Reply by the authors to Referee 2's comments on "Seasonal, interannual and decal variability of Tropospheric Ozone in the North Atlantic: Comparison of UM-UKCA and remote sensing observations for 2005–2018" (acp-2022-99)

Anonymous Referee 2 (RC2)

We wish to thank the Referees for their time and constructive comments, which have helped us improve the quality of the original manuscript. Our responses to these comments are provided below (the Referee's comments are shown in italic).

General comment:

This paper evaluates changes in North Atlantic O_3 (2005-2018) using satellite observations and a chemistry-climate model, with a detailed analysis of the drivers of variability in the model and how this differs from observations. The abstract and introduction introduce the importance of the topic very clearly. The methods are well explained, the use of satellite data to derive O_3 column in vertical layers provides a very useful tool for model evaluation. There is thorough and detailed analysis throughout the study. The scope of the manuscript is certainly relevant to this journal.

Specific comments are listed below. I would recommend the manuscript for publication after these minor issues are addressed.

Specific comments:

Abstract: The abstract introduces the intent of the paper, methodology and major findings very well, but would benefit from including quantitative results. e.g. model/observation bias, trend in model O_3 vs observations, variability attributed to lightning NOx/STT.

We have now included more quantitative information as suggested and modified the abstract as follows:

"Tropospheric ozone is an important component of the Earth System as it can affect both climate and air quality. In this work we use observed tropospheric column ozone derived from the Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS) OMI-MLS, in addition to OMI ozone retrieved in discrete vertical layers, and compare it to tropospheric ozone from UM-UKCA simulations (which utilise the Unified Model, UM, coupled to UK Chemistry and Aerosol, UKCA). Our aim is to investigate recent changes (2005-2018) in tropospheric ozone in the North Atlantic region, and specifically its seasonal, interannual and decadal variability and to understand what factors are driving such changes. The model exhibits a large positive bias (greater than 5 DU or ~50%) in the Tropical upper troposphere: through sensitivity experiments, timeseries correlation and comparison with the LIS-OTD lightning flash dataset, the model positive bias in the Tropics is attributed to shortcomings in the convection and lightning parameterisations, which overestimate lightning flashes in the Tropics relative to mid-latitude. Use of OMI data, for which vertical averaging kernels and a priori information are available, suggests that the model negative bias (6-10 DU or ~20%) at mid latitudes, relative to OMI-MLS tropospheric column, could be the result of vertical sampling. Ozone in the North Atlantic peaks in spring and early summer, with generally good agreement between the modelled and observed seasonal cycle. Recent trends in tropospheric ozone were investigated: whilst both observational datasets indicates positive trends of ~5 and ~10% in North Atlantic ozone, the modelled ozone trends are much closer to zero and have large uncertainties. North Atlantic ozone IAV in the model was found to be correlated to the IAV of ozone transported to the North Atlantic from the stratosphere (R = 0.77) and emission of NOx from lightning in the Tropics (R = 0.72). The discrepancy between modelled and observed trends for 2005-2018 could be linked to the model underestimating lower stratospheric ozone trends and associated stratosphere to troposphere transport. Modelled tropospheric ozone IAV is driven by IAV of tropical emissions of NOx from lightning and IAV of ozone transport from the stratosphere; however, the modelled and observed IAV differ. To understand the IAV discrepancy we investigated how modelled ozone and its drivers respond to large scale modes of variability. Using OMI height-resolved data and model idealised tracers, we were able to identify stratospheric transport of ozone into the troposphere as the main driver of the dynamical response of North Atlantic ozone to the Arctic Oscillation (AO) and the North Atlantic Oscillation (NAO). Finally, we found that the modelled ozone IAV is too strongly correlated to El Nino Southern Oscillation (ENSO) compared to observed ozone IAV. This is again linked to shortcomings in the lightning flashes parameterisation which underestimates/overestimates lightning flash production in the Tropics during positive/negative ENSO events."

L103: Briefly expand on why the North Atlantic region is particularly important as well as the citation for more detail.

We have now expanded lines 103-105 as follows:

"The North Atlantic is an interesting region where decadal changes in climate, spanning the atmosphere, ocean and cryosphere, interact to produce periods of faster warming and cooling, known as Atlantic multidecadal variability (AMV, Sutton et al., 2017). The AMV has been linked to a number of local and non-local impacts, ranging between rainfall anomalies, changes in the frequency of hurricanes and Greenland ice-sheet melt, to name just a few (Robson et al., 2018 and references therein). The leading mode of atmospheric variability in the North Atlantic climate system is the North Atlantic Oscillation (NAO), which drives interannual variability in tropospheric ozone, temperature and precipitation over Europe (Robson et al., 2018 and references therein). Understanding decadal changes in ozone and its drivers can help us predict future changes in North Atlantic ozone and how to mitigate its impact on, for example, exacerbating air quality problems."

L131-134: Would benefit from clarifying exactly what the authors consider a "recent" trend. A number of studies, in particular TOAR assessments, have shown statistically significant increasing O_3 trends in the NH and in sites around the North Atlantic since the late 20th century (Gaudel et al., 2018, Tarasick et al. 2018). The authors rightly mention the uncertainties introduced by spatial and temporal inconsistency of these measurements but there is a broad consensus in the literature here.

Similar concerns were raised by Reeree 1. We have therefore further expanded/clarified lines 124-135 as follows:

“Whilst there is consensus on the long term increase in global ozone burden, it is harder to pinpoint its magnitude due to the sparse nature and reliability of early ozone measurements. Using isotopic evidence from polar firn and ice and some model simulations, Yeung et al. (2019) estimated an ozone increase of less than 40% between 1850 and 2005. Tarasick et al. (2019) found surface ozone increases of 30-70% between historical (1877-1975) and present day (1975-2015) measurements at rural Northern Hemisphere stations; they also found that free tropospheric ozone has increased by ~50% between the same period for Northern Europe and the Eastern USA. CMIP6 model integrations are consistent with observations, with the multi-model ensemble mean producing an increase in tropospheric ozone burden of ~109 ± 25 Tg (~40%) between 1850–1859 and 2005–2014 (Szopa et al., 2021); this change in ozone has been attributed to an increase in anthropogenic ozone precursor emissions over the same time period (Szopa et al. 2021).

In most recent decades, between the mid 1990s and present day, we see a more marked ozone increase in tropical regions compared to mid-latitudes (Gulev et al., 2021). At northern mid-latitudes, surface and low altitude ozone trends are variable, with some positive and some negative trends, but more positive values are observed in tropical regions (Cooper et al., 2020; Gaudel et al., 2020), where changes are between 2-17% per decade (Gulev et al., 2021). Similarly, ozone in the tropical free troposphere has increased more compared to ozone in the mid-latitude free troposphere, with increases of 2-12% per decade and 2-7% per decade, respectively (Cooper et al., 2020; Gaudel et al., 2020; Gulev et al., 2021; Chang et al., 2022).

Ozone trends in the North Atlantic can be influenced by a variety of factors. Anthropogenic emissions of ozone precursors have been decreasing in North America and Europe since the 1990s as a result of air quality policies; this reduction is potentially contributing to lower tropospheric ozone trends at northern mid-latitude compared to equatorial regions, where anthropogenic emissions of ozone precursors have continued to increase (Archibald et al., 2020a). Due to the relatively long lifetime of free-tropospheric ozone, 20-30 days (Young et al., 2013; Monks et al., 2015), North Atlantic ozone concentrations can also be affected by hemispheric transport of ozone generated by emissions outside of the local region (e.g., Butler et al., 2018; Sorooshian et al. (2020)). Other potential factors contributing to North Atlantic ozone trends include changes in tropical biogenic and biomass burning emissions, tropical NOx emissions from lightning and transport of ozone rich air from the stratosphere.

Several studies have focused on ozone trends in Europe, USA and the North Atlantic region using surface measurements, sondes, aircraft and satellite observations (Cooper et al., 2014; Parrish et al., 2014; Oetjen et al., 2016; Heue et al., 2016; Gaudel et al., 2020; Cohen et al., 2018; Cooper et al., 2020; Chang et al., 2022). However, due to ozone’s large interannual variability, calculated trends can be influenced by the reference years; furthermore, due to ozone spatial heterogeneity and large seasonal variations, reported trends can differ in magnitude depending on the horizontal/vertical location and season (e.g. Cohen et al., 2018).

L302-304: O3 burden compares well to the observed values, but given the large overestimate in tropical TCO, this must be the result of negative bias elsewhere in the model, and therefore not indicative of good model performance relevant to the current study. Supplementary Figure 4f also supports this.

We totally agree with the referee here, and in fact the next sentence expands on this point; lines 306-308 currently read:

"Archibald et al. (2020b) have shown that the UKCA global tropospheric ozone burden is consistent with observations as a result of an overestimate of TCO in the Tropics and an underestimate of TCO at mid latitudes, which is in line with our findings."

In order to make this clearer and avoid any possible misunderstandings, we have modified lines 300-308 as follows:

"Despite this spread in the observed TCO values, UKCA TCO, calculated for the same latitude band and period described in Gaudel (2018), shows values in the range 35-39DU, which are outside the range of uncertainty of the combined observations. Gaudel et al. (2018) reported a mean ozone burden, from 5 satellite datasets between 60° S:60° N, of ~300 Tg +/- 6 % for the most recent satellite record (up to 2016). In our study the tropospheric ozone burden from OMI-MLS and UKCA for the 2005-2018 period are 297 and 301 Tg respectively. Although UKCA's ozone burden in the 60° S:60° N range shows a very good agreement with observations, Archibald et al. (2020b) showed that the UKCA global tropospheric ozone burden is consistent with observations as a result of an overestimate of TCO in the Tropics and an underestimate of TCO at mid latitudes, which is in line with our findings (see also supplementary Figure 4)."

Section 3.1: NOx emissions from soil and biomass burning also contribute to O_3 variability.

We believe this Referee's comment is intended for Section 3.2 (not 3.1), as this is where the relationship between ozone and its precursor emissions is discussed. We agree with the Referee that NOx emissions from soil and biomass burning also contribute to O3 variability. In our model runs, these emissions are combined with anthropogenic NOx emissions and are referred in this section as 'surface NOx emissions'. We have now made this clearer and specifically acknowledged the importance of these extra sources by modifying lines 356-361 as follows:

"Present day anthropogenic emissions are generally well constrained, and their geographical locations, seasonal variations and magnitudes are derived from emission inventories and inverse modelling techniques (Lamarque et al., 2010; Feng et al., 2020). In contrast, some natural emissions of ozone precursors can have quite large uncertainties; these include CO and NOx from biomass burning, soil NOx, biogenic isoprene and NOx from lightning. An overestimate of such ozone precursors emissions in the model could therefore result in an overestimate of tropospheric ozone. Please note that, with the exception of lightning, all other natural and anthropogenic sources of NOx are combined in the model and referred to as surface NOx emissions."

Section 4.1: More context from a modelling perspective would be very informative here. How does the UM-UKCA compare to other relatable modelling studies? Is the lack of an O3 trend a consistent problem across CCMs (if so why?) or is it just UM-UKCA?

To address the Referee's comment we have now added the following paragraph to lines 505-506 in section 4.1:

“In contrast to observed trends, UKCA ozone trends tend to be much smaller in magnitude and effectively zero (within the error) for both the tropical and mid latitude part of the domain. Skeie et al. (2020) investigated ozone trends in CMIP6 model simulations; while it is clear that modelled ozone has increased significantly between 1850 and 2010, it is hard to pinpoint the sign and magnitude of the ozone trends from CMIP6 models in the more recent decades. Figure 2 in Skeie et al. shows that observed ozone trends for 2000-2010 are less than 5% per decade, while modelled trends for the same period show many models have trends very close to zero, and generally within ± 2% per decade. Although the period we are investigating (2005-2018) is not the same as shown in Skeie et al. (2020), their findings suggest that UKCA ozone trends in the more recent decades are comparable to other CMIP6 models and that accurately estimating tropospheric ozone trends over relatively short time periods remains a challenge due to ozone’s large interannual variability.”

Technical corrections:

L122: Jet stream?

“Jet speed” has been replaced with “speed of the jet stream”.

Figure 3: If the shaded area is of no interest in all 4 panels perhaps remove it from the figure?

We would prefer to keep the figure as it is for consistency, as all other figures show the full North Atlantic domain. Also, although not specifically within the upper troposphere, comparison of model and satellite over the shaded area provides some further information for the reader regarding model biases higher up in the atmosphere.

Figure 8. No label on y-axis.

We thank the Referee for pointing this out. The label on the y-axis should be “Ozone anomalies (DU)”. This has now been corrected.

Table 2. Unit. % change per year or over whole period?

We thank the referee for pointing out that the percentage had not properly defined. This is the percentage change per decade, relative to the concentration of each species at the beginning of the period in question. This has now been clarified in the caption to Table 2.

Figure 9. The black boxes next to the shaded area don’t clearly highlight the area of interest. Changing the colour of boxes/shading could improve this so it’s easier to pick out the important regions.

The colour of the black boxes has now being changed to magenta and lines were made thicker to address the Referee’s comment.

Supplementary Figure 8. No label on y-axis.

We thank the Referee for pointing this out. The label on the y-axis should be “Ozone anomalies (DU)”. This has now been corrected.