Response to Reviewer #1 of submission of:

Changes to the chemical state of the northern hemisphere atmosphere during the second half of the twentieth century by Newland et al., 2017, submitted to ACPD

General Response

We thank the referee for giving their time to make comments helping to clarify and improve our manuscript. Responses to each point are given separately beneath that point. The referee's comments are bold and italic, the author's comments are inset in plain type.

With respect to the primary concern of the referee, we are not sure whether there has been some misunderstanding over the nature of our use of the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ or whether it is that the referee has concerns over the validity of the use of an average value for this ratio. We hopefully address both concerns in our responses.

While we present a lot of work in response to the referee's major concern, we do not feel that much of it belongs in the paper beyond clarifying that the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ is representative of a mean value during transport.

Major issues:

1) The fatal problem with this paper is that the derivation of Equations E2 and E3 is flawed. Thus, the conclusions of this paper that are based on these equations are also flawed. For this paper to be acceptable for publication, the authors must reconsider all analyses based on these equations, and revise the conclusions accordingly. Bertman et al. [1995] derived Equation E1 by integrating the differential equation for the time rate of change of the alkyl nitrate concentration, which required approximations that were valid in an NOx rich environment. To extend Equation E1 to yield Equation E2 as the authors have done is not valid. To be valid, the original differential equation of Bertman et al. [1995] must be properly modified to include Reaction 11, and then properly integrated.

> The ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ as used in Equation E2 (and subsequent rearrangements) represents an average over time, t, in the same way as [OH], which is embedded in the k_A and k_B terms in the Bertman et al equations. When referring to this ratio as an average over time in the following discussion we shall label it γ.
>
> Below we show how we get from the rate equation for [RONO$_2$] presented in Bertman et al., to Equation E2.
>
> Bertman et al. start with the following rate equation (Equation 11 in Bertman et al):

$$d[RONO_2]/dt = \beta k_A[RH] - k_B[RONO_2] \qquad (T1)$$

where $\beta = \alpha_{13}\alpha_{14}$, $k_A = k_{13}[OH]$, $k_B = k_{15}[OH] + j_{16}$ (numbers relate to our manuscript)

N.B. In our manuscript $k_B = k_{15}[OH]$, i.e. we ignore the photolysis sink of the alkyl nitrates.

This assumes a NOx rich environment, i.e. that all peroxy radicals produced react with NO. This will not be the case for an air mass in transport to the Arctic. The peroxy radical could also react with HO_2. (This is of course still a simplification - the peroxy radical could also react with other peroxy radicals etc.). To account for this additional sink for the peroxy radical, we simply need to introduce the ratio of the sink of the peroxy radical to [NO] to the total peroxy radical sink. This can be seen by considering the production of $[RONO_2]$ in Equation T1 in terms of $[RO_2]$ (Equation T2).

$$d[RONO_2]/dt = [RO_2][NO]k_{14}\alpha_{14} - k_B[RONO_2] \qquad (T2)$$

At steady state, which is a valid assumption given the very short lifetime of RO_2:

$$[RO_2] = k_A\alpha_{13}[RH] / (k_{14}[NO]+k_{11}[HO_2]) \qquad (T3)$$

i.e. we now include the loss of RO_2 via reaction with HO_2 as well as with NO.

 Substituting T3 into T2 gives:

$$d[RONO_2]/dt = \beta k_A[RH]k_{14}[NO] / (k_{14}[NO]+k_{11}[HO_2]) - k_B[RONO_2] \quad (T4)$$

Replacing the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ with γ (which assumes the ratio to be a constant):

$$d[RONO_2]/dt = \gamma\beta k_A[RH] - k_B[RONO_2] \qquad (T5)$$

Note that T5 is exactly the same as Equation 11 in Bertman et al., with the exception of the inclusion of the term γ. In the same way that Bertman et al. assumed k_A and k_B to be constant (note they are both a function of OH), we also assume γ to be constant. We discuss this assumption in a response to a later comment made by the reviewer. The first term on the right hand side of T5 is now [RH] multiplied by a constant equal to $\gamma\beta k_A$ as opposed to being multiplied by a constant equal to βk_A in Bertman's equation.

We can then begin to integrate Equation T5. For this we will use integrating factors:

I.e. if we have an equation of the form:

$$dy/dx + f_1(x)y = f_2(x)$$

then the integrating factor (IF) is:

$$e^{\int f1(x).dx}$$

and:

$$y = \int IF.f_2(x).dx \, / \, IF$$

Replacing [RH] in Equation T5 with $[RH]_0 e^{-kAt}$ we get:

$$d[RONO_2]/dt = \gamma\text{ß}k_A[RH]_0 e^{-kAt} - k_B[RONO_2] \quad\quad (T6)$$

For this equation then:

$$f_1(x) = k_B$$

$$f_2(x) = \gamma\text{ß}k_A[RH]_0.e^{-kAt}$$

$$IF = e^{\int kB.dt} = e^{kBt}$$

Hence:

$$[RONO_2] = \int e^{kBt}.\gamma\text{ß}k_A[RH]_0.e^{-kAt}.dt \, / \, e^{kBt} \quad\quad (T7)$$

Solving this gives:

$$[RONO_2] = \gamma\text{ß}k_A[RH]_0 e^{-kAt} \, / \, (k_B-k_A) + c.e^{-kBt} \quad\quad (T8)$$

At t=0:

$$[RONO_2]_0 = \gamma\text{ß}k_A[RH]_0 \, / \, (k_B-k_A) + c$$

$$c = [RONO_2]_0 - \gamma\text{ß}k_A[RH]_0 \, / \, (k_B-k_A)$$

So:

$$[RONO_2] = \gamma\text{ß}k_A[RH]_0 e^{-kAt} \, / \, (k_B-k_A) + e^{-kBt} ([RONO_2]_0 - (\gamma\text{ß}k_A[RH]_0 \, / \, (k_B-k_A))) \quad (T9)$$

Replacing $[RH]_0.e^{-kAt}$ with [RH]:

$$[RONO_2]/[RH] = \gamma\text{ß}k_A.(1-e^{(kA-kB)t}) \, / \, (k_B-k_A) + [RONO_2]_0.e^{(kA-kB)t} \, / \, [RH]_0 \quad (T10)$$

Assuming $[RONO_2]_0 = 0$:

$$[RONO_2]/[RH] = \gamma\text{ß}k_A.(1-e^{(kA-kB)t}) \, / \, (k_B-k_A) \quad\quad (T11)$$

I.e. Equation E2 from the paper.

Equations T10 and T11 are exactly the same equations as derived by Bertman et al. (Equations 12 and 13), with the exception of the inclusion of the constant γ. This result can be expected since the constant factor ßk_A has simply been replaced by the constant factor γßk_A.

We have edited Section 4 somewhat to reflect this discussion, demonstrating that Equation E3 comes from a version of the rate equation for $[RONO_2]$

As it stands, it is clear that Equation E2 cannot be correct. The left side of the equation is a ratio that is determined by long time scale evolution, while the right side depends on NO and HO₂ concentrations, which are variables that vary on very short time scales. For example, if the NO concentration were suddenly increased by fresh emissions, Equation E2 indicates that the [RONO2]/[RH] ratio would change, which is obviously incorrect.

As stated above, γ represents an average of the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ over the time t. The air that arrives at the firn drill site will inevitably be a mix of air masses and will have experienced emissions from a number of different source regions. Furthermore, the air extracted from the firn is also a mix of ages and a firn diffusion model is used to derive a time series. As such the $[RONO_2]/[RH]$ ratios derived from the firn air represent an integrated effect of the chemical and transport processes. What is really interesting and of value exploring is that these ratios have changed over the period of the firn air record, which suggests that there may have been a change in this rate of processing. In the paper we discuss what these changes might have been. Equations E2 and E3 illustrate that if the air arriving at the firn site in the 1970s had experienced a different mean $[NO]/[HO_2]$ to the air in the 1990s, this would affect the observed $[RONO_2]/[RH]$ ratios.

*It might be (or might not be) that a proper integration of the appropriate differential equation could yield an equation that looks like Equation E2, where the NO and HO2 concentrations would be weighted time integrals, but those weightings are likely to be different for each species. How those weightings might be calculated is not clear. Equation E3 is derived from Equation E2, so the problems in the latter propagate to the former. The left side is now a ratio of highly variable species, while the right side depends on concentrations that change relatively slowly.*

It is true that changes to the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ at different times along the air mass trajectory, t, will affect $d[RONO_2]/dt$ at that time differently because $d[RONO_2]/dt$ is also driven by $[RH]$ which is a function of time. However, the uncertainties introduced by the assumption of γ as a constant on $[RONO_2]/[RH]$ calculated at time = 10 days are on the order of 5 % (as demonstrated below), while the observed changes in $[RONO_2]/[RH]$ in the firn are considerably larger, on the order of a factor of 4 − 5. Hence we consider the assumption of γ as a constant to be a reasonable assumption for the sake of making the problem tractable and that the changes to $[NO]/[HO_2]$ that we calculate in the paper are not an artefact of this assumption.

Using Equation T4 (i.e. allowing the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$) to vary with time rather than it being a constant as in Equation T5) we can examine the magnitude of changes to [2-butyl nitrate]/[n-butane] in pairs of scenarios where γ is the same for each pair but the time evolution of the ratio ($k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$) is different. Figure T1 illustrates how the ratio ($k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$) varies over the 10 days for each of six hypothetical scenarios: A1, A2, B1, B2, C1, and C2. γ is the same for both A scenarios and likewise for both B scenarios and both C scenarios. The time evolution of [RH] is calculated as $[RH]_t = [RH]_0 e^{-k[OH]t}$, and [NO] as $[NO]_t = [NO]_0 e^{-t/\tau} + [NO]_{bg}$, where τ is the NOx lifetime (assumed to be 4 hours (e.g. Liu et al., 2016, ACP)) and $[NO]_{bg}$ is the assumed [NO] in the background atmosphere, and $[NO]_0 = 2.5 \times 10^{11}$ cm^{-3}.

The results are presented in Table T1 where we show the percentage difference in $[RONO_2]/[RH]$ at time t = 10 days for each pair of scenarios A, B and C. Scenarios A and B represent 'realistic' scenarios, in that the ($k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$) at time t=0 is ~ 1 and falls to some sort of background value during transport away from an NO source region. This is what can be expected since RH sources are generally co-located with NOx sources (i.e. emissions from motor vehicles) and hence [NO] concentrations are high close to the RH source. Scenario C represents an extreme scenario, but unlikely case, in which $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ is 0 for the first half of the transport time and 1 for the second half, or, 1 for the first half of the transport time and 0 for the second half. I.e. γ = 0.5 in both cases. Such a scenario is purely hypothetical and serves only to demonstrate the upper range of uncertainty that could arise from the assumption of an average ($k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$). We also consider the results using a range of [OH] in Table T1.

The results in Table T1 show that even though the time evolution of the ratio $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ varies, if the average value, i.e. γ, is the same there is only a small percentage difference between the resulting values for $[RONO_2]/[RH]$. The differences increase with increasing assumed [OH], but even for the largest value of [OH] for Scenarios A and B the greatest difference in $[RONO_2]/[RH]$ at time t = 10 days is < 5%. For Scenario C the difference is up to 35%. However, it is noted that the scenario, in which $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ is 0 for the first half of transport and 1 for the second half must be considered extremely unlikely.
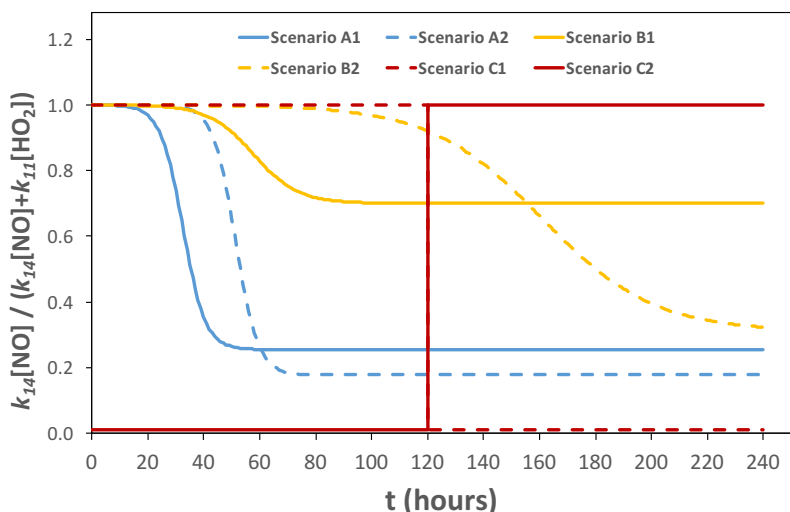
Figure T1 Six hypothetical scenarios with different time evolution of the $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ ratio but where γ is the same for both A scenarios and likewise the same for both B and C scenarios.

Table T1 Percentage difference in $[RONO_2]/[RH]$ after 10 days between the scenarios within each pair shown in Figure T1 for $[OH] = 2, 4$ and $6 \times 10^5 \, cm^{-3}$. $[RONO_2]/[RH]$ calculated using Equation T5. $[RH]$ calculated as $[RH]_t = [RH]_0 e^{-k[OH]t}$.

$[OH] \, / \, cm^{-3}$	2×10^5	4×10^5	6×10^5
Scenarios A (γ=0.36)	1.2	2.5	4.8
Scenarios B (γ=0.77)	1.3	2.4	3.4
Scenarios C (γ=0.5)	14.0	24.7	34.7

The authors are evidently implicitly assuming that the NO and HO2 concentrations are some sort of remote, hemisphere wide average (as indicated by the statement on lines 11-12 of page 13). However, this is far from clearly correct, as the production of the alkyl nitrates may well be dominated by relatively rapid photochemistry in the urban, NOx-rich, higher OH environment, rather than photochemistry in the remote environment. Hence, properly integrating the differential equations describing the photochemical processes is critical to correct analysis. That integration may not be possible to carry out analytically.

In urban areas, close to the RH source, there is no scope for a change in the production efficiency of alkyl nitrates because $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])) \sim 1$ (i.e. production is RH limited). In the rural / background environment, where $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])) < 1$, changes to [NO] would have an effect on the alkyl nitrate production efficiency.

That being said, an increase in urban NOx would have a small effect in terms of $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]))$ remaining at ~ 1 for a longer time during air mass

transport. Using Scenarios A1 and B1 (above), an increase of $[NO]_0$ of two orders of magnitude (i.e. to 2.5×10^{13} cm^{-3}) yields changes in $[RONO_2]/[RH]$ at time t = 10 days of 18% for Scenario A1 and 6% for Scenario B1, assuming $[OH] = 6 \times 10^5$ cm^{-3}.

Figure T2 shows the time evolution of $[RH]$ (in terms of % remaining compared to $[RH]$ at t=0), % of total RO_2 produced, and $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]))$, for Scenario A1 above ($[NO]_0 = 2.5 \times 10^{11}$ cm^{-3} (i.e. 10 ppb), the NOx lifetime in the air mass = 4 hours, background $[NO] = 1.9 \times 10^7$ cm^{-3}, $[HO_2] = 2 \times 10^7$ cm^{-3}, initial $[RH]$ = 800 ppt, $[OH] = 6 \times 10^5$ cm^{-3}). It is seen that the vast majority (i.e. 86%) of the RO_2 produced over the 10 days is produced when $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])) < 1$. (If $[OH]$ were lower than 6×10^5 cm^{-3} then the amount of RO_2 produced when $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])) < 1$ would be even larger.)

This shows that there is a large scope for changes in the ratio $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]))$ away from sources to affect alkyl nitrate production efficiency during air mass transport. Figure T3 shows the time evolution of the $[RONO_2]/[RH]$ ratio for the scenario shown in Figure T2. It also shows the $[RONO_2]/[RH]$ calculated assuming $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]))$ to remain at 1 for the whole transport time. Figure T2 shows that for the scenarios shown, an increase in γ has the potential to increase $[RONO_2]/[RH]$ at t = 10 days by a factor of 2.5.
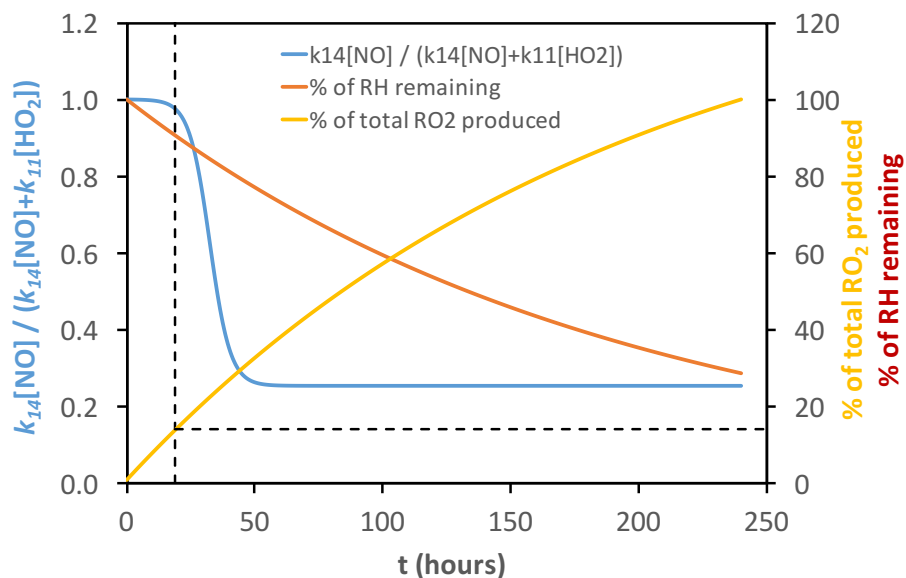
Figure T2 The time evolution of $[RH]$, % of total RO_2 produced, and $(k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]))$, calculated using $[NO]_0 = 2.5 \times 10^{11}$ cm^{-3}, NOx lifetime = 4 hours, and $[OH] = 6 \times 10^5$ cm^{-3}, background $[NO] = 1.9 \times 10^7$ cm^{-3}, and $[HO_2] = 2 \times 10^7$ cm^{-3}. The vertical dashed line represents the time when $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]$ drops below 0.98, and the horizontal line indicates that only 14% of the RO_2 is produced when $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2]$ is close to 1.
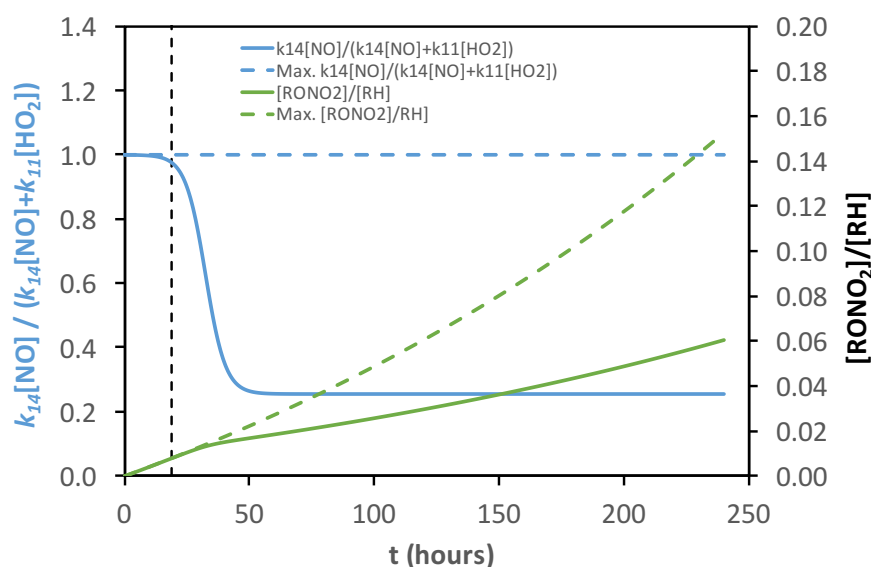
Figure T3 The time evolution of [RONO$_2$]/[RH] for the scenario shown in Figure T2. Solid blue line shows (k_{14}[NO]/(k_{14}[NO]+k_{11}[HO$_2$])) from Scenario A1, dashed blue line shows maximum possible (k_{14}[NO]/(k_{14}[NO]+k_{11}[HO$_2$])), i.e. equal to 1 for the whole transport time. Solid purple line shows [RONO$_2$]/[RH] calculated using (k_{14}[NO]/(k_{14}[NO]+k_{11}[HO$_2$])) from Scenario A1 ([RONO$_2$] calculated with Equation T4, [RH] calculated as [RH]$_t$ = [RH]$_0$e$^{-k[OH]t}$, as above). Dashed green line shows [RONO$_2$]/[RH] calculated using (k_{14}[NO]/(k_{14}[NO]+k_{11}[HO$_2$])) = 1 for the whole run.

In summary, in our response to Referee #1's point 1, we have demonstrated that the [RONO$_2$]/[RH] ratios seen at the firn air site can be influenced by the [NO]/[HO$_2$] ratios during transport. Thus changes to the background concentrations of NO or changes to its lifetime that lead to an overall change in the average value of k_{14}[NO]/(k_{14}[NO]+k_{11}[HO$_2$] can result in changes in the ratios in [RONO$_2$]/[RH] at remote locations. We accept that there are large assumptions in the methodology in our paper, but we believe that the assumptions we have made are clear, justified and consistent with other assumptions inherent in Bertman et al.'s original equations and therefore are appropriate for interpreting the firn air data.

The nature of these data means that they represent averages of air masses arriving at the firn site from different regions and over extended periods of time. We use simplified equations to illustrate that changes in the [NO]/[HO$_2$] ratios could contribute to the decadal changes in the firn [RONO$_2$]/[RH] data. We believe this is an important point to make and adds to the growing evidence for changes in atmospheric chemical processing during this period, but we acknowledge that this needs to be further fully investigated through the use of more sophisticated models that can then take account of the complexities of the chemistry and transport.

2) It is unclear whether the uncertainties in the estimated diffusion coefficients of the alkyl nitrates have an important impact on the atmospheric history reconstructions. This should be discussed.

Author response:

Figure T4 shows the three alkyl nitrate scenarios presented in Figure 2 in the paper. A red dashed line is included using diffusion coefficients calculated using the method of Chen and Othmer (1962) which gives diffusion coefficients some 10% lower than those calculated using the method of Fuller et al. (1966) which are used for the work presented in the manuscript. It is seen in Figure T4 that the difference between the scenarios derived using the two differently calculated diffusion coefficients are minor and well within the uncertainty envelopes.
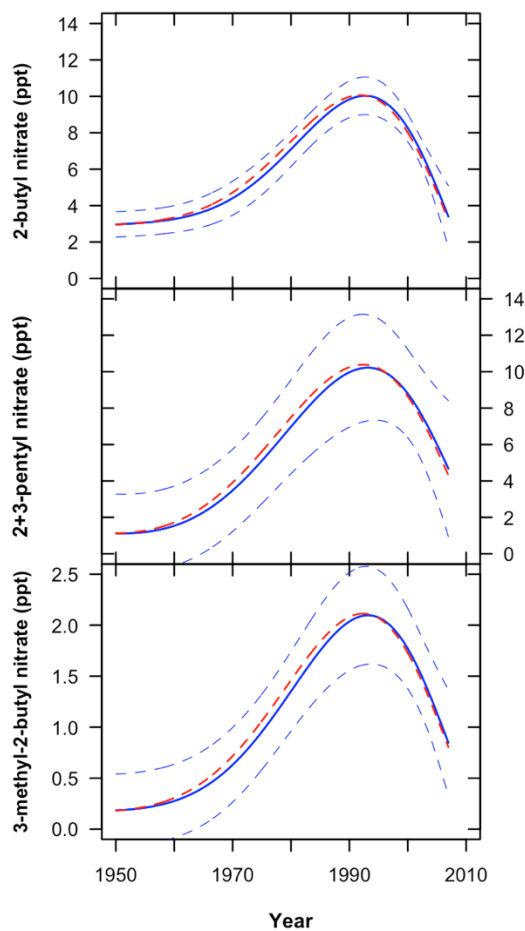
Figure T4 As Figure 2 in manuscript but including red dashed line showing effect of 10% decrease in diffusion coefficient used in firn modelling.

We have included the following additional text in the third paragraph of Section 2.3 (Firn Modelling):

"Model runs were also performed using diffusion coefficients for the alkyl nitrates calculated using the Chen and Othmer method. These coefficients are ~ 10% lower than those calculated using the Fuller method. However, the atmospheric scenarios derived from the modelling are very similar, well within the 2-σ uncertainty envelopes presented in Figure 2."

3) On page 7 "It is noted that the latter part (post-1995) of the model derived scenarios for 2+3-pentyl nitrate is rather sensitive to the inclusion or exclusion of the measurement at 34.72 m (the most shallow measurement used). The scenarios presented in this work are based on including this measurement." Is this an important issue? The authors should discuss the effect on the final conclusions if this measurement is excluded.

There is not a strong argument for removing the data point, however we felt the need to make clear that there has been some consideration as to the possible effect of seasonality on the alkyl nitrate measurements at this depth. In Helmig et al. 2012, a series of tests were performed using the firn model to examine the effect of seasonality on the alkane measurements at the NEEM sites between 30 and 40 m depth (Figure 6, Helmig et al. 2014). The alkanes have a similar diffusion coefficient, and similar atmospheric seasonality, to the alkyl nitrates and so any seasonality effect would be very similar. It is seen in this figure that the effect at 35 m for the butanes and pentanes (the most similar alkane molecules to the alkyl nitrates presented here) is < 5 %.

If the measurement were affected by seasonality, the correct thing to do would be to apply a minor correction rather than to remove the point. These two processes would have very different effects. Removing the measurement has a significant effect on the derived reconstruction because of the limited size of the data set in that region. Correcting for any seasonality on the other hand would have a very minor effect on the derived reconstruction.

As stated in the paper, removing the measurement at 34.72 m would have a significant effect on the derived scenarios post-1995, particularly for 2+3-pentyl nitrate. This is because we are removing a constraint on a data set of limited size - the decreasing concentrations post-1995 for the two pentyl nitrates are constrained by relatively few measurements partly because of the increased scatter in the peak region (55 - 65 m) compared to 2-butyl nitrate. However, this is already demonstrated in our data presentation by the relative sizes of the 2-sigma uncertainty envelopes for the pentyl nitrates compare to 2-butyl nitrate (~ 10% at the peak for 2-butyl nitrate compared to 25 - 30% for the pentyl nitrates).

Minor issues:

1) Abstract lines 18-21 - I disagree with the statement "Due to their short atmospheric lifetimes, NOx and HOx are highly variable in space and time, and so the measurements of these species are of very limited value for examining long term, large scale changes to their budgets." If measurements of either HOx or NOx were measured in a given region in an extensive enough manner to characterize average ambient concentrations (e.g., during an intensive aircraft deployment such as NASA DC-8 studies), and if those extensive measurements were repeated after the passage of a decade or so, then it should be possible to quantify the long term, large scale changes to their concentrations. I suggest that this statement be removed, as it is superfluous to the paper.

Author response:

We have removed the word 'very' from the statement highlighted. However, we maintain the essence of the statement to be correct.

The study that the referee suggests would, in their words, require measurements that are in an "extensive enough manner" and if these were repeated then it would be possible to make an estimate of a long term trend. However, such a study requires such a large spatial and temporal coverage to be able to give meaningful results as to make it either prohibitively expensive or an extremely rare experiment. By far the majority of the measurements of NOx and HOx are not like this and so we stand by our statement that "measurements of these species are of limited value for examining long term, large scale changes to their budgets". We believe that this is a justifiable reason for alternative methods of examining the problem. This is entirely consistent with the rationale of using methyl chloroform for determining long term changes in OH.

2) Page 2, lines 13 and 14. I also disagree with the phrase "which is positively correlated with NOx concentrations in the background atmosphere through the photolysis of NO2 (Reactions R1-R2)." Ozone is certainly correlated with total NOy concentrations, but since ozone and NOx have very different lifetimes, they are in general only poorly correlated, because any correlation resulting from production through the photolysis of NO2 is destroyed by removal or fresh emissions of NOx, even in the remote troposphere. Here and elsewhere throughout the paper, I suggest that very careful consideration be given to the veracity of each sentence.

Author response:

Our intended meaning was that "ozone production" was positively correlated to NOx concentrations. We see that our statement was ambiguous so we will change the

sentence to "NO_X and HO_X are linked through ozone production, which is positively correlated with NO_X concentrations in the background atmosphere through the photolysis of NO_2 (Reactions R1-R2)."

3) Page 2, lines 23 and 24 - The phrase "The main removal processes for HOx are the reaction of OH with NO2 (Reaction R9) ..." has an error. Evidently this reaction has not been separately listed in the paper.

Author response:

The OH + NO2 reaction is already included but the referee is correct it is not Reaction R9, it is Reaction R5. This has been changed.

4) Page 4, line - Should be "on-road" vehicles.

Author response:

This has been changed to read '…from motor vehicles.'.

5) Page 6 - Sentence on lines 18-21 is not clear.

Author response:

The second 'samples' has been removed from line 19. The sentence hopefully now makes sense.

6) Page 8 - Line 12 - The Worton et al. (2010) reference is not in the References list; should be Worton et al. (2012)?

Author response:

The referee is correct. Changed.

7) Page 12, lines 19-22 are not exactly correct: In Equation E1, taken from Bertman et al. (1995), [OH] is assumed to be the average over the time t. This is exactly what the authors are doing as well. This should be clarified, i.e. the term [OH]t represents the time integral of [OH] over the period during transport from the source region to the Arctic.

Author response:

Lines changed to:

"In Equation E1, taken from Bertman et al. (1995), [OH] is assumed to be a constant. Similarly for the purposes of this work, [OH] is assumed to be constant and represents an average [OH] to which the air mass is exposed during transport from the source region to the Arctic."