

*We greatly appreciate the editor's guidance and the reviewer's insightful comments. We have followed that guidance, and incorporated each of the suggestions into our revised manuscript. In the following we reproduce the original comments in black regular font, and include our responses in blue italic font. Revisions made to the manuscript are indicated in blue bold font.*

### **Editor's Comments:**

Please make the changes suggested by the reviewer. Given that this is a contentious topic (it always has been!), it is really important that the points raised by Cooper et al are clearly addressed in the manuscript. In my opinion your work marks a significant step forward: however it is important not to overstate the case or it will be seen as just another contribution to a long-running debate. So please pay close attention to the wording of the statements.

**We have made all of the changes suggested by the reviewer; these changes addressed several of the points raised by Cooper et al. Additionally, the following sentence has been added to the penultimate paragraph of the Results section:**

**“Notably, Tarasick et al. (2019) conclude that baseline ozone increased in the NH by a smaller amount (30-70%, with large uncertainty) between the period of historic and present day observations; however Parrish et al. (2021a) show that their comparison of historic data (collected at a set of primarily baseline representative sites at coastal and mountain locations) with modern data, collected at rural, low elevation sites within the European continental boundary layer, introduced several biases into their comparison, all of which caused systematic underestimates in their derived differences between the historic and modern data. Parrish et al. (2021a) discuss these biases in detail and give estimates of their magnitudes, which are large enough to account for this apparent disagreement.”**

*We think these changes address all of the major points raised by Cooper et al.*

*We have reviewed the wording throughout our manuscript, and are generally comfortable with the degree of certainty indicated, thanks in part to comments received in the first round of reviews. We have made three additional changes to ensure we do not overstate our case:*

- **Line 135: “... the most suitable method ...” replaced with “... a suitable method ...”**
- **Line 215: “We quantify the MBL baseline ozone mixing ratios as accurately as possible from these limited data, to allow a comparison of ...” replaced with “We quantify the MBL baseline ozone mixing ratios as accurately as these limited data allow, in order to compare ...”**
- **Line 289: The original wording: “The cause of the greater increases in the NH and reversal of the natural interhemispheric ozone gradient is attributed ...” was qualified by adding “likely” before “reversal”.**

### **Reviewer's Comments:**

I would like to thank the authors for their thorough responses to my comments. Taking into account those as well as their reply to the comments by Cooper et al., I feel that some minor amendments are still needed to provide further context to the conclusions drawn as well as making the differences with other studies (e.g., TOAR) clearer to the reader.

In particular, there are very interesting sections in the author response that, if included in the main text, would enrich the manuscript greatly, by giving further elucidation on the underlying reasoning and making it more accessible to readers.

*Thank you for your careful reading and useful suggestions.*

More specifically, I would like the discussion on ozone lifetime to appear in some form in the main paper (start of page 5 on the collated Authors' Response, "Simple transport and ozone lifetime considerations support this conclusion; in the free troposphere at northern mid-latitudes the net lifetime of ozone is estimated as 100 days, which is considerably longer than either the circum-global transport time (~30 days) or the vertical overturning time scale (~ 20 days). Consequently, even though the many sources and sinks of ozone are heterogeneously distributed, and each may possibly change over long time scales, the relatively rapid mixing and transport ensures that those changes are reflected in average baseline ozone concentration changes throughout northern midlatitudes. In the presence of relatively rapid transport and mixing, there simply is no mechanism that can maintain heterogeneity in the long-term changes in the zonal baseline ozone concentrations. Parrish et al. (2020; 2021b) discuss these considerations in more detail"). This would lend confidence to the approach taken by the authors in analysing the relative trends in ozone (so it would fit nicely in the Methods section).

**We included this material as a separate paragraph in the Methods section:**

**"The HTAP-based analysis approach utilized here relies on the concept that baseline ozone concentrations followed the same relative long-term changes throughout northern mid-latitudes. Simple transport and ozone lifetime considerations support this picture; in the free troposphere at northern mid-latitudes the net lifetime of ozone is estimated as 100 days, which is considerably longer than either the circum-global transport time (~30 days) or the vertical overturning time scale (~ 20 days). Consequently, even though the many sources and sinks of ozone are heterogeneously distributed, and each can possibly change differently over long time scales, the relatively rapid mixing and transport ensure that those changes are all reflected in approximately constant average baseline ozone concentration changes throughout northern midlatitudes. In the presence of relatively rapid transport and mixing, there simply is no mechanism that can maintain heterogeneity in the long-term changes in the zonal baseline ozone concentrations. Parrish et al. (2020; 2021b) discuss these considerations in greater detail."**

I found the discussion on the differences between HTAP and TOAR in the response to the comments by Cooper et al. and in Parrish et al. 2021 very interesting, and I feel some of it needs to be included in the text to provide further context, perhaps in the shape of a brief summary on the findings of Parrish et al (2021) regarding the choice of sites to include in the analysis. This could appear somewhere in the long paragraph between lines 143 and 163 of the revised manuscript with track changes.

**We revised the first 3 sentences of the suggested paragraph in the Methods section to give a brief summary regarding the choice of sites that were included in the HTAP analysis:**

**"The HTAP analysis followed the suggestion of Crutzen (1988): "it would be very interesting to compare certified old data with modern data taken at the same sites as where the 'ancient' data were taken." The "ancient" data considered are the sparse record of early measurements made at baseline representative sites throughout Europe, which extend back to 1950, with two summer measurement periods from the 1930s. Tarasick et al. (2019) "certified" the "ancient data" to the extent possible by carefully evaluating the early measurements; these results were compared to modern data collected at those same sites."**

Lastly, I would recommend amending the title slightly, as in the absence of measurements from the pre-industrial era the anthropogenic reversal of mid-latitude ozone remains an intriguing hypothesis. Perhaps something along the lines of "Investigations on the anthropogenic reversal etc."

**We have changed the title as suggested:**

**“Investigations on the Anthropogenic Reversal of the Natural Ozone Gradient between Northern and Southern Mid-latitudes”**