Problems with calculation of archiving lifetime and number of recyclings

When I wrote my main review of this paper, I confessed to some conceptual uncertainty about the meaning of τ_{arch}, and some surprise about the number of recyclings estimated by the authors (page 6892, para 2 and many subsequent parts of paper). I have now thought some more about this,, and have convinced myself there is a problem that the authors need to address. I hope that others who have considered this problem might come in, either to explain more formally what the correct calculation is, or to explain why I have got it completely wrong.

Tau-arch of course can be calculated, and (m_{50cm}/FA) is certainly the time it would take to deplete the 50 cm box purely by archiving nitrate. However the authors assert that this is the lifetime of nitrate in the top 50 cm and therefore use this to estimate the number of photochemical recyclings each nitrate ion has undergone. What struck me as odd about this calculation is that is blind to the magnitude of the primary input (FPI=FT+FS), or f_{exp}. So let's do some thought experiments in which the some of the values in Table 5 are varied, but still within physically allowed ranges. I have included the value of m_{50cm} and a calculated f_{exp}. All units are mg (or 10^{-6} kg) N m^{-2}, or mg N m^{-2} a^{-1} as in the paper. Case A is the one in the paper. Cases B and C are the extreme ends of what is possible. In each case the amount in the top 50 cm and the amount archived (FA) are kept the same as in the base case. But FPI is varied between the minimum that allows mass balance (FA), and a very large number. I simply have to alter FE (and hence f_{exp}) to balance the calculation.

	A (as in paper)	B	C
m_{50cm}	8.3	8.3	8.3
FP	32.00	32.00	32.00
FD	32.16	32.16	32.16
FE	8.04	999.84	0
FA	0.16	0.16	0.16
FPI	8.2	1000	0.16
f_{exp}	20%	Close to 100%	0%

Because of the fast photochemistry, the 50 cm firn box and the atmosphere are essentially a single box as far as mixing with the primary input is concerned. In case B there is a huge primary input. When nitrate in the firn is photolysed, it is quickly swamped by new (FPI) input, and whatever is deposited is almost entirely new input. In this case the lifetime of nitrate in the combined firn/atmosphere box (ignoring the physical transit time for an ion to be advected with the snow from the skin layer to below 50 cm) is close to zero, and all the ions that survive have likely never been recycled (zero recyclings).

In case C on the other hand, the primary input balances FA and the only way an ion can escape the combined box is by being archived. In this case the lifetime in the combined box is indeed 50 years, but that is calculated as FP/FPI, not as FP/FA as in the calculation of τ_{arch} in the paper. The number of recyclings in this case is very high and may well be 120.

So, here is the point. The lifetime of nitrate (or perhaps I should say of NOy) in the combined 50cm/atmosphere box is NOT m_{50cm}/FA, but m_{50cm}/(FA+FE) = m_{50cm}/FPI, and this is what determines how old the nitrate in the box is and how many recyclings it has experienced. For the base case in

the paper this is then of order 1 year and the average number of recyclings will be of order 10 I suppose.

The critical point here is that the lifetime in a box is based on all the processes involved (Jacob 1999, eq 3.1). What we need to know the age of NO_y (the sum of nitrate + NO_x, for which we don't care about photolysis to NO_x except in so much as it mixes the 50 cm and atmosphere box) is, analogous to Jacob's eq 3.1

$\tau = m_{50cm}/(F_{out}+D)$ (L is zero in this case as there is no chemical loss of NO_y), ie

$\tau = m_{50cm}/(FE+FA)$.

What the authors calculate as τ_{arch} is m_{50cm}/FA which is the lifetime against archiving, but is not the one you need to estimate the age of the archived nitrate!

Please tell me how I am wrong, but I think I am not! I am unsure why your model takes 20 years to equilibrate, but given that it takes around 15 years for surface snow to sink through the 1 metre snow model domain, it may simply be that. If I am right, the paper needs changing in several places including Fig 1, page 6938, 6936 and especially 6892, but the whole paper should be checked for errors based on this.