

Interactive comment on “Changes to the chemical state of the northern hemisphere atmosphere during the second half of the twentieth century” by Mike J. Newland et al.

Anonymous Referee #2

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General comments. This manuscript is an ambitious work that derives alkyl nitrate trends from firn air measurements and combines them with similarly-derived (already published) parent alkane data to characterize possible scenarios for large changes in the chemical state (especially the $[\text{NO}]/[\text{HO}_2]$ ratio) of the northern hemisphere during the 1970s to late 1990s. The manuscript explores the possibility that the data could indicate significant changes in NO_x sources and sinks (driven by trends in sulfate aerosol precursors). Other potential explanations (such as changes in OH and transport) are also considered. The manuscript makes some very wide-ranging/large scale conjectures using this relatively limited new data. However, I consider that the authors are careful to back up their suggestions where possible and provide appropriate caveats

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where due. I recommend that the manuscript be accepted basically "as is", but I request that the authors consider making minor revisions as suggested below.

Specific comments. Given the potential importance of the trends in SO_2 emissions and their implied influence on the NO_x 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 NO_x 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.

Technical corrections typing errors, etc. Page 19, line 17 – I do not find the “Aydin et al.” reference in the reference section Page 23, Line 23 – I do not find the “Dlugokencky et al.” reference in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-927, 2016.

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