Response to Reviewer #2 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.

Specific comments. Given the potential importance of the trends in SO2 emissions and their implied influence on the NOx budget (as outlined in section 5.1.2) I suggest that this topic be represented in a little more depth.

Specific suggestions. Section 5.1.2 NOx sinks: Given the potential significance of this section to the overall conclusions, it seems to be somewhat truncated compared to other parts. As the timing of the various trends appear to be crucial to the various evidence lines presented in this work, maybe the authors could add a figure (or perhaps combine with existing Fig 5?) to convey the time series for sulfate aerosol trends (eg like in Fig 8 in Smith et al., 2011) to more concretely illustrate when sulfur emissions peaked.

> Author response:
>
> We have included a new Figure in the manuscript (Figure 6) to show the SO_2 emissions 1950 – 2005 presented in Smith et al. (2011, ACP) and observed winter-time sulfate aerosol mass concentration in Europe (Turnock et al., 2015, ACP).
>
> We have altered the text of Section 5.1.2 to reflect the inclusion of this figure and have been a little more explicit in the regional nature of sulfate aerosol in relation to SO_2 emissions.

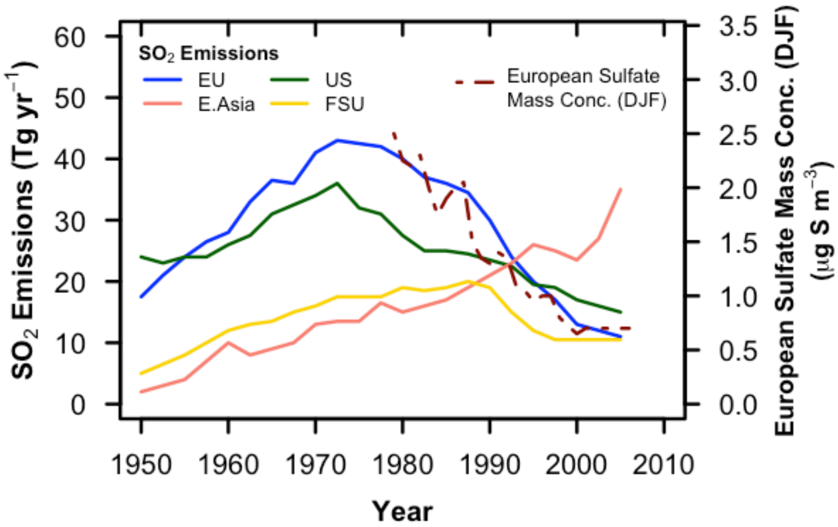

Figure 6 SO_2 emissions (Tg yr^{-1}) 1950-2005 from Smith et al. (2011), and mean European sulfate mass concentration (μg S m^{-3}) in winter (DJF) from Turnock et al. (2015). SO_2 emissions: Blue – Europe; Green – N. America (US + Canada); Gold – Former Soviet Union (Russia, Ukraine, others); Pink – E. Asia (China, Japan, S. Korea, others). Brown dashed line - mean European sulfate mass concentration in winter (DJF).

"A modelling study by Dentener and Crutzen (1993) predicted that changes to the loss of NO_x via sulfate aerosol could have a significant effect on northern hemisphere NO_x concentrations and that these changes would also affect O_3 and OH concentrations. Subsequent modelling studies, though often focusing on remaining uncertainties in the uptake coefficients, have broadly agreed with the magnitude of the changes suggested by Dentener and Crutzen (Brown and Stutz, 2012).

There has been a large decrease in sulfate aerosol observed in Europe and the United States since 1980 (Berglen et al., 2007; Turnock et al., 2015). Figure 6 shows the measured trend in winter-time (DJF) sulfate mass concentration presented in Turnock et al. (2015), with decreases of about 75% from 1979 to 2005. This decreasing trend has been driven by a ~70 % decrease in SO_2 emissions (Smith et al., 2011) from these regions (Figure 6). It is noted that while global SO_2 emissions have only decreased about 15% from the peak in the 1970s, due to rapidly increasing emissions in East Asia in recent decades, sulfate aerosol has a lifetime of about 5 days in the troposphere (and SO_2 of about 1 day) (Stevenson et al., 2003) and so aerosol concentrations will be largely driven by regional SO_2 emissions.

These large decreases in sulfate aerosol in Europe and the US (the main source regions for air masses arriving in the Arctic in the winter) would be expected to have led to a decrease in NO_X removal by N_2O_5 hydrolysis, and hence to an increase in the NO_X lifetime and atmospheric [NO_X]. The time period of decreasing SO_2 emissions and sulfate aerosol is broadly in line with the derived steep increase in the [NO]/[HO_2] ratio."

Stevenson, D. S., C. E. Johnson, W. J. Collins, and R. G. Derwent, The tropospheric sulphur cycle and the role of volcanic SO2, in Volcanic Degassing edited by C. Oppenheimer, D. M. Pyle and J. Barclay, Geol. Soc. Lond. Spec. Pub., 213, 295-305, 2003.

Technical corrections typing errors, etc.

Page 19, line 17 – I do not find the "Aydin et al." reference in the reference section

Author response:

Reference added.

Page 23, Line 23 – I do not find the "Dlugokencky et al." reference in the text.

Author response:

I think this was just a case of mis-spelling. Dlugokencky is now spelled correctly throughout the manuscript and appears in the text and references.

Additional Changes in Response to both Referees' Reviews

The following sentence has been added to the end of paragraph 1 of Section 5:

"*Since the term $k_{14}[NO]/(k_{14}[NO]+k_{11}[HO_2])$ is an average across the whole transport time it reflects both the urban and remote environments."*

The last paragraph of Section 5.0 has changed from:

"*To investigate the drivers that might have led to these changes in [NO]/[HO2] ratio, we shall now examine how the NOx and HO_2 concentrations may have changed."*

to:

"This analysis suggests that the observed changes to the $[RONO_2]/[RH]$ ratio in the firn could be explained by changes to the average $[NO]/[HO_2]$ ratio experienced by air masses in transport to the Arctic. We now investigate whether trends in processes that could drive this ratio are consistent with this scenario, i.e. how NO_x and HO_2 concentrations may have changed."