

Interactive comment on “Factors controlling Arctic denitrification in cold winters of the 1990s” by G. W. Mann et al.

G. W. Mann et al.

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- > Interactive comment on "Factors controlling Arctic denitrification in cold winters of the 1990s" by G. W. Mann et al.
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- > Anonymous Referee 1
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- > General Comment
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- > This paper describes the complex vortex-dynamical conditions that are necessary for severe Arctic denitrification to develop. Since denitrification has a controlling effect on Arctic chemical ozone depletion, it is important to study the conditions where this could occur.

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> In particular, it is important for larger-scale models (e.g. chemical-climate models) to be able to represent denitrification for better estimates of chemical ozone depletion in a future climate which would require more accurate parameterizations of denitrification. Although this study does not provide directly applicable parameterizations the paper, in a very comprehensive way, points out the necessary conditions for denitrification to develop. It is beyond the scope of the present paper to make the necessary comparison with observed denitrification, but the presented results offer good opportunities to pursue this. The developed concept of closed-flow, which is shown to have close links with denitrification, would also be an interesting parameter to use in analyses of chemical ozone depletion in addition to other parameters e.g. PSC-areas. The paper is clearly written and of interest to the broad stratospheric research community. I can recommend it to be published after some minor comments have been considered.

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> Specific (line-by-line) comments:

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> p. 2558, l. 26 : I would remove "NAT", and write on p. 2559, l. 2 after the parenthesis "presumably composed of NAT". It is being said so often that the large particles are composed of NAT that people begin to take this for a fact.

We have clarified this sentence to more accurately reflect this concern.

> p. 2559, l. 23 : which winters is the word "these" referring to?

We have clarified the references to observations of denitrification in previous winters.

> p. 2560, l. 26 : I think it would be relevant to mention, that if the PSCs were characterized by mixtures of liquid STS particles and higher number

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- > concentrations of smaller solid type PSCs, the denitrification would be
- > much smaller. I guess it is only because you have such low number
- > concentrations of NAT particles in competition with the liquids for the
- > available HNO_3 that the NAT particles grow so large in size.

We have added a figure showing the sensitivity of denitrification to the nucleation rate and thus also to the change in number density and particle size (discussed at the end of the Winter 1999/2000 section). We feel, when considered in addition to the reference to the Jensen et al. (2003) paper, we have taken account of this point.

- > p. 2561, l. 21 I guess denitrification means removal to total HNO_3
- > (gas+condensed). Also I assume that the passive runs include particle
- > formation (but no sedimentation). This is not completely clear and has a
- > bearing on the next point.

Yes, denitrification does mean removal from total HNO_3 (gas + condensed). Also, yes the passive runs do include particle formation but not sedimentation. We have clarified both these in the paragraph which described these aspects of the model.

- > p. 2563, l. 2: I guess the fields of NAT-supersaturation (panels b in
- > Figure 1-4+6) are calculated using the actual partial pressure of HNO_3
- > (and not the initial values). But then how can fields of
- > NAT-supersaturation develop when you have strong denitrification (e.g. Fig
- > 1 around day 25)? I am confused here, because you mention on p. 2570, l.
- > 6-9 that denitrification causes areas to develop with very low mixing
- > ratios of HNO_3 , preventing the NAT particles to form (which is true, of
- > course). This seems to contradict co-located fields of
- > NAT-supersaturation.

Firstly, yes the fields of NAT-supersaturation are calculated using the actual (modelled) partial pressure of HNO_3 (not the initial values). The referee asks how fields of NAT supersaturation can develop under conditions of very strong denitrification. At the loca-

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tions where very strong denitrification has occurred (leaving concentrations of HNO_3 1 ppbv) the air will often no longer be able to be supersaturated with respect to NAT — this effect is included in our calculation of NAT area. However, at any given level, even when the vortex average denitrification has been very strong, there are usually significant areas with sufficient HNO_3 to have NAT supersaturation. The reduction in HNO_3 by denitrification is however incorporated into the calculation of TNAT and the areas with $T < \text{TNAT}$. This NAT area would often be higher later in the winter if we had calculated TNAT using the initial field of HNO_3 . We have clarified this in the text.

> I think the confusion arises because it seems that you do not discriminate
> in the discussion between the cold pool and NAT-supersaturated regions.
> They are of course co-located early in the winter, but when
> denitrification develops the NAT supersaturated diminish inside the cold
> pool. You begin the discussion on page 2567, l. 23 with a comparison of
> the vortex location and the NAT-supersaturated region, but then define the
> fraction of the cold pool that is in closed flow (p. 2569, line 13) from
> the temperature fields and continue the discussion based on this
> parameter. The discussion is much clearer to me if you by the
> NAT-supersaturated region mean where temperature occur below T-NAT; T-NAT
> being based on the initial HNO_3 mixing ratios, i.e. simply low temperature
> regions.

We understand this point and agree that our interchange of the terms "NAT-supersaturated region" and "cold pool" may be confusing and not strictly correct. We have preferred however to keep the NAT-area term to describe the area where air is supersaturated with respect to NAT as this incorporates the feedback that strongly depleted HNO_3 areas cause in suppressing further denitrification. As such we have replaced all references to cold pool with NAT area or NAT region to further clarify the concept of closed flow fraction.

> p. 2570, line 16: Regarding the vertical structure of the closed flow, I

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> think it would be interesting to plot the c-flow parameter into figures
> 1-4+6. I assume that the depth of the c-flow parameter would have an
> influence on denitrification in the sense that a larger vertical depth of
> high c-flow would allow particles to sediment over longer vertical
> distances without evaporation. Could the depth of the c-flow reduce the
> scatter in Fig. 8?

Yes, the depth of closed flow is an important factor. We have, to some extent, taken account for this when we take the mean of the denitrification rate and the closed flow fraction over the depth of the non-renitrifying part of the NAT region (i.e. where there is no renitrification and where $T < TNAT$). If at that level all of the air is in through flow (and hence only denitrifies a little), the c-flow fraction will be zero but will still be included in the mean of the closed flow — so a shallower closed flow area will have a lower mean c-flow fraction.

But, we have tried to improve the figure further by modifying the vertical region over which we take the mean of the closed flow and the denitrification rate to be the mean over the entire vertical range between 350 and 600K

(except for levels which are net renitrifying which we ignore). This increases the extent to which we incorporate the effect of depth in the scatter plot and also reduces the scatter because particles forming in a shallow closed flow region in a shallow NAT region (for example between days 40 and 60 in 1999/2000) would in the previous plot have had a higher mean closed flow fraction but a low denitrification rate because the layer is so shallow. Now, since we have extended the range over which the mean is taken, the mean closed flow fraction is reduced for such shallow layers (the closed flow fraction is zero where there is no NAT area) and the outliers are "moved to the left" in Figure 8 (now Figure 9).

Note that we have also made a change to Figure 8 (now Figure 9) in response to Refree 2's comments by changing the mean denitrification rate to be mean denitrification

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rate (as a % of available HNO_3) rather than absolute denitrification rate. This has also helped to reduce the scatter in Figure 8 (now Figure 9) which now has an improved correlation. The remaining scatter results from 2 factors: i) the time scale over which the mean closed flow fraction changes can be much less than it would take for the system to respond to the change. For instance a sudden change from a partially closed flow to a near-complete closed flow and then back again (say over 2 days) would not result in a sufficiently high increase in denitrification rate because it takes a number of days for the particles to grow, sediment and cause denitrification in response to the closed flow situation. To remain in near-complete closed flow for just one or two days is not sufficient even when a time lag of 3 days is included. ii) the variation of closed flow in the vertical — a situation where the NAT region is in closed flow between 400 and 450K and between 500K and 550K (but in through flow between 450 and 500K) would not create large denitrification even though it will produce a relatively high mean closed flow fraction because particles which are formed between 500 and 550K will evaporate when they fall below 500K before they can enter the lower closed flow volume. So the topology of the closed flow volume may also be important.

- > p. 2560, line 16-18: and p. 2572 l. 13-21: The authors might discuss in
- > somewhat more detail what meteorological conditions cause the vortex and
- > the cold pool to be concentric, e.g. could there be a connection to the
- > NAO or some other large-scale parameters?

We have added a little more discussion on p. 2572 suggesting that further research into what meteorological conditions could give rise to a prolonged concentric vortex in future climates would be interesting.

> Technical corrections:

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- > Fig 8: Figure 8 in the printed version is not very clear. Perhaps using
- > filled and somewhat larger symbols could improve this.

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We have made the symbols in Figure 8 (now Figure 9) filled.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 2557, 2002.

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2, S1068–S1074, 2002

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