Author Responses to Reviewer Comments

We thank the reviewers for their useful and constructive comments/feedback. We also thank Owen Cooper for his useful comments on our manuscript in relation to the TOAR-II special edition. We have reproduced their comments below in black text, followed by our responses in red text. Please note, we have numbered the reviewers' comments (where not already done so) for clarification when providing comments. Any additions to the manuscript are in blue text and our reference to line numbers is based on the originally submitted manuscript. Also, as several of the reviewers' comments overlap, we have responded to one of the connected comments and then point the reviewers with similar comments to our initial response.

Reviewer #1 (William Collins):

1. The authors need to make it clearer what the major advances are over previous studies, such as Rap et al. 2015. Is it simply that they have now been able to calculate TO3RE for a longer timespan? Is there a significant advantage of IASI over TES as used by Rap et al.? Would the same timespan for TES give a similar result? Or is the bias correction using sondes the key improvement?

We consider all of these reasons to make this a worthwhile advance on the previous paper. The TES ozone record goes from 2004 to 2018 (i.e. 14-years), however, the actual data volumes were severely reduced after 2010 meaning a well-sampled data record of just 6-years. On the other hand, IASI, on MetOp-A, has maintained a radiometrically stable long-term record and uniform geographical sampling over the full mission (2008-2020, i.e. years with full coverage). TES did not scan across-track, so took only one nadir sounding of footprint 5.3 × 8.3 km swath at each point along the orbit, providing homogeneous though sparse coverage approximately every 16 days (Rap et al., 2015). The IASI swath in comparison is a 2200 km-wide providing homogeneous global coverage with its four 12 km diameter fields of view forming a 50 km by 50 km square scanned across track twice per day (Clerbaux et al., 2009). Therefore, the data density and data record for IASI are much larger than TES and thus provide a much more comprehensive ozone data set for analysis in this study. The application of the bias correction factors also adds an improved novelty as it harmonises the absolute value of TCO_3 for the three products used, meaning a more robust pool of data to derive the TO_3RE.

In relation to the bias correction factors, the importance of this is already discussed on Page 4 Lines 160-164: "*This is an important exercise as it provides a more accurate absolute range in satellite retrieved TCO_3 (and the ozone values used to derive the TO_3RE) but as the ozonesondes generally have poor spatial coverage, the global coverage and spatial distribution of the satellite data are critical in our analysis.*".

To make these points about IASI clearer, we have updated the following on Page 1 Lines 31-33 from:

"*Previous studies have shown the short-term (i.e. a few years) globally weighted average TO3RE to be 1.17±0.03 W/m^2, while our analysis suggests that the long-term (2008-2017) average TO3RE to be 1.21-1.28 W/m^2.*" to:

"Previous studies have shown the short-term (i.e. a few years) globally weighted average TO_3RE to be 1.17±0.03 W/m^2. However, from our analysis, using decadal (2008-2017) ozone profile datasets from the Infrared Atmospheric Sounding Interferometer, average TO_3RE ranges between 1.21 and 1.26 W/m^2.".

On Page 3, Lines 96-99, we update “Following the methodology adopted in Rap et al. (2015), we exploit satellite ozone profile data from IASI, on the MetOp-A satellite, which has a long-term record and substantial spatial coverage, in combination with the TOMCAT CTM, to improve the TO_3RE estimate and investigate its long-term variability and implications for climate.” to:

“Following the methodology adopted in Rap et al. (2015), we exploit satellite ozone profile data from IASI, on the MetOp-A satellite, which has a longer-term record and considerably denser spatial coverage than TES, in combination with the TOMCAT CTM, to improve the TO_3RE estimate and provide the first quantification of its decadal variability.”.

On Page 4 Line 130 we have added:

“Overall, IASI provides substantially denser spatial sampling and a longer-term record than its predecessor instruments. For instance, TES provided homogenous global coverage, albeit with sparse spatial sampling, every 16 days (Rap et al., 2013) over a 6-year period (2005-2010), while IASI on MetOp-A provided comparatively dense global coverage twice per day between 2008 and 2020 (though we focus on 2008-20217 where the IASI products have consistent records). Thus, making it suitable to investigate decadal average spatial patterns in TO_3RE and decadal scale interannual variability.”.

The two references listed above are:

Clerbaux, A., Boynard, A., Clarisse, L., et al. 2009. Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder. *Atmospheric Chemistry and Physics*, **9**, 6041–6054, doi: 10.5194/acp-9-6041-2009.

Rap, A., Richard, N.A.D., Forster, P.M., et al. 2015. Satellite constraint on the tropospheric ozone radiative effect. *Geophysical Research Letters*, **42**, 5074–5081, doi: 10.1002/2015GL064037.

2. Many of the statements made seem to conflate decadal trends with interannual variability. Since the lack of trend (0%/yr) is due to the compensation of increases due to emissions by decreases due to meteorology, it must be that emissions (0.2%/yr) and meteorology (-0.3% /yr) make comparable magnitude contributions to the decadal trend. However, it does seem clear (figure 3) that the year-to-year variability is dominated by meteorology.

This is true. Contributions from precursor emissions and meteorology are leading to a near cancellation and lack of any substantial TO_3RE trend. The meteorological interannual variability is most prominent in later years of the decade which coincides with the large-scale ENSO+ event of 2015/2016. Therefore, we have reorganised the analysis and conclusions to indicate that changes in meteorological factors and precursor emissions have balanced the TO_3RE tendency but large-scale variability from ENSO is driving a slightly larger trend (e.g. the fixed met run peaks in 2015 when ENSO+ occurred and the control and fixed emissions runs both have minimum values). The updates to the results can be seen in the updated Sections 3.1 and 3.2 at the bottom of this document. The conclusions and abstract have been updated accordingly.

3. Given that the main advance in this study seems to be the long time series available, more explanation is needed for the science reasons behind the trend and variability. For instance, some quantification of the emission trends over this period (i.e. the drivers behind fig 4(e)), and some explanation of the meteorological variability is needed. The text mentions “changes in global circulation”. Do the authors literally mean circulation as in

upwards/downwards transport of ozone? Or do they mean more generally changes in meteorological patterns including water vapour and temperature? The dip in the TC-Ems timeseries seems to coincide with the 2015-2016 El-Nino which presumably would affect tropical water vapour and humidity?

First, we note that TOMCAT is a CTM so there is no feedback from changes in ozone to radiation and thus meteorology in the simulations. However, changes in the precursor emissions and meteorology will influence the simulated ozone and thus the derived TO_3RE. The two TOMCAT sensitivity experiments were designed to provide an estimate of the top-level processes (i.e. emissions vs. meteorological) controlling the decadal interannual variability and trend in TO_3RE. The aim of the manuscript was not to dive down into individual processes controlling the decadal TO_3RE. While this would be scientifically valuable, to do so would require much more work beyond the scope of this study and a follow-up paper required. For instance, our recent study (Pope et al., 2023) investigating the processes behind the summer 2018 European ozone pollution events investigated that in detail. As can be seen from Figure 11 of that study, there was no clear relationship between temperature (i.e. one of the variables listed in Reviewer 1's comment #28) and ozone, primarily because factors controlling ozone are complex resulting in non-linear relationships with key variables. Several further sensitivity experiments would be required to untangle this, which is not our objective here. Therefore, while we agree that what the reviewer proposes would be very useful (in a follow-up study), we politely suggest this is beyond the scope of this current study.

Pope et al., (2023) reference:

Pope RJ, Kerridge BJ, Chipperfield MP, Siddans R, Latter, BG, Ventress LJ, Pimlott MA, Feng W, Comyn-Platt E., Hayman GD, Arnold SA and Graham AM.: Investigation of the summer 2018 European ozone air pollution episodes using novel satellite data and modelling. *Atmospheric Chemistry and Physics*, 23 (20), 13235—13253, doi: 10.5194/acp-23-13235-2023, 2023.

4. I guess the only influence of meteorology included here is on the ozone concentrations. Meteorology will also affect TO_3RE through the radiative transfer, particularly through cloud cover and through the surface and atmospheric temperatures. Presumably the Rap radiative kernels are based on a fixed climatology? This should be explained.

The reviewer is correct that the radiative kernel is based on a long-term average. The water vapour, temperature and surface albedo are based on a long-term climatology from ECMWF ERA-Interim reanalyses and uses cloud fields from 2000 from the International Satellite Cloud Climatology Project. We have added the following text on Page 5 Line 194:

"Meteorological inputs (temperature, water vapour, surface albedo) into SOCRATES to derive the radiative kernel are based on climatological ECMWF ERA-Interim reanalysis. Cloud fields are based on 2000 data from International Satellite Cloud Climatology Project data (Rossow and Schiffer, 1999), while aerosols have been ignored.".

We have added the Rossow and Schiffer (1999) reference to the reference list:

Rossow, W.B. and Schiffer, R.A.: Advances in understanding clouds from ISCCP, *Bulletin of the American Meteorological Society*, **80**(11), 2261–2287, doi:10.1175/1520-0477(1999)080<2261:aiucfi>2.0.co;2,1999.

5. As the authors discuss, the TO3RE is very sensitive to the vertical distribution of ozone. So it seems surprising that their headline numbers are derived from the (effectively) single tropospheric point from the satellite retrievals. This contrast with the Rap et al. paper who used the modelled profiles to derive the TO3RE and only used the satellite columns to constrain this. The authors should provide in the supplement examples of the ozone vertical profiles derived from the satellite retrievals and from TOMCAT before and after applying averaging kernels. I couldn't spot what the TO3RE was from the non-AK TOMCAT data (tables of values are needed). To me it would make most sense to combine the satellite (+sonde) derived kernel-averaged TCO_3 with the vertical distribution from TOMCAT to generate a blended ozone dataset.

Firstly, we have employed the same method used by Rap et al., (2015) with TES satellite ozone profile data to derive the TO_3RE (see bottom of Table 1 in their study). Therefore, we have confidence in this approach of using satellite data to derive the TO_3RE. Regarding the blended dataset, that is probably a paper in itself and beyond the scope of this study. Secondly, it is not possible to create a blended data set for all the satellite data, ozonesondes and TOMCAT because the AKs from each of the three individual satellite products would need to be applied to the model (yielding three different model-satellite data sets) for like-for-like comparisons. The different vertical sensitivities of the three satellite data sets also means it is not sensible to blend them. Regarding a table, we have added a new Table 1 summarising the statistics from Figures 1-3. This now includes the TCO_3, TO_3RE and NTO_3RE from the IASI products and the TOMCAT control run. The trend metrics are also included as well as the satellite degrees of freedom of signal for the troposphere and UTLS. Discussion of these metrics has been included in the updated Sections 3.1 and 3.2. We have added these new sections and figures/tables at the bottom of this response document. Regarding the vertical sensitivity, we have added the DOFS metrics in **Table 1** to show that there is substantial information in the IASI retrievals in the troposphere and UTLS (where the O_3 radiative effect is most effective).

6. The TOMCAT+AK values are not useful in their own right, since they throw away vertical information. It needs to be made clearer that these data are only useful for comparison with the satellites, and are not estimates of the "true" TO3RE. This is evident from the strong dependence of TO3RE on the AK. Some explanation needs to be given of why TOMCAT+IMS is so different. This doesn't seem related to the TCO3.

Please see response to Reviewer 1's comment #5. We consider the satellite TO_3RE data to be valuable (e.g. given the relatively high DOFS values) and note that they have similar values and variability to that of the model. However, we agree that Figure 2 and TOMCAT+AK time-series in Figure 3 need updating. See response in comment #5.

7. Numbers need to be presented in tables. It is difficult looking through all the figure panels to find numbers and compare them.

We have added a new table (**Table 1**) to include the TO_3RE numbers and other metrics:

Dataset	TCO_3 (DU)	TO_3RE (W/m^2)	NTO_3RE ($W/m^2/DU$)	TO_3RE Trend (%/yr)	TO_3RE CoV (%)	$DOFS_{trop}$	$DOFS_{utls}$
FORLI	32.6	1.23	37.8	-0.64 (-0.99, -0.28; p = 0.00) -0.21 (-0.66, 0.23; p = 0.35)*	2.5 (1.2)*	1.1	1.2
SOFRID	29.9	1.21	40.4	-0.01 (-0.14, 0.12; p = 0.94)	1.1	0.9	1.0
IMS	29.8	1.21	40.6	-0.13 (-0.36, 0.10; p = 0.25)	1.1	1.2	1.0
TC-CLT	30.7	1.26	41	-0.05 (-0.40, 0.30; p = 0.78)	1.5		
TC-EMS	30.6	1.25	40.8	-0.23 (-0.59, 0.13; p = 0.20)	1.7		
TC-MET	30.1	1.27	41	0.26 (0.13, 0.39; p = 0.00)	0.9		

Table 1: Summary statistics of the satellite and TOMCAT TCO_3, TO_3RE and NTO_3RE global average (2008-2017) metrics and the corresponding linear trends and covariance of variation (CoV) from **Figures 1-3**. TC-CTL, TC-EMS and TC-MET represent the control, fixed emissions and fixed meteorology runs, respectively. The global average (2008) degrees of freedom of signal (DOFS) for the IASI products are shown for the troposphere (approximately the surface to 200 hPa) and the upper troposphere – lower stratosphere (UTLS – approximately 400-100 hPa). * represents the IASI-FORLI trends for 2011-2017.

8. Lines 43-46: The mention of upper troposphere needs to be removed here since all these effects are happening at all altitudes. The longwave effect is most pronounced in the upper troposphere, but it occurs everywhere.

This is where the radiative effect is most pronounced, so we believe it is still appropriate to include this sentence. However, the reviewer is of course correct that it occurs everywhere. Therefore, on Page 2 Line 45, we have reworded the text as:

"As a SLCF, it influences both the incoming solar short-wave radiation and the outgoing long-wave radiation throughout the troposphere but has the largest radiative impact in the upper troposphere where the balance between the two yields a net positive (i.e. warming) effect at the surface.".

9. Line 49: "estimate these model TO3RE estimates". It is not clear what the authors mean here.

We have replaced, on Page 2 Line 9, "*However, satellite retrievals of tropospheric ozone in recent decades have provided the opportunity to estimate these model TO3RE estimates.*" with "However, satellite retrievals of tropospheric ozone now have decadal records and provide the opportunity to quantify the TO_3RE and complement estimates based on model simulations.".

10. Line 59: Suggest to cite AR6 (Forster et al. 2021).

This reference has now been added.

11. Line 64-65: This needs to be more specific as to what year is used for PI. The model studies used 1850 whereas the IPCC use 1750. This could be updated to IPCC AR6 (Forster et al. 2021, based on Skeie et al. 2020).

We have updated the text on Page 2 Lines 64-65 to "The PI to present day (PD) radiative forcing (RF) from TO_3 is estimated by the Intergovernmental Panel on Climate Change (IPCC) to be 0.47 Wm^{-2} (Forster et al., 2021) with an uncertainty range of 0.24-0.70 Wm^{-2}.".

12. Line 74: "TO3RE which is used to derive the TO3RF" is it not obvious how the TO3RE from this study can be used to derive the TO3RF.

We do not use the TO_3RE to derive the TO_3RF as we have not undertaken any PI model runs. However, satellite data can be used to derive the TO_3RE in the present and help constrain model estimates of the same quantity. Therefore, when used in conjunction with PI TO_3RE estimates, it can help estimate the TO_3RF. To make this clearer, we have altered the text on Page 2 Lines 71-75 to "Therefore, satellite ozone profile datasets from infrared instruments, in combination with off-line ozone radiative kernels (e.g. Bowman et al., 2013; Rap et al., 2015), can be used to quantify the PD TO_3RE. This can then either constrain model estimates of PD TO_3RE or be used directly with modelled PI TO_3RE to derive the TO3RF.".

13. Line 169: Does TOMCAT include stratospheric chemistry? If not how is ozone calculated above the tropopause?

On Page 5 Line 169 we have added:

", which extends up to the model lid. Here, climatological fields of trace gases/aerosols are used as the vertical boundary conditions (including stratospheric ozone).".

14. Line 175: Hoesy et al. 2018 would be a better reference for the emissions.

This reference has been updated accordingly.

15. Line 177: Be more specific about where the BVOC emissions come from. Presumably UKESM+JULES wasn't run specifically for this study. Was UKESM+JULES forced by the ERA-interim, or with AMIP SSTs, or free-running CMIP6 historical?

The BVOC emissions (not monoterpenes or isoprene) are climatological and come from CCMI, as mentioned in the text with the corresponding reference. The isoprene and monoterpene emissions are from a CMIP6 historical free-running simulation. Therefore, we have updated the text on Page 5 Line 178 "*within the free-running UK Earth System Model (UKESM, Sellar et al., 2019)*" to "within the free-running UK Earth System Model (UKESM Sellar et al., 2019) from a CMIP6 historical setup.".

16. Line 192-194: This sentence isn't quite correct as I don't think SOCRATES is used directly at all in this study. Rather the Rap kernels were used.

This is correct. We have updated the text:

"*The TO_3RE was calculated using the SOCRATES off-line radiative transfer model (Edwards and Slingo, 1996) in combination with TOMCAT and the three IASI ozone products.*" to:

"The TO_3RE was calculated using a radiative kernel, derived from the SOCRATES off-line radiative transfer model (Edwards and Slingo, 1996), in combination with TOMCAT and the three IASI ozone products.".

17. Line 225: "Appears to have a limited impact …". This is a strange phrasing for something that is purely a geometric effect. It has a limited impact because the area of a latitude band is zero at the pole. It is little or nothing to do with TO3.

Yes. We have deleted the text "*where TO_3 appears to have limited impact on the TO_3RE*".

18. Line 225: "supports this" – again a strange phrasing. If you multiply two numbers by cos(90) you get zero in each case whatever the original numbers were, so this doesn't support anything.

Yes. On Page 6 Lines 225-228, we have updated the text:

"*The bottom panel of **Figure 1** supports this as the zonally average profiles, weighted by the cosine of degrees latitude, show that TCO3 is near-zero in the high-latitudes, approximately 15.0-20.0 DU in the mid-latitudes, peaking at 28.0-33.0 DU in the sub-tropics and then decreasing by several DU at the tropics.*" to:

"The bottom panel of **Figure 1** shows the zonally average profiles weighted by the cosine of latitude (similar to Rap et al., 2015). This accounts for area weighting in the derived TO_3RE for different latitude bands on the global weighted average. Here, TCO_3 is near-zero at high-latitudes, approximately 15.0-20.0 DU at mid-latitudes, peaking at 28.0-33.0 DU in the sub-tropics and then decreasing by several DU in the tropics.".

19. Line 239-241: This doesn't seem to quite make sense. If NTO3RE in the S. Pacific is similar to other ozone regions then it is not true that "the South Pacific is more effective". It must be similarly effective.

The reviewer is correct here. We have therefore reworded the text on Page 6 Lines 234-241:

"*The NTO_3RE (**Figure 1 right column**) provides an estimate of where the TO_3RE is most sensitive to changes in TCO_3 (i.e. the unit of TO_3RE per unit of TCO_3). Peak NTO_3RE (>45.0 $mW/m^2/DU$) occurs in similar locations to the peak TO_3RE (e.g. sub-tropics, Africa and Australia), while the minimum values (10.0-20.0 $mW/m^2/DU$) occur in the high-latitudes. However, while the South Pacific TCO_3 values (23.0-30.0) are lower than other ocean regions (e.g. >30.0 DU), the NTO_3RE values are of similar magnitude (approximately 50.0 $mW/m^2/DU$). Therefore, while the sub-tropical/mid-latitude oceans have reasonably large TCO_3 and TO_3RE values, the South Pacific is more effective at contributing to the TO_3RE, despite its lower TCO_3 values (i.e. more positive radiative effect per unit of TO_3).*" to:

"The NTO_3RE (**Figure 1 right column**) provides an estimate of where the TO_3RE is most sensitive to changes in TCO_3 (i.e. the unit of TO_3RE per unit of TCO_3). Peak NTO_3RE (>50.0 $mW/m^2/DU$) occurs at similar locations to the peak TO_3RE (e.g. Africa and Australia), while the minimum values (10.0-20.0 $mW/m^2/DU$) occur at high-latitudes. Over the sub-tropical oceans, there are NTO_3RE values of similar magnitude (approximately 45.0 $mW/m^2/DU$). Therefore, despite some regions having lower TCO_3 and TO_3RE values (e.g. the South Pacific vs. the South Atlantic and Indian Ocean), the sensitivity to ozone perturbations (i.e. radiative effect per unit of TO_3) is similar in these regions. ".

20. Line 245-247: The different ozone profiles need to be shown here (or in the supplement) to explain this point.

We refer the reviewer to our response to his/her comment #5.

21. Line 247-253: This is a long sentence, but doesn't seem to be complete.

This text has been rewritten as:

"As the IASI-FORLI NTO_3RE is lower, while having the highest global average TCO_3 and TO_3RE, it suggests that IASI-FORLI has more TO_3 in the mid-troposphere where the radiative kernel has less sensitivity. Further to this, as the IASI ozone products only have approximately 1.0 DOFS, the harmonisation of the products using the ozonesondes can only be done on a tropospheric column level. As a result, the scaling of the satellite derived TO_3RE is done based on the relationship between the original IASI and IASI-sonde corrected TCO_3. Thus, a limitation is that though the upper troposphere is the most sensitive region to ozone radiative properties, the scaling of the TO_3RE is applied based on the satellite-ozonesonde TCO_3 relative differences.".

22. Line 272: Again, getting zero when multiplying by cos(90) isn't necessarily consistency.

We have replaced the text on Page 7 Lines 271-274:

"*In the bottom panel of* ***Figure 2****, the zonal profiles (weighted by cosine of degree latitude) for TCO_3 (TO_3RE) are consistent with IASI as high-latitude values are near-zero, mid-latitude values range between 10.0 and 20.0 DU (0.5 to 1.0 W/m^2) and sub-tropical values range between 30.0 and 38.0 DU (1.5 and 1.7 W/m^2).*" to:

"In the bottom panel of **Figure 2**, the zonal profiles (weighted by cosine of latitude to highlight the relative influence on the global weighted average) for TCO_3 (TO_3RE) have similar values to that of IASI. Here, the TOMCAT high-latitude values are near-zero (constrained by cos(90^o) = 0), mid-latitude values range between 10.0 and 20.0 DU (0.5 to 1.0 W/m^2) and sub-tropical values range between 30.0 and 38.0 DU (1.5 and 1.7 W/m^2).".

23. Line 280: But line 239 says the radiative efficiency of the south pacific is similar.

We have deleted lines 278-280 to account for this and refer the reviewer to our updated response to Reviewer 1's comment #19.

24. Page 8: Relevant numbers on this page need to be in a table.

We refer the reviewer to our response to his/her comment #7.

25. Line 318-319: This squashing of the interannual variability by applying the AKs seems worrying. This suggests that a considerable cause of the variability in TO3RE is due to changes in the vertical distribution which is washed out by the averaging.

We refer the reviewer to our response to his/her comment #5.

26. Line 328: Are all emissions (including biomass burning and BVOCs) fixed?

Yes, all the emissions are fixed. We have updated "*TOMCAT was run using repeating emissions and repeating meteorology for 2008*" to "TOMCAT was run using repeating emissions (from all sources) and repeating meteorology for 2008".

27. Line 334-335: The contributions of emissions (0.2%) and meteorology (-0.3%) seem to have comparable effects on the decadal trend. This is different from the year-to-year variability which does seem to be driven more by meteorology.

We refer the reviewer to our response to his/her comment #2.

28. Line 337: This discussion (and figure 4) needs to be clearer that it is comparing a single year to a decadal average that includes that year. For the meteorology it might have been more instructive to compare 2008 with 2015 (when the dip in TO3RE is strongest). It would be useful to show some meteorological variables (in the supplement) such as q, T, w to see what is changing. Similarly maps of emission changes should be shown in the supplement

too so we could see whether fig 4(e) is due to anthropogenic, biomass burning or BVOC changes.

We have updated the manuscript to make it clearer that we are comparing TOMCAT ozone using a single year (2008 repeating) to the long-term TOMCAT ozone average (2008-2017). This has been included in the updates to Sections 3.1 and 3.2 in line with our response to Reviewer 1's comments #5 and #6. As for Figure 4, we are interested in the decadal effect that meteorology and emissions have on controlling the TO_3RE. Therefore, we are not sure of the benefits of comparing 2008 with a low TO_3RE year (e.g. 2014 or 2015). This is because 2008 represents a relatively average year and not a high year. Comparing a low and high year would show the maximum difference the meteorological using fixed emissions (i.e. for both years). However, we are interested in decadal impacts and temporal change, as presented in the original **Figures 3** and **4**. Finally, the two TOMCAT sensitivity experiments were designed to provide an estimate of the top-level processes (i.e. emissions vs. meteorological) controlling the decadal trends in TO_3RE. The aim of the manuscript was not to dive down into individual processes controlling the decadal TO_3RE. While this would be scientifically valuable, to do so would require much more work beyond the scope of this study and a follow-up paper. For instance, our recent study (Pope et al., 2023) investigating the processes behind the summer 2018 European ozone pollution events investigated this in detail. As can be seen from Figure 11 of that study, it showed no clear relationship between temperature (i.e. one of the variables listed in Reviewer 1's comment #28) and ozone, primarily because ozone is a complex non-linear relationships with temperature and other variables. Several further sensitivity experiments would be required to untangle this, which is not the objective here. Therefore, while we agree that what the reviewer suggests would be very useful (e.g. a follow-up study), we politely suggest this is beyond the scope of this current study.

Pope et al., (2023) reference:

Pope RJ, Kerridge BJ, Chipperfield MP, Siddans R, Latter, BG, Ventress LJ, Pimlott MA, Feng W, Comyn-Platt E., Hayman GD, Arnold SA and Graham AM.: Investigation of the summer 2018 European ozone air pollution episodes using novel satellite data and modelling. *Atmospheric Chemistry and Physics*, 23 (20), 13235—13253, doi: 10.5194/acp-23-13235-2023, 2023.

29. Line 347: How do the authors know this is a circulation effect rather than due to water vapour and temperature changes?

The reviewer makes a fair point that we have not diagnosed the meteorological processes driving the changes. From Figure 4c, spatial changes have relatively steep gradients. Therefore, unlike more spatially homogeneous fields of variables like temperature (from the model and not in the radiative kernel), these differences appear to be more consistent with global circulation patterns (e.g. Rossby waves/weather systems). However, we have removed this statement "*though there is considerable spatial variation due to changes in the global circulation.*".

30. Line 372-373: This comparison with Rap et al. is presented without comment. Do these numbers supersede Rap (because they are better)? Or are they just representative of different years?

On Page 9 Line 373, we have added "This represents an update on the estimates from Rap et al., 2015) using an improved version of the TOMCAT model (as in Monks et al., (2017) compared to Richards et al., 2013)) and improved satellite products with better spatial and temporal coverage.

However, these two studies do cover different time periods, which may be contributing to the differences between the studies.".

31. Line 378: The emission changes have had a similar effect (0.2% vs -0.3%) to meteorology on the trend.

We refer the reviewer to our response to Reviewer 2's comment #2.

32. Line 381: This phrasing of "stabilising the tropospheric ozone contribution" seems to overstate the case. It is not obvious whether the compensation of the meteorology and emissions is a coincidence for this particular time period (that includes an El Nino near the end) or whether it is a more general compensation due to a climate trend.

We refer the reviewer to our response to Owen Cooper's comment #4.

Reviewer #2:

1. The paper is well written and logically presented and I have no significant concerns with the analysis or the discussion. My one significant concern is the ability of the IASI observations to constrain the vertical distribution of ozone in the troposphere, particularly given the importance of the vertical distribution for the radiative effect. This issue does come up a couple of times through the manuscript. There is some discussion of the limitations of the IASI observations of tropospheric O3 on lines 242 – 253 in reference to differences in the normalized TO3RE across the three different retrievals. There is also discussion of the differences in TO3RE when the IASI kernels are applied to the TOMCAT model, producing larger values of average TO3RE and, importantly, a marked reduction in the interannual variability as compared to directly using the original ozone distribution of TOMCAT over lines 313-326. The effects of the IASI kernel on the estimates of TO3RE are clearly shown in the figures and discussed where appropriate, but there is no dedicated discussion of the effect, which must surely be well known in the tropospheric ozone satellite observation community. I feel the manuscript would be improved if an overview of the limitations of the IASI observations was presented as part of the introductory material, perhaps as part of Section 2.4 or its own section before the results are presented.

The IASI instrument measures top-of-atmosphere infrared spectra to a high degree of radiometric accuracy and stability. The same vibration-rotation bands which determine ozone longwave absorption, and hence radiative effect, notably the strong ν_3 band centred near 9.6 μm, are used for IASI ozone height-resolved retrievals. Vertical sensitivities of IASI ozone retrievals, peaking as they do in the mid-upper troposphere, are therefore well matched to that of TO_3RE. Characteristics of the three IASI retrieval schemes are described in the respective references in the manuscript (i.e. Boynard et al., 2018; Barrett et al., 2020; Pope et al., 2021). Also, please see our response to Reviewer 1's comment #5 and #6.

Barret, B., Emili, E., Le Flochmoen, E. 2020. A tropopause-related climatological a priori profile for IASI-SOFRID ozone retrievals: improvements and validation. *Atmospheric Measurement Techniques*, **13**, 5237–5257, doi: 10.5194/amt-13-5237-2020.

Boynard, A., Hurtmans, D., Garane, K., et al. 2018. Validation of the IASI FORLI/EUMETSAT ozone products using satellite (GOME-2), ground-based (Brewer-Dobson, SAOZ, FTIR) and ozonesonde measurements. *Atmospheric Measurement Techniques*, **11 (9)**, doi: 10.5194/amt-11-5125-2018.

Pope, R.J., Kerridge, B.J., Siddans, R., Latter, B.G., Chipperfield, M.P., Arnold, S.R., Ventress, L.J., Pimlott, M.A., Graham, A.M., Knappett, D.S and Rigby R.: Large Enhancements in Southern Hemisphere Satellite-Observed Trace Gases Due to the 2019/2020 Australian Wildfires. *Journal of Geophysical Research: Atmospheres,* **126**(18), e2021JD034892, doi: 10.1029/2021JD034892, 2021.

The Pope et al., (2021) reference has also been added to the reference list as missed initially.

2. I would also suggest some caution in the presentation of the effect of meteorological variability on trends. As stated in the abstract, 'the meteorological variability in the tropical/sub-tropical upper troposphere is dampening any tendency in TO3RE from other factors (e.g. emissions, atmospheric chemistry).' I will note that large differences between the TOMCAT control simulation and the run with repeating meteorology as shown in Figure 3, are only really apparent in the 2014, 2015, 2016 period. At least 2015 – 2016 are years with an exceptionally strong El Nino and since this period falls towards the end of the 2008 – 2017 analysis period it has a significant effect on trends. It is not a significant objection, and the authors do state that the effects of meteorological variability have a strong effect on the trends for the period analysed, but I would urge the authors to be careful about leaving any impression of larger significance to the finding of meteorological variability. In particular, at lines 333 – 336 the authors state 'On the other hand, meteorological factors, while not dramatically altering the absolute simulated TO3RE values, are more important as fixing the meteorology yields a steady and significant increase (0.3%/year). Thus, without year-to-year variability in meteorology, temporal variability in TO3 would likely have a more substantial impact on the present-day climate.' If most of the differences in TO3RE are due to El Nino in 2015-2016 then large effect found for meteorological variability may be very particular to the exact period being analysed.

The reviewer makes a good point and we have updated the text in line with our response to Reviewer 1's comment #1 and #2.

3. Line 40: the authors state 'Here, the meteorological variability in the tropical/sub-tropical upper troposphere is dampening any tendency in TO3RE from other factors (e.g. emissions, atmospheric chemistry).' I am a bit unclear how to interpret this statement. Is it that meteorological variability is adding noise and making trends less statistically insignificant or is meteorological variability producing trends that are opposite to those imposed by emissions and atmospheric chemistry?

In line with Reviewer 1's comment #2, we have now updated the conclusions and underlying message on the fixed met and fixed emissions trends. Thus, the text referenced here by the reviewer has been updated in accordance with our response to Reviewer 1's comment #2.

4. Lines 58 – 65: a number of the references in this paragraph seem dated now. While they are not substantially different, why not refer to more current literature for quantities like the pre-industrial to present-day change in O3 radiative forcing, including the 6th IPCC Assessment Report?

We agree with the reviewer and direct him/her to our response to Owen Cooper's comment #7.

5. Line 73: unnecessary brackets around the year in 'Bowman et al., (2013)'.

The brackets are correct, but the Rap 2015 reference should be Rap et al., (2015).

6. Line 98: I think it should be TOMCAT in 'with the TOMCT CTM'

This has been corrected.

7. Lines 158 – 159: 'Here, we generated annual-latitude (30° bins) bias correction factors (BCF) which were applied to the gridded satellite records (see SI-2) to harmonise the retrieved TCO3'. Do the authors have any reason to believe the biases in the satellite data would be solely a function of latitude? I understand the sampling limitations of the ozonesonde data, which I assume led to the decision to derive broad latitude-dependent bias corrections, but could the authors present some information on the ozonesonde-IASI differences in TCO3 at individual stations to give the reader some idea of how regionally-dependent the biases are? Perhaps a plot of station locations, similar to Figure S1, but coloured according to the magnitude of the bias for the different IASI retrievals?

We appreciate the reviewer's point of view, and if there were more sites which were more evenly distributed, then we would focus on regional biases. Unfortunately, the spatial limitation of the ozonesondes means we struggle to do this. And if we use ozonesondes on a site-by-site basis, then it is going to be restricting the satellite correction to only regions with lots of data (e.g. Europe) and will potentially introduce sharp unrealistic spatial gradients in the satellite ozone fields. The approach used in this study has also been used in other studies (e.g. Russo et al., 2023), so we are confident in the method we have used here which is consistent with that paper. The corresponding reference is:

Russo MR, et al.: {Seasonal, interannual and decadal variability of tropospheric ozone in the North Atlantic: comparison of UM-UKCA and remote sensing observations for 2005-2018, *Atmospheric Chemistry and Physics*, 23 (11), 6169-6196, doi: 10.5194/acp-23-6169-2023

8. Lines 320 – 322: It seems that some parts of the sentence are missing: 'Interestingly, without the application of the AKs, the TOMCAT TO3RE time-series has similar temporal variability (e.g. peaks in 2008, 2010 and 2017 and troughs in 2009 and 2014.'

Sections 3.1 and 3.2 have now been re-written in response Reviewer 1's comments #5 and #6.

9. Lines 344 – 345: I couldn't help but notice that one pattern in the differences between the control run and the run with fixed meteorology (Figure 4c) looks like a wave-1 pattern across the tropics with strong positive differences centered over the Pacific and negatives values over the Atlantic and equatorial Africa. A bit of speculation, but I wonder if the pattern of the differences in the tropics is related to the effects of El Nino which would have been an important effect in the 2015 – 2016 period when the largest differences are seen between the control and constant meteorology simulations.

The reviewer makes a good point and this is addressed in our response to Reviewer 1's comment #2.

Comments from Owen Cooper:

1. The TOAR-II Recommendations for Statistical Analyses: The aim of this guidance note is to provide recommendations on best statistical practices and to ensure consistent communication of statistical analysis and associated uncertainty across TOAR publications. The scope includes approaches for reporting trends, a discussion of strengths and weaknesses of commonly used techniques, and calibrated language for the communication of uncertainty. Table 3 of the TOAR-II statistical guidelines provides calibrated language for

describing trends and uncertainty, similar to the approach of IPCC, which allows trends to be discussed without having to use the problematic expression, “statistically significant”.

We have now removed terms like statistically significant from the manuscript and have added the 95 % confidence on the TO_3RE trends and the corresponding p-values.

2. Several of the authors on this paper have another paper under review with the TOAR-II Community Special Issue which reports the long term (1996-2017) ozone trends across the globe based on a composite satellite product (Pope et al., 2023a). The lower-mid tropospheric column (surface to 450 hPa) ozone increases reported by this analysis are quite strong and in some latitude bands greatly exceed the observed increases reported by IPCC AR6 (Gulev et al., 2021). Below I have inserted a comparison between the Pope et al. 2023a ozone trends (ppbv decade-1) and the trends reported by IPCC AR6. In their reply to the referees the authors of Pope et al. (2023a) stated that they are confident in their reported positive trends and that they believe that the ozone decreases reported by IASI ozone products during the first phase of the Tropospheric Ozone Assessment Report (e.g. Gaudel et al., 2018) are not reliable. Based on studies that have appeared since the first phase of TOAR and based on the assessment by IPCC AR6, which have shown that tropospheric ozone has increased during the first part of the 21st century (Skeie et al., 2020; Szopa et al., 2021; Griffiths et al. 2021; Fiore et al., 2022; Wang et al., 2022; Liu et al., 2022), I agree that the tropospheric ozone burden has continued to increase. After the publication of Gaudel et al. (2018), Boynard et al. (2018) conducted a careful evaluation of the IASI ozone product and concluded that “The observed negative drifts of the IASI-A TROPO O3 product (8% – 16% decade-1) over the 2008–2017 period might be taken into consideration when deriving trends from this product and this time period.” It’s not clear if this new study has applied a bias correction to the IASI data to correct for the negative drift. If a bias correction has been applied, how does the corrected IASI record over 2008-2017 compare to the Pope et al. 2023a composite trend over the same period? If a bias correction has not been applied, then this study needs to discuss the impact of the negative drift on their analysis, and the authors also need to reconcile the IASI decreasing trend with the strong increasing trend reported by Pope et al. 2023a.

Firstly, the paper mentioned above, which has been accepted, focussed on the lower tropospheric column from UV-Vis sounders in the 1996-2017 period, whereas this paper focuses on the tropospheric radiative effect, with peak sensitivity in the upper troposphere and comparatively low sensitivity near the surface in the 2008-2017 time period. Also given the use of ozonesondes in both papers, we are confident the results should be consistent. As highlighted by Owen Cooper, a key issue concerns the IASI FORLI data used here. On Page 7 Lines 285 and 292, we discuss this. As advised by Anne Boynard (i.e. Boynard et al., (2018) and co-author on this manuscript), we have focused on the data trend from 2010 onwards after the recorded step change occurred when the product record is more stable and suitable for trend analysis. This has been presented in Section 2 and Figure 3 of the original manuscript.

3. There are many instances in the paper in which ozone changes over 2008-2017 are referred to as long-term. Typically, long-term ozone changes are thought of in terms of two or more decades. To draw a distinction between this study (2008-2017) and studies that examine

multi-decadal ozone trends, it would be helpful if the authors refer to their time period as decadal, rather than long-term.

We agree with this comment and have now replaced the use of "long-term" with "decadal".

4. Line 35 The abstract states that the tropospheric ozone radiative contribution to climate has remained stable with time, but these two processes are acting on very different time scales. Climate change is the response to radiative forcing, which can take decades to play out. While the analysis finds that ozone's radiative effect was constant over 10 years, there was no analysis to quantify the climate response to this period of stagnation, compared to a period in which ozone increased. While the authors can argue that radiative forcing did not increase, they provided no analysis of the climate response.

The reviewer is correct that we have not charactered the climate response from this stagnation in the radiative effect. We have not characterised the radiative forcing as suggested the comment above, however. As we are using a chemistry transport model instead of a chemistry climate model, we cannot undertake the analysis to quantity the climate impact (e.g. impacts on temperature, water vapour etc.). Therefore, we have modified our conclusions on the associated climate impacts. For example, we have replaced, on Page 10 Line 382, "*has been important in stabilising the tropospheric ozone contribution to climate, via radiative properties, in the recent past (i.e. satellite-era).*" With "has been important in stabilising tropospheric ozone and its short-term radiative impacts, in the recent past (i.e. satellite-era).".

5. Line 37 I don't think it can be stated that emissions had a limited impact on TO3RE. Presumably the model shows that ozone increased due to emissions increases over the period 2008-2017. Therefore, in the absence of meteorological variability, emissions would have caused an increase of TO3RE. I think this statement needs to be rephrased to say that meteorological changes masked the expected increase in TO3RE due to emissions increases.

The reviewer makes a good point and this is addressed in our response to Reviewer 1's comment #2.

6. The paper would benefit from a discussion of the reasons why changing meteorology reduced TO3RE. Is it due to changes in clouds, or to the redistribution of ozone in the atmosphere? For example, as TO3RE peaks in the upper troposphere in the sub-tropics, it seems reasonable that if ozone in the UT of the sub-tropics was pushed poleward, and/or downward, then a reduction of TO3RE would follow.

Please see our response to Reviewer 1's comment #3.

7. Lines 58-61 When reviewing the impacts of ozone on health, vegetation and climate you could also cite the key TOAR papers (Fleming and Doherty et al., 2018; Mills et al., 2018, Gaudel et al., 2018). References to IPCC should be updated to AR6, e.g. Forster et al., 2021; Gulev et al., 2021; Szopa et al., 2021.

We have updated the IPCC reference to those of Forster et al., (2021), Gulev et al., (2021) and Szopa et al., (2021). We have added the Mills et al., (2018) reference for the ozone-vegetation link. We have also added Fleming et al., (2018) in reference to the health effects from ozone. We have updated the original text on Page 2 Lines 58-65 from:

"Tropospheric ozone (TO_3) is a short-lived climate forcer (SLCF). It is the third most important greenhouse gas (GHG; Myhre et al., 2013) and a hazardous air pollutant with adverse impacts on human health (WHO, 2018) and the biosphere (e.g. agricultural and natural vegetation; Sitch et al., 2007). Since the pre-industrial (PI) period, anthropogenic activities have increased the atmospheric loading of ozone (O_3) precursor gases, most notably nitrogen oxides (NO_x) and methane (CH_4), resulting in an increase in TO_3 of 25-50% since 1900 (Gauss et al., 2006; Lamarque et al., 2010Young et al., 2013)., the TO_3 The PI to present day (PD) radiative forcing (RF) from TO_3 is estimated to be 0.4 (0.2-0.6) Wm^{-2} (Myhre et al., 2013; Stevenson et al., 2013) based on model simulations." to:

"Tropospheric ozone (TO_3) is a short-lived climate forcer (SLCF; Forster et al., 2021; Szopa et al., 2021). It is the third most important greenhouse gas (GHG; Forster et al., 2021; Myhre et al., 2013) and a hazardous air pollutant with adverse impacts on human health (WHO, 2018; Fleming et al., 2018) and the biosphere (e.g. agricultural and natural vegetation; Mills et al., 2018; Sitch et al., 2007). Since the pre-industrial (PI) period, anthropogenic activities have increased the atmospheric loading of ozone (O_3) precursor gases, most notably nitrogen oxides (NO_x) and methane (CH_4), resulting in an increase in TO_3 of 25-50% since 1900 (Gauss et al., 2006; Lamarque et al., 2010; Szopa et al., 2021; Young et al., 2013). More recently, since the mid-twentieth century, northern hemispheric TO_3 has increased by 30-70%. The PI to present day (PD) radiative forcing (RF) from TO_3 is estimated by the Intergovernmental Panel on Climate Change (IPCC) to be 0.47 W m^{-2} (Forster et al., 2021) with an uncertainty range of 0.24–0.70 W m^{-2}.". The corresponding references are:

Fleming ZL, Doherty RM, von Schneidemesser E, Malley CS, Cooper OR, Pinto JP, Colette A, Xutt X, Simpson D, Schultz MG, Lefohn AS, Hamad S, Moolla R, Solberg S and Feng Z.: Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health. *Elem Sci Anth*, **6**(12), doi: 10.1525/elementa.273, 2018.

Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.- L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D., Watanabe, M., Wild, M., and Zhang, H.: The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity, in: Climate Change 2021: The Physical Science Basis, Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J. B. R.,Maycock, T. K., Waterfield, T., Yelekçi, O., Yu, R., and Zhou, B., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 923– 1054, doi:10.1017/9781009157896.009, 2021.

Gulev, S.K., P.W. Thorne, J. Ahn, F.J. Dentener, C.M. Domingues, S. Gerland, D. Gong, D.S. Kaufman, H.C. Nnamchi, J. Quaas, J.A. Rivera, S. Sathyendranath, S.L. Smith, B. Trewin, K. von Schuckmann, and R.S. Vose.: Changing State of the Climate System. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, edited by Masson-Delmotte, V., P. Zhai, A. Pirani, S.L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M.I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J.B.R. Matthews, T.K. Maycock, T. Waterfield, O. Yelekçi, R. Yu, and B. Zhou., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 287–422, doi:10.1017/9781009157896.004, 2021.

Mills G, Pleijelt H, Malley CS, Sinha B., Cooper OR, Schultz MG, Neufeld HS, Simpson D, Sharps K, Feng Z, Gerosa G, Harmens H, Kobayashi K, Saxena P, Paoletti E, Sinha V and Xu X.: Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. *Elem Sci Anth*, 6(47), doi: 10.1525/elementa.302, 2018.

Szopa, S., V. Naik, B. Adhikary, P. Artaxo, T. Berntsen, W.D. Collins, S. Fuzzi, L. Gallardo, A. Kiendler-Scharr, Z. Klimont, H. Liao, N. Unger, and P. Zanis.: Short-Lived Climate Forcers. In Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, edited by Masson-Delmotte, V., P. Zhai, A. Pirani, S.L. Connors, C. Péan, S. Berger, N. Caud, Y. Chen, L. Goldfarb, M.I. Gomis, M. Huang, K. Leitzell, E. Lonnoy, J.B.R. Matthews, T.K. Maycock, T. Waterfield, O. Yelekçi, R. Yu, and B. Zhou., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 817–922, doi:10.1017/9781009157896.008, 2021.

8. Line 212 Bethan et al. (1996) is a fine paper, but when referencing the WMO tropopause the original WMO document should be cited.

This has been corrected and the following WMO reference added and in text reference updated.

WMO, Meteorology—A three-dimensional science, World Meteorological Organisation, Bulletin 6, (Oct), 134–138, 1957.

Updated text for Sections 3.1 and 3.2:

3. Results

3.1. Tropospheric Ozone Radiative Effect

Figure 1 shows the IASI derived TCO_3, TO_3RE and normalised TO_3RE (NTO_3RE, i.e. the TO_3RE divided by its TCO_3 as in Rap et al., (2015)). For the TCO_3, the three harmonised IASI products have good spatial agreement in the decadal (2008-2017) average, with a background north-south hemisphere gradient of approximately 30.0-40.0 to 15.0-25.0 DU. Peak TCO_3 (>40.0 DU) occurs over East Asia, the Middle East and in ozone outflow from central Africa (e.g. production from lightning and biomass burning precursor gases (Moxim & Levy, 2000)). The global average TCO_3 values for IASI-FORLI, IASI-SOFRID and IASI-IMS are 32.6 DU, 29.9 DU and 29.9 DU, respectively (**Figure 1 left column** and **Table 1**). From **Table 1**, degrees of freedom of signal (DOFS) are approximately 1.0 for the troposphere (i.e. $DOFS_{trop}$) and also in the upper troposphere – lower stratosphere (UTLS, $DOFS_{utls}$) – i.e. the vertical region where the O_3 radiative effect is most prominent). These DOFS are derived on a global scale using IASI data for 2008. They show there to be sufficient information in the troposphere from IASI to derive radiative effect metrics. Therefore, like in Rap et al., (2015), we are confident in our approach to directly use the satellite data to derive the observational TO_3RE.

When the TO_3RE is calculated (**Figure 1 middle column**), peak values occur over the sub-tropics, Africa and Australia ranging consistently between approximately 2.0 and 2.5 W/m^2 for each IASI product. The minimum values are found at high latitudes ranging between 0.0 and 0.8 W/m^2. The bottom panel of **Figure 1** shows the zonally average profiles weighted by the cosine of latitude (similar to Rap et al., 2015). This accounts for area weighting in the derived TO_3RE for different latitude bands on the global weighted average. Here, TCO_3 is near-zero at high-latitudes, approximately 15.0-20.0 DU at mid-latitudes, peaking at 28.0-33.0 DU in the sub-tropics and then decreasing by several DU in the tropics. The corresponding TO_3RE profiles follow a similar pattern with near-zero values at high-latitudes, approximately 0.5-1.0 W/m^2 at mid-latitudes, peaking at 1.5 W/m^2 in the sub-tropics and then decreasing to 1.1-1.2 W/m^2 in the tropics. Therefore, the sub-tropics have the largest contribution to the global TO_3RE. The global weighted TO_3RE averages for IASI-FORLI, IASI-SOFRID and IASI-IMS are 1.23, 1.21 and 1.21 W/m^2, respectively (**Figure 1** and **Table 1**).

The NTO_3RE (**Figure 1 right column**) provides an estimate of where the TO_3RE is most sensitive to changes in TCO_3 (i.e. the unit of TO_3RE per unit of TCO_3). Peak NTO_3RE (>50.0 mW/m^2/DU) occurs at similar locations to the peak TO_3RE (e.g. Africa and Australia), while the minimum values (10.0-20.0 mW/m^2/DU) occur at high-latitudes. Over the sub-tropical oceans, there are NTO_3RE values of similar magnitude (approximately 45.0 mW/m^2/DU). Therefore, despite some regions having lower TCO_3 and TO_3RE values (e.g. the South Pacific vs. the South Atlantic and Indian Ocean), the sensitivity to ozone perturbations (i.e. radiative effect per unit of TO_3) is similar in these regions.

Overall, the global weighted average NTO_3RE is 37.78, 40.43 and 40.60 mW/m^2/DU for IASI-FORLI, IASI-SOFRID and IASI-IMS, respectively. It is likely that differences between the three ozone retrieval schemes could be causing the differences between globally averaged NTO_3RE values. As the IASI-FORLI NTO_3RE is lower, while having the highest global average TCO_3 and TO_3RE, it suggests that IASI-FORLI has a larger fraction of TO_3 is located in the mid-troposphere, where the radiative kernel has less sensitivity than the upper troposphere. Further to this, as the IASI ozone products only have approximately 1.0 DOFS in the troposphere (**Table 1**), the harmonisation of the products using the ozonesondes can best be done on a tropospheric column level. As a result, the scaling of the satellite derived TO_3RE is done based on the relationship between the original IASI and IASI-sonde corrected TCO_3. Thus, a limitation being that though the upper troposphere is the most sensitive region to ozone radiative properties, the scaling of the TO_3RE is applied based on the satellite-ozonesonde TCO_3 relative differences.

TOMCAT allows for a further quantification of the TO_3RE in the satellite-era and the ability to run sensitivity experiments to explore some important top-level processes. Evaluation of the model using the IASI products and ozonesondes (see **SI-2**, **Figure S3** & **S4**) shows the model generally captures the TCO_3 spatial pattern and absolute values. In the tropics (mid/high-latitudes), the model underestimates (overestimates) by approximately 10-20% on average. These biases are comparable with other modelling studies evaluating models against satellite TO_3 observations (e.g. Archibald et al., 2020; Monks et al., 2017; Nassar et al., 2009; Young et al., 2013), indicating TOMCAT to be suitable for this study.

The global mean TCO_3 from TOMCAT (2008-2017) (**Figure 2 – top panel**) is 30.7 DU and consistent with the IASI data sets in **Figure 1**. When translated into TO_3RE, described above, the peak values from TOMCAT range between 2.0 and >2.5 W/m^2 over Africa, Australia and the sub-tropics. The global area-weighted TO_3RE for TOMCAT is 1.26 W/m^2, thus slightly larger than for IASI (1.21-1.23 W/m^2). As TOMCAT has a positive TCO_3 bias with respect to the observations in the sub-tropics, where the TO_3RE influence is most pronounced, this probably explains the slightly larger model TO_3RE value. In the bottom panel of **Figure 2**, the zonal profiles (weighted by cosine of latitude to highlight the relative influence on the global weighted average) for TCO_3 (TO_3RE) have similar values to that of IASI. Here, the TOMCAT high-latitude values are near-zero (constrained by $\cos(90^o) = 0$), mid-latitude values range between 10.0 and 20.0 DU (0.5 to 1.0 W/m^2) and sub-tropical values range between 30.0 and 38.0 DU (1.5 and 1.7 W/m^2). There is a decrease to approximately 25.0 DU (1.0-1.3 W/m^2) in the tropics. In terms of the NTO_3RE, the TOMCAT global area-weighted average is 41.0 mW/m^2/DU, which is similar to IASI. The peak NTO_3RE values are over the oceans (50.0-60.0 mW/m^2/DU) and over Africa/Australia (>60.0 mW/m^2/DU).

3.2. Temporal Evolution of the Tropospheric Ozone Radiative Effect

As IASI has daily global coverage (Clerbaux et al., 2009), we are able to derive annual average 3D ozone fields between 2008 and 2017, thus providing the first assessment of interannual variability and decadal tendency in satellite derived TO_3RE. **Figure 3** shows the annual TO_3RE time series for all three IASI products. First thing to note is that the Eumetsat meteorological data used to retrieve ozone for the IASI-FORLI product is subject to discontinuities (Boynard et al., 2018; Wespes et al., 2018). As a result, we include decadal analysis of the IASI-FORLI data for the full time period (2008-2017) and then a sub-time period (2011-2017) given the large discontinuity in September 2010 reported by Boynard et al., (2018) and Wespes et al., (2018). Here, we can derive the TO_3RE to quantify the absolute values (e.g. are they generally similar year to year?) and how they compare between products over the two time periods. In the near future, a new consistent IASI-FORLI ozone climate data record will be available using a more stable set of level-2 Eumetsat meteorological data retrieved from MetOp IASI and microwave sounders.

For IASI-SOFRID and IASI-IMS, the annual TO_3RE values range between 1.19 and 1.24 W/m^2 across the 2008-2017 decade. IASI-FORLI has somewhat larger values at the start of the record (1.26-1.28 W/m^2) before tending to that of IASI-SOFRID/IASI-IMS from 2011 onwards. Correlations (squared) in the annual TO_3RE time-series between IASI-FORLI and IASI-SOFRID (IASI-IMS) are poor at R^2=0.148 (R^2=0.132). However, IASI-SOFRID and IASI-IMS have a much stronger agreement with R^2=0.591 sharing nearly 60% of the temporal variability. We also calculate the coefficient of variation (CoV, i.e., time series standard deviation divided by its mean) to assess the inter-annual variability. For IASI-SOFRID and IASI-IMS, this is 1.1%, but for IASI-FORLI it is 2.5%. Therefore, there is more year-to-year variability in the IASI-FORLI TO_3RE record. However, when focussing on IASI-FORLI data for 2011-2017, the CoV drops to 1.2% in-line with IASI-SOFRID and IASI-IMS. The correlation (squared) values are now $R^2_{FORLI-SOFRID}$=0.496 and $R^2_{FORLI-IMS}$=0.137, which shows improved agreement between IASI-FORLI and IASI-SOFRID, but slightly surprisingly not with IASI-IMS. This may potentially be due to the lower sampling sizes of the IASI-IMS data record. Using ordinary least squares fit regression, IASI-FORLI, IASI-SORFRID and IASI-IMS have global average weighted TO_3RE linear trends of -0.64 (-0.99, -0.28; 95% confidence interval) %/year, -0.01 (-0.14, 0.12) %/year and -0.13 (-0.36, 0.10) %/year (see **Table 1**). As the IASI-FORLI product has known discontinuities (hence the larger CoV), the near-zero IASI-SOFRID and IASI-IMS trends are more robust. This is supported by IASI-FORLI when only considering 2011-2017 with a linear trend of -0.21 (-0.66, 0.23) %/year. Therefore, this suggests negligible change in the contribution of TO_3 to the tropospheric radiative effect over the recent past (i.e. 2008-2017).

TOMCAT global average weighed TO_3RE ranges between 1.24 and 1.29 W/m^2 between 2008 and 2017. The CoV is 1.5% for TOMCAT and is comparable to the IASI products (i.e. IASI-FORLI for later years). The TOMCAT TO_3RE time-series also has similar temporal variability (e.g. peaks in 2008, 2010 and 2017 and troughs in 2009 and 2014 to that of the IASI products. The underlying TOMCAT TO_3RE decadal trend is -0.05 (-0.40, 0.30) %/year and consistent with the IASI products. So, between 2008 and 2017, there has been limited overall change in TO_3, despite reasonable interannual variability, and thus its decadal impact on the TO_3RE has been relatively minor.

To investigate the importance of emissions and meteorology on the decadal TO_3RE trends, TOMCAT was run twice for the full time-period, once using repeating emissions and once using repeating meteorology for 2008 (i.e. start of the time-series). Using fixed emissions reduced the TO_3 burden and the TO_3RE values dropped to 1.22 to 1.28 W/m^2 (i.e. minima in 2014 and 2015 more pronounced). However, the trend in TO_3RE (-0.23 (-0.59, 0.23) %/year) remained small indicating that temporal changes in emissions yield a relatively small influence on the decadal tendency in TO_3RE. By comparison with the fixed meteorology run, temporal changes in meteorological processes over the period 2008-17 were found not to dramatically alter the TO_3RE values either, but there is an increase to 1.26 to 1.30 W/m^2 when the model meteorology is fixed to 2008. The corresponding TO_3RE trend in the fixed meteorology run is 0.26 (0.13, 0.39) %/year leading to a steady increase in TO_3RE, though with a similar magnitude to that of the fixed emissions experiment. Therefore, temporal changes in pre-cursor emissions and meteorological processes appear to be balancing each other leading to the near-zero TOMCAT control run TO_3RE trend. However, the largest changes in TO_3RE between the control and fixed meteorology runs are towards the end of the decade, coinciding with the 2015/2016 El Niño event (i.e. TO_3 spatiotemporal variability has previously been linked to El Niño activity – e.g. Ziemke et al., (2015) and Rowlinson et al., (2019)). The largest difference between the TOMCAT control and fixed meteorology runs is 0.6 W/m^2 in 2015 Overall, the year-to-year variability in meteorology appears to be contracting any decadal TO_3RE trend arising from temporal changes in precursor emissions with the net result being no significant underlying change in TO_3RE over the 2008-2017 decade.

Figure 4 shows the horizontal and vertical impact of the two sensitivity experiments on TOMCAT O_3 radiative effect (note the different colour bar scales). Consistent with **Figures 1** and **2**, the TOMCAT control TO_3RE has peak values (>2.50 W/m^2) over northern Africa and throughout the sub-tropics (approximately 2.0 W/m^2, **Figure 4a**). Vertically, the TOMCAT peak ozone radiative effect (>0.25 W/m^2) is in the upper troposphere (**Figure 4b**) with the largest impact in the sub-tropics of both hemispheres (500-200 hPa). Similar values extend through mid-latitudes of both hemispheres but in a smaller pressure range (400-300 hPa).

In **Figure 4c**, TO_3RE is seen to be higher in the fixed meteorology run than the control by 0.1 to >0.2 W/m^2 throughout the tropics and sub-tropics, although there is considerable spatial variability, including an area in the sub-tropical Pacific where TO_3RE is lower in the fixed meteorology run by -0.15 W/m^2. In high and mid-latitudes, TO_3RE is lower than the control by between -0.1 and 0.0 W/m^2. In the upper troposphere (**Figure 4d**), the zonal averaged contribution to TO_3RE in the fixed meteorology run is consistently higher than the control, by up to 0.02 W/m^2 at approximately 200 hPa in the tropics and sub-tropics and persisting at approximately 0.01 W/m^2 down to 600 hPa in the same latitudinal range. Poleward of 50°N and 50°S TO_3RE is lower in the fixed meteorology run, peaking at -0.02 to -0.015 W/m^2 at 300 hPa and extending down to 500 hPa at -0.005 W/m^2.

With fixed emissions, TO_3RE is higher at northern mid- and high latitudes by up to 0.02 W/m^2, consistent with a decline in anthropogenic precursor emissions, whereas in the tropics/sub-tropics and southern mid-latitudes it is generally lower than in the control run by up to -0.02 W/m^2 (**Figure 4e**). However, over tropical Asia, Indonesia and Australia, TO_3RE is seen to be lower by a more substantial amount, -0.05 to -0.04 W/m^2. In regard to its height dependence, contributions to TO_3RE are seen in **Figure 4f** to be lower in the fixed emissions run by up to -0.005 W/m^2 in the tropics/sub-tropics between 600 and 200 hPa, and also in a tongue stretching to southern high latitudes at around 300hPa (**Figure 4f**). In the northern hemisphere, on the other hand, TO_3RE in the layer between 400 and 600 hPa is seen to be higher by up to 0.003 at latitudes from the pole to 50°N, and down to higher pressures at latitudes below 50°N.

In summary, the two model sensitivity experiments indicate that, except for southern high latitudes, precursor emissions and meteorology exerted counteracting influences of comparable magnitude on TO_3RE in the 2008-17 decade, and this is specifically so in the sub-tropical regions of the upper troposphere, where contributions to global average area weighted TO_3RE are largest. At southern high latitudes, precursor emissions and meteorology are seen to have both increased TO_3RE over this period, specifically through contributions in the uppermost troposphere, although area weighting minimized their combined impact in the global averaged TO_3RE.

New/Updated Figures and Tables:

Dataset	**TCO_3 (DU)**	**TO_3RE (W/m^2)**	**NTO_3RE ($W/m^2/DU$)**	**TO_3RE Trend (%/yr)**	**TO_3RE CoV (%)**	**$DOFS_{trop}$**	**$DOFS_{utls}$**
FORLI	32.6	1.23	37.8	-0.64 (-0.99, -0.28; p = 0.00) -0.21 (-0.66, 0.23; p = 0.35)*	2.5 (1.2)*	1.1	1.2
SOFRID	29.9	1.21	40.4	-0.01 (-0.14, 0.12; p = 0.94)	1.1	0.9	1.0
IMS	29.8	1.21	40.6	-0.13 (-0.36, 0.10; p = 0.25)	1.1	1.2	1.0
TC-CLT	30.7	1.26	41	-0.05 (-0.40, 0.30; p = 0.78)	1.5		
TC-EMS	30.6	1.25	40.8	-0.23 (-0.59, 0.13; p = 0.20)	1.7		
TC-MET	30.1	1.27	41	0.26 (0.13, 0.39; p = 0.00)	0.9		

Table 1: Summary statistics of the satellite and TOMCAT TCO_3, TO_3RE and NTO_3RE global average (2008-2017) metrics and the corresponding linear trends and covariance of variation (CoV) from **Figures 1-3**. TC-CTL, TC-EMS and TC-MET represent the control, fixed emissions and fixed meteorology runs, respectively. The global average (2008) degrees of freedom of signal (DOFS) for the IASI products are shown for the troposphere (approximately the surface to 200 hPa) and the upper troposphere – lower stratosphere (UTLS – approximately 400-100 hPa). * represents the IASI-FORLI trends for 2011-2017.

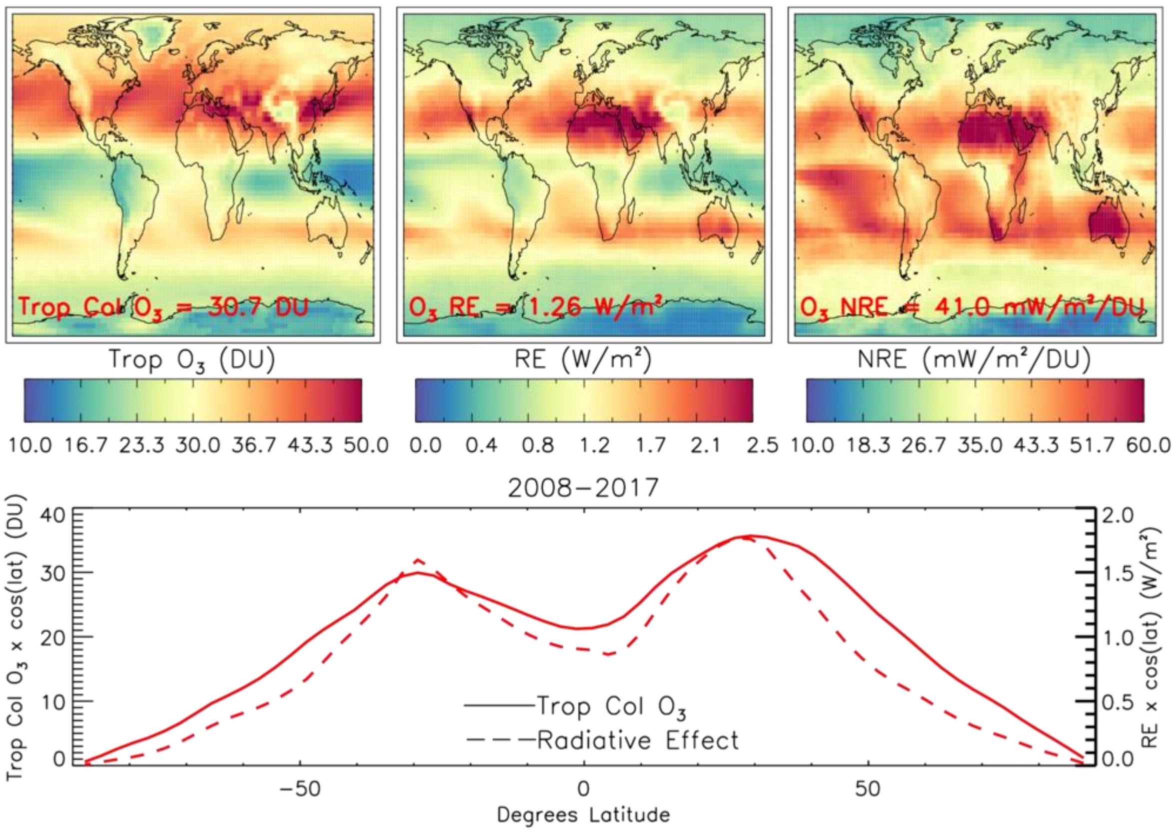

Figure 2: *TCO_3 (DU), TO_3RE (W/m^2) and NTO_3RE ($mW/m^2/DU$) averaged for 2008 to 2017 for TOMCAT. Zonal averages of TCO_3 (DU, solid lines) and TO_3RE (W/m^2, dashed lines), both weighted by cosine of latitude, is shown in the bottom panel from TOMCAT.*

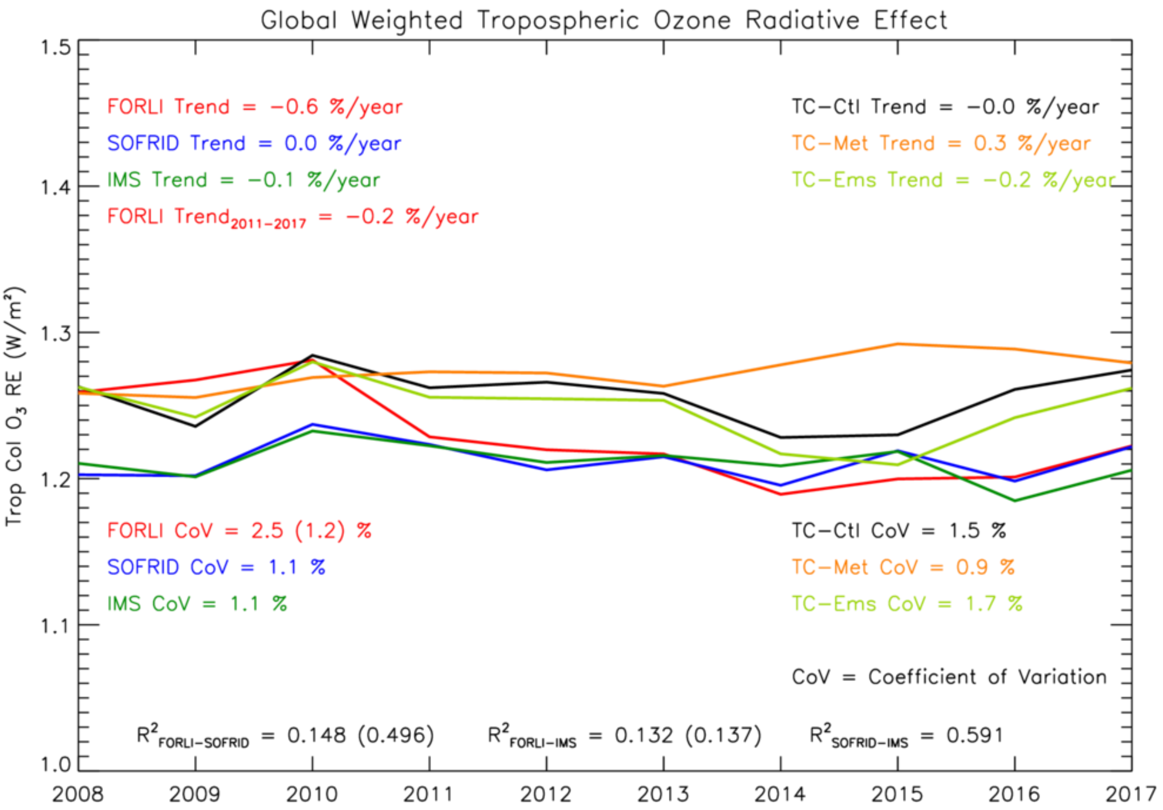

Figure 3: *Annual global mean time-series of TO_3RE (W/m^2), between 2008 and 2017, for IASI-FORLI (red-solid), IASI-SOFRID (blue-solid) and IASI-IMS (green-solid). TOMCAT simulation (black-solid), TOMCAT with fixed emissions (lime-solid) and TOMCAT with fixed meteorology (orange-solid) are also shown. The linear trend (%/year) is shown as well as the percentage coefficient of variation (CoV). The correlation between IASI time-series are shown by the R^2 values. TC represents TOMCAT. The IASI-FORLI trend for 2011 to 2017 is also shown as well as the CoV and R^2 in brackets in addition to the statistical metrics over the full time period due to record inhomogeneities prior to 2011 (Boynard et al., 2018).*

New References:

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Ziemke JR, Douglass AR, Oman LD, Strahan SE and Duncan BN.: Tropospheric ozone variability in the tropics from ENSO to MJO and shorter timescales. Atmospheric Chemistry and Physics, **15** (14), 8037-8049, doi: 10.5194/acp-15-8037-2015, 2015.