. The MIPAS2D retrievals seem to be of high quality, and what they show about the extraordinary stratospheric conditions of the 2010/2011 winter is of general interest. However, in my opinion the manuscript contains numerous inaccuracies and flaws in either the analysis or the exposition of it (or both), and it will require substantial revision before it is suitable for publication in ACP. Both major substantive issues and minor wording comments are enumerated below.

Substantive comments:

– p33193: L12-14: "... chlorine is converted into active radicals such as Cl and ClO, which destroy most of the vortex ozone at 14-20km altitude (Solomon et al., 1986; Molina et al., 1987)". Solomon et al. and Molina et al. are not the best references for the second part of the sentence, for which a recent WMO report would be more appropriate.

– p33193, L17-18: "Denitrification of the Antarctic stratosphere through sedimentation of HNO3 prevents the reactive NOx to promptly recapture the available chlorine into reservoirs". This sentence needs to be more carefully worded. Denitrification only prevents chlorine deactivation through the formation of ClONO2; it has no impact on the production of HCl, which is the primary deactivation pathway in the Antarctic.

– p33193, L24-25: "The total ozone depletion in the Arctic winter was found to be linearly dependent on the volume of PSCs integrated over the winter (Rex et al., 2006)...". This relationship has also been confirmed for the Antarctic by Tilmes et al. [GRL, 2006].

– p33194, L12: "Eventually, Arctic ozone hole conditions were reported for the 2010-2011 Arctic winter (Manney et al., 2011)". Using the word "Eventually" here makes it sound as though it was inevitable that ozone hole conditions were reached in the Arctic. I don’t think that’s true. It might be better to say something along the lines of "Recently, Arctic ozone hole conditions were reported for the first time for the 2010-2011 Arctic winter (Manney et al., 2011)".

– p33196, section 2.2: The MIPAS2D retrievals at the core of this work are briefly described in this section, but no information is provided about their quality, that is, accuracy, precision, and resolution (the retrieval vertical grid is specified, but that does not necessarily reflect the true vertical resolution of the data). It is stated that the MIPAS2D database is "thoroughly described in Dinelli et al. (2010)", but readers should not have to look up another paper to obtain information that is critical for evaluating the reliability of the results presented here.
... regions of temperature below TNAT lasting almost continuously until late March”. In fact, in at least some meteorological analyses, minimum temperatures were below TNAT continuously from early December until the beginning of April. It is only the MIPAS data that are discontinuous in this interval, not the low temperatures.

Temperature minima dropped persistently below 185K ... The presence of these persistent cold regions is reproduced in the vortex average temperature ...”. First, I do not think that vortex average temperature is a particularly meaningful quantity. I think it would be much more informative to show minimum temperature in Fig. 1, rather than the vortex mean value. Second, it is rare for minimum temperatures to drop below 185 K in the Arctic. Although 2011 was a cold winter, temperatures were not extraordinarily low (they were just moderately low for an extraordinarily long time, as shown in Manney et al. [2011]). Thus I am not convinced that such low temperatures were reached at all in 2011, and certainly not persistently for days at a time, as is asserted here. Had temperatures really been that low for that long, many more ice PSCs would have formed and a greater degree of dehydration would have been observed. This leads me to suspect the quality of the MIPAS temperature data (as noted above, no information on data quality has been provided in the manuscript for most of the MIPAS data products). How reliable are the MIPAS temperatures? How well do they compare with, for example, ECMWF? My guess is that the ECMWF temperatures are more suitable for polar processing studies than the MIPAS retrievals. Since ECMWF temperatures are used elsewhere in this paper, why not use them here too instead of the MIPAS data?

Large concurrent increases in temperature and the N2O tracer showed the Arctic return to typical conditions.” It’s not clear when this occurred. The previous sentence referred to late April, so perhaps this one does as well, but that should be clarified. The reader cannot judge either of these sentences because the plots in Figs. 1 to 4 only extend to late March or mid-April.

A total of 2920 MIPAS scans detected PSCs over the period 1 December 2010 to 18 March 2011, corresponding to 82% of days with PSCs out of 92 days of observation”. Earlier (Section 2.3) it was stated that MIPAS identifies about 70% of the PSCs detected by CALIPSO in the Arctic. So clearly some PSCs are missed. Are there false positive detections as well? These statistics have implications for results discussed later in the manuscript.

... close to the cold core of the vortex for most of the winter, tracing the occasional distortion of its shape”. I am confused by this wording. What is meant by “tracing”? Perhaps “following” would be a better word. Is it the whole vortex or just the cold core that gets distorted? “The only evident deviation was observed between 20 and 29 January, when detected PSCs shifted from the cold core of the vortex to form a ring surrounding it”. I assume that this ring of PSCs surrounds the cold core of the vortex, and not the vortex itself (i.e., it does not lie outside of the vortex). Perhaps the implications of the PSC formation region shifting from the coldest part of the vortex core to warmer areas could be discussed more. Did this change in the location of PSC formation occur for both the upper and the lower altitude PSC regions?

Although the majority of PSCs were vertically distributed where the average temperature was consistently around the threshold for STS formation (Fig. 1b), temperatures associated to the PSCs of this period were the highest of the 4 periods”. Again, because PSCs are highly localized, the vortex average temperature is almost totally irrelevant. In fact, although one can tell from Fig. 5 that period (i) was warmest overall, Fig. 1a shows that in a vortex-average sense, temperatures in the first half of January were actually higher than they were in the last half of December. This underscores the point that vortex mean temperatures are much less useful in this context than minimum temperatures.

For period (ii) it is stated in L2 that "a very few cases" were classified as ice PSCs. I see only one point clearly in the “ice” category in Fig. 5b. In addition, in L16 the statement is made: "The few cases showing ice composition (top
left of the panel, including also those around CI=1.5), ... represent the only ice PSCs detected during the whole season”. I am confused by this statement, because the points near CI=1.5 lie to the right of the “ice” line and thus are classified as “STS/Mix” in Fig. 5b.

– p33204, L2-5: “The bulk of STS/Mix PSCs in the diagram was consistently associated with temperature below STS formation, and was observed between theta=450 and 550 K (Fig. 1b). At theta from 550 to 700 K, the STS/Mix PSCs had more scattered color ratio values and associated temperatures that reached values above TNAT”. It’s not clear to me how the reader is supposed to know at which altitudes the points plotted in Fig. 5 occur. This sentence points to Fig. 1b, but I do not see any easy way to associate the two figures. So while the reader can look at Fig. 5b and see that most of the STS/Mix PSCs formed at temperatures below TSTS, there is no way to judge that these occurred between 450 and 550 K, nor any way to tell that the points displaying greater scatter in Fig. 5b were located at 550 to 700 K.

– p33204, L9-11: "The very high altitude clouds (CTH at theta > 700K) detected during the very first days of January had either a clear STS signature (see bottom right of Fig. 5b) or extended towards the ice region (top left of the panel)”. I am skeptical that PSCs, especially low-CI (implying thick ice clouds) ones, formed between 700 and 950 K (i.e., up to ~35 km altitude). In order for the authors to credibly assert PSC formation at such extraordinarily high altitudes, they need to provide far more support for their evidence than they do in this manuscript. There is no historical precedence for such high-altitude PSCs (see comment below about p33210, L4-7). What do CALIPSO measurements (or any other PSC observations, for that matter) show for January 2011? Do they provide any indication of PSC activity at these altitudes during this period? As I mentioned above, some statistics on the number of “false positive” PSC detections in MIPAS spectra are needed. It would indeed be a noteworthy discovery if MIPAS has detected Arctic PSCs up to 35 km, but the current presentation of the data has left me unconvinced that these signatures are real.

– p33204, L6-7 and L11-13: “suggesting PSC formation within cold mesoscale temperature perturbations that were not reproduced by ECMWF temperatures” and “In the clear STS cases, associated temperatures were largely above TNAT, again supporting the importance of mesoscale perturbations that were likely missed by ECMWF temperatures”. I think more explanation and/or description of the ECMWF temperatures is needed, probably in section 2.4, which is labelled "ECMWF meteorological products" but which discusses only sPV (temperature is not mentioned in that section even though it is used in this study). I would have thought that the full-resolution ECMWF fields would capture mesoscale temperature fluctuations, so exactly what ECMWF data are being used should be specified. Particularly in the second case (L11-13), rather than supporting the occurrence of unrepresented mesoscale temperature perturbations, I think a more likely explanation is artifacts in the data that are being erroneously classified as PSCs.

– p33205, L3-5: "Fig. 5e–h report some events with a remarkable consistency of the classified PSC and the expected NAT and STS regions also under very inhomogeneous conditions”. Although good, I am not sure that the consistency of the PSC classes and STS/NAT formation regions can really be characterized as "remarkable", especially as some of the white diamonds do lie outside of the white contour lines. Also, I do not know exactly what is meant by "very inhomogeneous conditions" in this context. In the Arctic, low temperature regions are typically small, shifted off the pole, and not concentric with the vortex, so the conditions depicted in Fig. 5(e-h) do not look unusually inhomogeneous to me.

– p33205, L8 and L16-17: "each of the PSCs we classified as NAT was composed of at least 40% of such particles ... The overall fraction of 16% of NAT classified PSCs in the 2010–2011 Arctic winter can be therefore considered a good estimate of PSCs with dominant NAT composition". On p33199, the percentage of small NAT particles in PSCs classified as "NAT" was given as "30-40%”. In either case, it seems like a fairly low percentage for such PSCs to be characterized as having a “dominant NAT
composition". Also, it wasn’t clear where the overall estimate of 16% NAT PSCs during the winter came from, since the NAT fraction for the four individual periods was given as 10%, 20%, 18%, and 6%, respectively.

– p33206, L3-4: "The low HNO3 occurred where temperatures were below the STS formation threshold suggesting a capture by the NAT component of the STS/Mix PSCs". I didn’t really follow this argument – why would the temperatures being below TSTS necessarily imply that NAT particles in mixed clouds were taking up HNO3? Why wouldn’t there be HNO3 uptake by the STS droplets themselves?

– p33206, L12-16: "As expected by the lack of ice PSCs, H2O (Fig. 2a) showed only a marginal decrease in late March in the lower stratosphere. ... the overall trend in H2O was consistent with the diabatic descent. The lowest H2O was reached in the second half of March at theta = 400-450 K, in coincidence with minima in HNO3 and O3, and increase in ClONO2". The analysis presented here suggests that very few water ice PSCs formed in this winter. Consequently, I doubt that the degree of dehydration was large enough to be discernible in vortex-average H2O abundances. In addition, even the few ice PSCs that were detected occurred earlier in the winter; no ice PSCs were still present in late March when the minimum in H2O was measured. It’s not clear how the fact that the mimima in HNO3 and O3 and the increase in ClONO2 occurred at the same as the minimum in H2O is relevant, since they all have different causes. Finally, even slow descent should have led to small increases, not decreases, in H2O in the lower stratosphere. So I think that the decrease in MIPAS H2O values in Fig. 2a needs to be explained better.

– p33207, L13-15: "Snapshots ... reported in Fig. 3 show depletion of O3 occurring within the vortex since January". Since the first snapshot in Fig. 3 is from early February, it really cannot be judged from this figure exactly when O3 depletion started. Depletion that may have taken place between December and February would not be discernible in this figure. Moreover, chemical loss cannot be diagnosed merely through examination of "dot plots" of O3 such as those in Fig. 3 in any case. Replenishment of O3 via diabatic descent could have offset any loss and made the O3 in these maps appear to be constant or even slightly increasing even though some loss took place.

– p33209, section 4.1: This section compares the MIPAS measurements from the 2010-2011 winter to those from previous Arctic winters. Although I understand that the MIPAS data used in this manuscript differ from the IMK retrievals discussed in previous MIPAS papers on earlier Arctic winters, I think it would have been courteous for the authors to acknowledge that some of those papers exist (e.g., some studies by von Clarmann, Oelhaf, and others). In fact, stating how consistent the MIPAS2D data shown here are with previously-published results for some of these past winters might have been useful.

Another overall comment on this section is that the second paragraph comes across as a collection of random thoughts with no thread tying the sentences together. It would benefit from some reordering and reorganization. For example, the paragraph starts off discussing temperatures and then moves on to descent. This is followed by a return to temperature in the sentence: "The cold core of the vortex traced by the TNAT region persisted until early April at a slightly higher altitude" (higher than what?). The TNAT sentence is followed by "Since in many past Arctic winters the vortex disappeared before spring, it should be noted that the distribution in time and the multi-year average of 2003-2010 data at a certain date is given by only those years for which the Arctic vortex was defined". This is a perfectly valid point to make, but one wonders why it is stuck in the middle of this paragraph. Immediately following this note, PSCs are discussed.

– p33209, L11-12: "The figure also reports averages over the reference TNAT region (green) introduced in Sect. 3." I do not see the relevance of calculating averages over the TNAT region for most of these quantities. Perhaps for HNO3 the green line provides some useful information, but it is largely meaningless for the other products shown in Fig. 7. What would have been helpful to the reader would have been to show the multi-
year average, which is referred to several times in the text but which is not depicted in any of these panels.

– p33210, L4-7: “The altitude range covered by PSCs during 2010-2011 Arctic winter was also anomalous, with PSCs reaching altitudes above 30 km, as compared to maximum altitudes of 29 km previously observed (e.g. Poole & Pitts, 1994; Fromm et al., 1999; Massoli et al., 2006)”. As discussed above, I believe that the apparent MIPAS detections of high-altitude PSCs in January 2011 need to be validated before they are credible. In addition, I feel that the authors have mischaracterized the historical record in the sentence quoted here. The Arctic PSC sighting probabilities reported by Poole & Pitts [1994] drop to zero above about 26 km. Similarly, Fromm et al. [1999] report no Arctic PSC observations above about 25-26 km. Thus neither of these references can be used to support the assertion that PSCs have been observed in the Arctic up to 29 km. Finally, although PSCs in the Ny-Alesund record mainly appeared in the altitude range 20-24 km, they have been found as high as 28 km [Massoli et al., 2006] (but not 29 km). Thus the MIPAS detections of PSCs as high as 32-35 km are indeed “anomalous”, and, I would argue, dubious (at least until backed up by correlative measurements).

– p33210, L17-20: “NO2 at theta=450 K (Fig. 7f) showed sporadic, very high values, likely associated with evaporating PSCs, more often than previous years. In particular, the highest NO2 values were reached in the last week of February consistently with a period of minimum HNO3”. I don’t follow this argument. For one thing, 450 K seems a little high to me for renitrification, which has typically been reported at significantly lower altitudes. Also, wouldn’t evaporating PSCs lead to “higher” HNO3, not minimum values? The largest NO2 peaks occurred at the end of February/ beginning of March, which seems too early in the season for there to have been substantial HNO3 photolysis on such a rapid timescale.

– p33210, L22-26: “... the period of highest ClO (see Fig. 2d) which reached its peak value on 15 March. ClO then started to be converted into its reservoir ClONO2, as shown by the prompt increase of ClONO2 reaching previous years’ values. This prompt ClONO2 change points to reconversion of ClO into ClONO2 reservoirs rather than (or concomitant to) HCl”. I do not agree with the interpretation that the MIPAS data suggest reconversion into ClONO2 rather than HCl. Fig. 7 shows that ClONO2 in 2011 did not actually reach previous years’ values during the initial phase of chlorine deactivation. Although the ClONO2 values matched those in other years in early April, in previous winters that time period was well after the initial deactivation phase, at a stage when chlorine is being slowly repartitioned from ClONO2 into HCl (note the decrease in the values of the grey points between mid-March and early April in all other years). In contrast, in 2011 deactivation did not get underway to an appreciable degree until mid-March, as the authors note and Fig. 2d shows. Considering the strong ClO enhancement in 2011, had formation of ClONO2 been the primary initial deactivation pathway, ClONO2 values would have been very high by early April, similar to the highest values observed in mid-March in some other years. On the basis of Aura MLS measurements, Manney et al. [2011] argue that HCl reformation played a greater role in the chlorine deactivation process in 2011 than is typical for the Arctic, and to me the results of Fig. 7 support that argument. I think it is clear that, unlike in typical Arctic winters when ClONO2 reformation dominates, in spring 2011 both chlorine reservoir species were playing important roles.

– p33211, L1-19: I have several comments on this paragraph. "These characteristics are similar to the behaviour of the Arctic vortex in the winters 1995-1996 ... 1996-1997 ... and 1999-2000". What specific characteristics are similar? Depending on what is meant by that statement, 1996/1997 may not belong on the list, since it was not a particularly cold winter until very late in the season, and chlorine activation and consequent ozone loss were not as severe or extensive as in the other years. In the statement "although this winter the O3 reduction in the lower stratosphere (theta=450-500 K) was deeper and more broadly extended", does "deeper" refer to altitude (which wouldn’t make sense, since the sentence is specifically referencing a narrow theta range) or magnitude? Similarly, does "broadly extended" refer to the vertical or horizontal direc-

C14813
In 2004/2005, it is stated that the PSC season "was halted in mid February". The authors need to provide a citation for that, because I think that the PSC season lasted into late February at least; certainly chlorine remained activated at some levels in the lower stratosphere into early March (as the Santee et al., 2008 paper they reference shows). In the statement "the delayed reconversion of ClO into the ClONO2 reservoir", I would delete "ClONO2" and change "reservoir" to "reservoirs", since, as mentioned above, HCl also played a role. Again, what exactly does "wider PSC coverage" mean—horizontally or vertically or both? The authors note that "The observed 2011 denitrification appears to have had a greater role than in previous years". They do not need to speculate, since Manney et al. (2011) (which should be cited in this sentence) demonstrated conclusively that the more severe denitrification in 2011 partly contributed to the greater ozone loss. Finally, they state that "the much larger fraction of STS/Mix PSC observed suggests their active contribution in driving the lower stratospheric chemistry". The recent WMO report pointed out that chlorine activation primarily occurs on liquid aerosols, especially in the Arctic, and it would be good to reference that report here also.

– p33212, L3-5: "Averages were performed on pressure levels and respectively over the 75-90 N and 75-90 S geographical latitude, so as to reflect both chemical and dynamical changes of the vortex". I am not sure why the authors would consider mixing together the effects of chemical and dynamical processes, as is done in these broad latitude band averages, to be helpful in comparing the two hemispheres. I would think that would just complicate the comparisons and render them much less meaningful, especially since the Antarctic vortex is much larger (and generally more symmetric with latitude) than the Arctic vortex.

– p33213, L18-19: "In the middle stratosphere (theta=700-850 K) O3 was depleted by 25% down to 3.3 ppmv, at the lower edge of the 2003-2010 range". Although this high-altitude ozone depletion was noted in the O3 subsection (3.3.3), the comparison to previous Arctic winters was not discussed in the relevant section (section 4.1), so it seems odd to mention it in the Conclusions. In addition, since this depletion is presumably caused by gas-phase processes (not heterogeneous chemistry on PSCs), do the authors have an explanation for why it is larger in 2010/2011 than in most other years?

– p33213, L26 - p33214, L1: In discussing the Manney et al. (2011) results based on Aura MLS measurements, the authors state that: "Comparison of our ClONO2 to their HCl trends suggests 2011 Arctic ClO deactivated into ClONO2 rather than HCl which did not show the same prompt increase". I think the authors are misinterpreting the MLS results slightly. Manney et al. did not argue that HCl was the only or even the primary reservoir to be formed during chlorine deactivation in 2011 — only that it played a bigger, more Antarctic-like role than in typical Arctic winters.

– Overall comment on the Conclusions: Since this manuscript was submitted, another
paper on the 2010/2011 Arctic winter has been published [Sinnhuber et al., GRL 38, L24814, doi:10.1029/2011GL049784, 2011]. The authors may not have known about this GRL paper prior to its publication, but since it is also based on MIPAS measurements (albeit from a different retrieval), I really feel that some mention of how their results and these agree is necessary.

– p33224, Fig. 5: I do not understand the relevance of the insolation snapshots provided in Fig. 5(e-h). The information on PSC classification relative to the potential NAT/STS formation region is of some interest, but the average number of sunlit hours for these particular dates seems almost completely irrelevant. Evidently the authors did not find these maps of daily sunlit hours particularly useful either, as they did not discuss them at all in the text of the manuscript.

Typos and other minor wording issues:

– throughout the manuscript: "associated to" should be "associated with".
– p33193, L16: The term "lowermost stratosphere" has a specific definition (the region between the tropopause and the 380 K isentrope) that I do not believe is the intended meaning here. It should just be "lower stratosphere".
– p33194, L22: "anomalies induced by horizontal gradients adopting 1-D codes". "adopting" is not the right word here. Perhaps something along the lines of "anomalies induced by horizontal gradients not accounted for by conventional 1-D codes" would be better.
– p33195, L4: It would be better to change "injected" to "inserted" and "angle" to "inclination".
– p33196, L8: "assumes the atmosphere homogeneous" should be "assumes that the atmosphere is homogeneous".
– p33198, L10-11: "into MIPAS spectra" should be "in MIPAS spectra".
– p33199, L7-8: "because of their spectral signature similar to" should be "because of the similarity of their spectral signature to".
– p33199, L26: "adopted vortex averages" should be "calculated vortex averages".
– p33200, L18: "fraction to" should be "fraction of".
– p33201, L1-2: "Only when PSCs disappeared in March the regions of deepest O3 depletion around $\Theta=450$ K could be fully observed" should be "Only when PSCs disappeared in March could the regions of deepest O3 depletion around $\Theta=450$ K be fully observed".
– p33201, L18: "centred" should be "pole-centred".
– p33201, L21: "monotonical" should be "monotonic".
– p33203, L17: "panels f to i" should be "panels e to h"
– p33203, L20: "Beside" should be "Besides".
– p33203, L23: "largely scattered" – it would be better to say "highly variable".
– p33203, L20 to p33204, L29: This is a very long paragraph. It would be easier for the reader if this discussion were broken up into separate paragraphs for periods (i), (ii), and (iii, iv).
– p33204, L18: "82% PSCs" should be "82% of PSCs".
– p33204, L24-25: "scales" should be "scale" and "continue" should be "continuous".
– p33204, L26-27: Since for all of the other periods the STS/Mix percentage was stated first, and then the NAT percentage, it would be better to keep that same ordering for period (iv) as well.
– p33205, L9: "On the contrary" should be "In contrast".
– p33205, L22-23: "As a result of PSC formation, HNO3 was significantly removed
from the lower stratosphere from January to April. I think it is necessary to add “and subsequent sedimentation of PSC particles” after “PSC formation”, since PSCs form in all cold Arctic winters, but rarely is HNO3 actually removed from the lower stratosphere through denitrification (usually it is returned to the gas phase when the PSCs evaporate).

– p33205, L26: "successive" should be "subsequent".

– p33206, L23-25: "activation of ClO in the vortex, reaching a maximum in mid-March (Fig. 2d). This is also shown by ClO data at theta=550 K”. This wording is slightly confusing because Fig. 2d also shows ClO at 550 K.

– p33206, L25: "ClO sporadic high values" should be "sporadic high ClO values".

– p33207, L10: "showed a very stable O3" would read better as "showed little change in O3".

– p33207, L20-23: "Comparison ... results in a chemistry-driven depletion" should be "Comparison ... results in an estimate of the chemistry-driven depletion".

– p33208, L19: "a relative flat distribution" should be "a relatively flat distribution".

– p33208, L25-28: "Loss of HNO3 severely acted on a fraction of vortex air in late January and February, with greatly scattered low values, especially after the SSW on 3 February. It then turned into a more homogeneous low vortex HNO3 in March and early April. I’m not sure what “severely acted on” means, maybe “had a large effect on”? Also, why only “a fraction of vortex air”? The last sentence would be better written as "Low HNO3 was then more homogeneously distributed in the vortex in March ...”.

– p33209, L22-23: "the weak ascending trend observed in typical years”. It would be clearer to say "ascending trend observed in H2O in typical years".

– p33210, L11: "persisting also when temperatures rose" would be better as "and remained low when temperatures rose".

C14819

– p33212, L5: "Next to" should be "In addition to".

– p33212, L19-20: "Beside very similar ..." should be "Despite very similar", "pronounced" should be "pronounced", and "associated with" should be "consistent with".

– p33213, L16: "84% PSCs" should be "84% of PSCs".

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 33191, 2011.