

## Reply to RC1

*We are grateful for the referee's supportive comments. 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.*

### General comment

In my view, this is an excellent paper that combines observations and model outputs in a manner that illustrates the complex phenomena driving ozone changes since pre-industrial times, and highlights what should become a standard test for current Earth System Models aimed at estimating anthropogenic climate forcing. After minor revisions, I warmly recommend publication in ACP.

*Thank you for your supportive comments*

### Specific comments

- In the text, there are several comparisons among trends derived from different observational and model output sets. Please specify the approach for calculating these trends and their corresponding error.

*Thank you for this suggestion. We have added the following text to the first paragraph of the Methods section:*

**Herein we discuss linear and polynomial fits to time series of ozone measurements; these fits with associated confidence limits are derived from standard least-squares fitting procedures, such as discussed in Chapters 6 and 7 of Bevington and Robinson (2003). All fits were performed with the Wavemetrics IGOR Pro software package.**

- Develop further the properties of the model STOCHEM-CRI that make it particularly useful for analyzing low NO<sub>x</sub> chemistry.

*And, thank you for this suggestion. The following text has been added to our revised manuscript:*

**The Condensed Reactive Intermediates (CRI) mechanism is a condensed version of the highly detailed and explicit Master Chemical Mechanism (MCM) v3.3.1 (<https://mcm.york.ac.uk>). Both rely entirely on evaluated laboratory chemical kinetics studies for their rate coefficient and product yield data, which is important in the present context of the pre-industrial atmosphere because low NO<sub>x</sub> conditions are not accessible in the smog chamber studies that have been an important source of mechanistic data for the chemistry of polluted atmospheres. Derwent et al. (2021) document the fidelity of the MCM and CRI mechanisms in a chemical mechanism intercomparison focused on the low NO<sub>x</sub> conditions of the pre-industrial troposphere.**

- Based on available long-term records in the Southern Hemisphere, you argue that the zonal variability of annual ozone means in the marine boundary layer is relatively small. However, Cape Grim, Ushuaia and Cape Point are subject to substantially different weather and (natural and anthropogenic) chemical regimes. Is this homogeneity also captured in models? What happens when you consider the seasonality of those data? Perhaps the similarity among ozone levels is just the result of compensating but quite different drivers.

*We agree that the three Southern Hemisphere sites are subject to substantially different weather and chemical regimes. However, at mid-latitudes in the free troposphere the lifetime of ozone (~3 months) is longer than either the mean circum-global transport time (~1 month in the prevailing westerly winds) or the average vertical overturning time (also ~1 month). This zonal and vertical transport implies that mean baseline ozone concentrations are relatively long-term and large spatial averages over the different ozone source and sink regimes throughout southern mid-latitudes. As a result, mean free tropospheric ozone concentrations and their seasonal changes are expected to be zonally similar. Entrainment of ozone from the free troposphere is the primary ozone source to the marine boundary layer (MBL) at the relatively remote baseline locations of these three sites. Consequently, the zonal similarity in the free troposphere is expected to be reflected in similar long-term changes and seasonal cycles at these MBL sites.*

Parrish et al. (2016) compare measured and model simulated ozone concentrations at these three sites (see their Figure 4); the expected zonal similarity is seen in the measurements, and to a greater or lesser extent in simulations from three chemistry climate models. Figure 1 compares the seasonal cycles measured at the three sites (taken from Figure 4 of Parrish et al., 2016), and Figure 2 compares those same measurement curves with similar curves derived from the seasonal cycles calculated by the 6 ESMs discussed in our manuscript. These ESM simulations generally reproduce the qualitative features of the measurements (i.e., wintertime maximum and summertime minimum), but also exhibit some quantitative disagreements.

Overall, the zonal variability of both the annual ozone mean and the seasonal cycle in the marine boundary layer is relatively small in the observations. **We now mention the similarity of seasonal cycles in the discussion of the small zonal variability of ozone in the marine boundary layer of the Southern Hemisphere.**

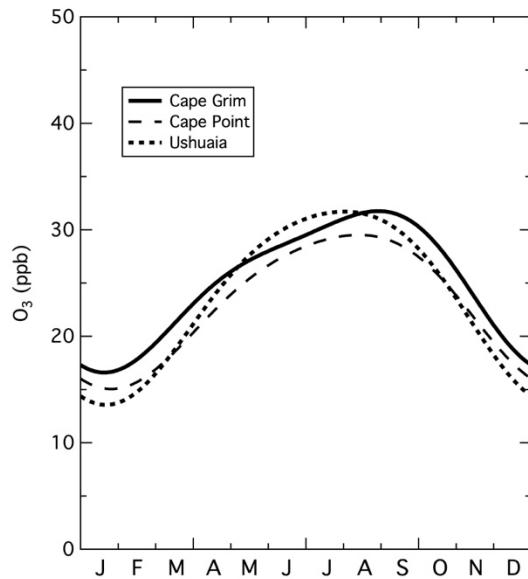


Figure 1. Sum of fundamental and second harmonic fits to measured seasonal cycles of  $O_3$  mixing ratios at the three southern hemisphere marine boundary layer sites identified in the annotation. Curves are taken from Figure 4 of Parrish et al. (2016).

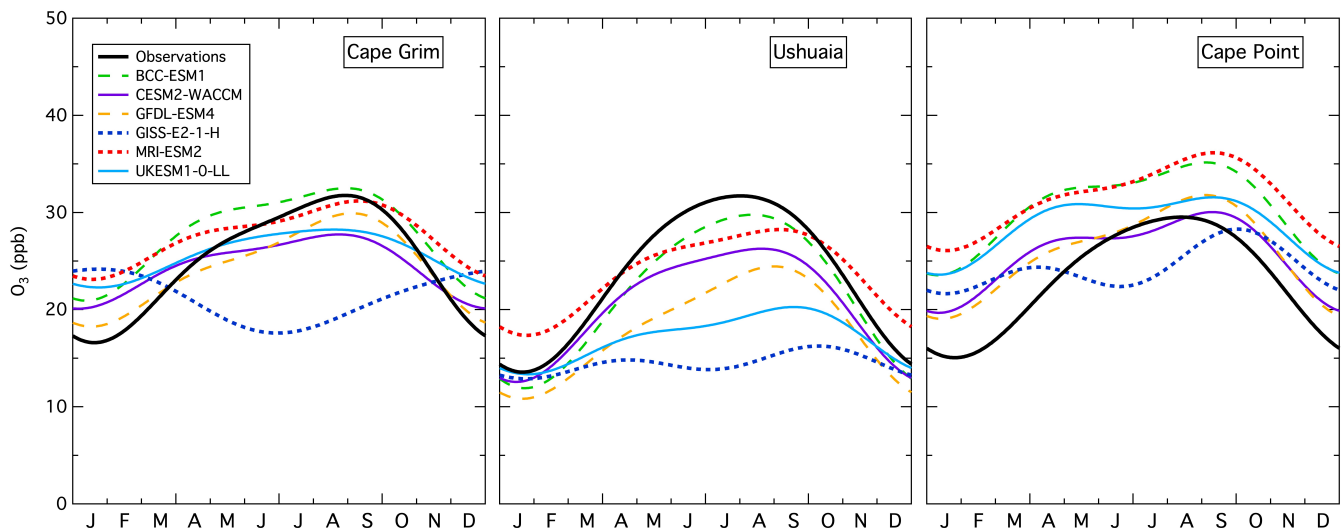


Figure 2. Sum of fundamental and second harmonic fits to measured and modeled seasonal cycles of  $O_3$  mixing ratios at the three southern hemisphere marine boundary layer sites included in Figure 1. Measurement curves are the same as in Figure 1. Model simulated seasonal cycle are from the 6 CMIP6 models discussed in our manuscript.