We thank Owen Cooper, Chris Smith and an anonymous reviewer for their useful comments that have helped improve the manuscript. To address these comments we have made substantial additions to the text, particularly to add discussion, figures and tables covering the radiative impacts of the ODS changes. The effective radiative forcing from the stratospheric ozone recovery is significantly larger than expected from previous studies and from calculations based on the traditional stratospheric-temperature adjusted RF. To further understand the physical processes behind the meteorological adjustments, we have added further discussion, figures and tables covering changes in humidity and surface albedo. We hope that these additions to the paper will make the importance of the scientific outcomes clearer to readers.

## Owen Cooper

### General comments:

The paper begins with an excellent review of ozone radiative forcing, which will be a very helpful reference for the scientific community (especially Figure 1).

## Thank you.

## Abstract.

Here the main findings on ozone RF are reported as: "We find robust changes in ozone due to future changes in ozone precursors and ODSs. These lead to a positive radiative forcing of  $0.27\pm0.09$  Wm-2 ERF,  $0.24\pm0.021$  W m-2 offline SARF,  $0.29\pm0.10$  Wm-2 online IRF." But to be clear, these numbers are the changes in RF between 2015 and 2050, correct? Should these results be reported as "delta ERF", for example? Would ERF for 2050 be estimated as 0.47 + -0.23 W m-2 (from IPCC AR6) plus  $0.27\pm0.09$  W m-2 (this study), to equal 0.74 + -0.31 W m-2?

# We have now clarified that the forcings are 2015 to 2050 $\,$

In the shared socioeconomic pathway SSP-3-7.0scenario, wWe find robust increases changes in ozone due to future increases changes in ozone precursors and decreases in ODSs<sub>2</sub>. These leading to a positive radiative forcing increase from 2015 to 2050 of  $0.2\underline{687} \pm 0.0\underline{849}$  Wm<sup>2--2</sup> ERF,  $0.24\underline{4} \pm 0.0\underline{5724}$  W m<sup>-2</sup> offline-SARF, and  $0.2889 \pm 0.101$  Wm<sup>-2</sup> online-IRF.

In this study we do not report these as changes since 1750, but use 2015 as the baseline. Care would be needed in combining our results with those from IPCC AR6, so we do not recommend this in this paper.

# Line 74

It would helpful to provide a little more background on the SSP3-7.0 scenario, which is generally described as being driven by "regional rivalry". While NOx and CH4 emissions go up in this scenario through 2050 (see figure 6.18 of IPCC AR6 WG-I), there are clear regional differences, especially for NOx (see figure 6.19 of IPCC AR6 WG-I). For example, NOx emissions decrease quite strongly by 2050 in North America, Europe, Russia, Central Asia, and Pacific OECD.

## We have added more detail here:

This scenario is chosen as it has the largest increase in tropospheric ozone (Keeble et al., 2021; Turnock et al., 2020) through increases in methane, NO<sub>x</sub> and other ozone precursors - although note NO<sub>x</sub> emissions decrease in OECD countries (Szopa et al., 2021).

The information in Figure 2 is a very handy reference, but I think it would work better as a table so you can list important items for each estimate such as: TOA vs tropopause, ERF vs IRF, starting and ending year. There seems to be an error in the mid-point of ozone RF for IPCC AR 2, which is plotted as 0.5 W m-2.

However, Page 20 of IPCC AR2 states: "Changes in tropospheric ozone have potentially important consequences for radiative forcing. The calculated global average radiative forcing due to the increased concentration since pre-industrial times is 0.4 (±0.2) Wm-2."

Thank you for your suggestion, we have added the information in figure 2 as a table in the supplement.

| Report | Nominal years | Tropospheric ozone                 | References                  |
|--------|---------------|------------------------------------|-----------------------------|
|        |               | forcing                            |                             |
| SAR    | 1850 to 1990  | $0.4 \pm 0.2 \text{ Wm}^{-2}$      | Hauglustaine et al. (1994)  |
| TAR    | 1850 to 1990  | $0.35 \pm 0.15 \ Wm^{-2}$          | Berntsen et al. (1997,      |
|        |               |                                    | 2000; Brasseur et al.,      |
|        |               |                                    | 1998; Hauglustaine et al.,  |
|        |               |                                    | 1998; Haywood et al.,       |
|        |               |                                    | 1998; Kiehl et al., 1999;   |
|        |               |                                    | Lelieveld & Dentener,       |
|        |               |                                    | 2000; Roelofs et al., 1997; |
|        |               |                                    | Stevenson et al., 1998;     |
|        |               |                                    | Van Dorland et al., 1997)   |
| AR4    | 1850 to 2000  | 0.35 [0.25, 0.65] Wm <sup>-2</sup> | (Gauss et al., 2006;        |
|        |               |                                    | Hauglustaine & Brasseur,    |
|        |               |                                    | 2001; Liao & Seinfeld,      |
|        |               |                                    | 2005; Mickley et al.,       |
|        |               |                                    | 2001, 2004; Shindell et     |
|        |               |                                    | al., 2003, 2005; Wong et    |
|        |               |                                    | al., 2004)                  |
| AR5    | 1750 to 2010  | $0.4 \pm 0.2 \ Wm^{-2}$            | (Skeie et al., 2011; Søvde  |
|        |               |                                    | et al., 2011; Stevenson et  |
|        |               |                                    | al., 2013)                  |
| AR6    | 1750 to 2019  | $0.45 \pm 0.225 \; Wm^{-2}$        | (Skeie et al., 2020)        |

Table S1: Tropospheric ozone radiative forcing (calculated as SARF) from the second (SAR) to sixth IPCC Assessment Reports.

## The value for SAR has been corrected – thank you for spotting this.

In terms of comparing the model output to observed ozone changes (1995 through 2021) a new paper by the HEGIFTOM Working Group will soon be available for open review on EGUsphere: Van Malderen, R., Z. Zang, K.-L. Chang, R. Bjorklund, O. R. Cooper, J. Liu, C. Vigouroux, E. Maillard Barras, I. Petropavlovskikh, T. Leblanc, V. Thouret, P. Wolff, P. Effertz, A. Gaudel, H.G.J. Smit, A. M. Thompson, R. M. Stauffer, D. E. Kollonige, D. Tarasick (2024), Global Ground-based Tropospheric Ozone Measurements: Regional

tropospheric ozone column trends from the HEGIFTOM homogenized ground-based profile ozone datasets, submitted to ACP (TOAR-II Community Special Issue) The analysis provides long-term ozone trends for many regions around the world, based on merged datasets (ozonesondes, lidar, IAGOS, FTIR). In my opinion, this is the best observation-based summary of global tropospheric ozone trends. The results are similar to the findings of IPCC AR6 (Gulev et al., 2021), except there is now evidence for a decrease of tropospheric column ozone in the Arctic, and there is a clear drop in ozone in 2020 and 2021 that coincides with the COVID-19 economic downturn.

Thank you for drawing our attention to this dataset which will be useful for the community. As we do not compare against observed changes in our study we will not cite this paper here.

Line 333 Please provide more information on the time-slice definition. Is this a 10-year time-slice centered on 2015 (i.e. averaged over 2010-2019)?

We have now clarified that this is a continuous year 2015

The control experiment (called pdClim-control) is a time-slice simulation for a continuousthe year 2015

#### **Chris Smith**

This paper provides results from a multi-model experiment of ozone radiative forcing (using three metrics: instantaneous, stratospheric adjusted and effective) for 2050 relative to 2015 under the SSP3-7.0 scenario. This addresses an important gap in the literature that was not available during the IPCC Sixth Assessment on the likely future evolution of ozone radiative forcing, and will be a valuable resource to the community that will be referenced for years to come.

## Thank you for your comment.

The title and abstract don't make it clear that only a single scenario is considered: we trust that the future will follow SSP3-7.0. Either call out SSP3-7.0 specifically or frame in more general terms: "Climate forcing due to future ozone changes in a high emissions scenario..." for example. Why was this scenario selected – presumably because it had the biggest signal? In the abstract, please state the 2050 relative to 2015 timeframe.

Since the focus of the paper is the metrics and methods rather than the specifics of the scenario we prefer not to lengthen the title. The key point of the paper is not the final radiative forcing number, but a comparison of the different methods to calculate radiative forcing.

We now explain in section 1 why this scenario is chosen.

This scenario is chosen as it has the largest increase in tropospheric ozone (Keeble et al., 2021; Turnock et al., 2020) through increases in methane,  $NO_x$  and other ozone precursors - although note  $NO_x$  emissions decrease in OECD countries (Szopa et al., 2021).

## We now clarify the timeframe:

<u>.</u>- These leading to a positive radiative forcing increase from 2015 to 2050 of  $0.2\underline{687} \pm 0.0\underline{849}$  Wm<sup>2--2</sup> ERF,  $0.24\underline{4} \pm 0.0\underline{5721}$  W m<sup>-2</sup> offline-SARF, and  $0.2\underline{889} \pm 0.10\underline{1}$  Wm<sup>-2</sup> online-IRF

Line 56: I think that tropospheric ERF is AR6 for 1750-2019 was +0.41 W m-2 and not +0.45 W m-2. Not that we mentioned it in the AR6 text, but I did do the trop/strat split. Does the Skeie et al. number include climate effects on ozone forcing? More generally, I am supposing that the experimental design here, using 2015 SSTs, does not include the climate effects on ozone either. SSP3-7.0 is a degree or more warmer in 2050 than in 2015 in many models so the effects are probably not insignificant.

AR6: "The contributions to total SARF in CMIP6 (Skeie et al., 2020) are  $0.39 \pm 0.07$  and  $0.02 \pm 0.07$  W m-2 for troposphere and stratosphere respectively ... The dataset is extended over the entire historical period following Skeie et al. (2020), with a SARF for 1750–1850 of 0.03 W m-2 and for 2010–2018 of 0.03 W m-2" – so this gives 0.45 W m-2 for the troposphere.

The Skeie et al. number does include climate effects. The experimental design here does not. This could be important for the comparison with CMIP6 ozone in section 4.2. We have added the caveat:

although the CMIP6 ensemble includes changes in climate which are excluded in the TOAR-RF simulations.

The section starting 1.1 on radiative forcing should possibly be promoted to a level-1 section (section 2). Possibly also section 1.2, but could sit under the radiative forcing header. I also thought that section 1.1 was a bit textbook and may not be required for this paper, but the coordinator of TOAR-II likes it, so it's probably a matter of taste and my familiarity with the topic.

Thank you for the suggestion. We have renumbered as suggested.

Line 270: "present day" in GISS means 2015? In all cases where "present day" is mentioned, please be specific on the year(s) (e.g. line 337).

"present day" has been changed to "2015" throughout

Line 337: where do these climatologies come from? Are they model-specific or centrally provided?

DMS concentrations climatologies are model-specific.

Other boundary conditions, such as ocean concentrations of <u>dimethyl sulphide (DMS)</u>, etc. are also prescribed <u>by each model</u> as climatologies appropriate for the present day

Lines 348-349: "the models' respective radiation and cloud schemes...": will changes in aerosols (that are not fixed) affect clouds, which will affect the ERF?

The aerosols only affect the chemistry schemes. Connections between aerosols and radiation and aerosols and cloud microphysics have been turned off. This has been clarified:

The models' respective radiation and cloud <u>microphysics</u> schemes continue to see year-2015 atmospheric concentrations <u>of greenhouse gases</u>, <u>aerosols and cloud condensation nuclei</u>, except for ozone.

Line 473:  $298.3 \pm 8.3$  DU – any observations to compare this to? Figure 6 has data from NIWA, perhaps this could be compared?

Lines 545-548: I'd also say the historical trend of CMIP6 models compared to the obs is quite good, even if biased high.

This comparison has been added to section 4.2.

The historical trend in CMIP6 ozone agrees well with that observed, Hhowever, bothlike the CMIP6 ensemble mean and, the TOAR-RF ensemble mean are systematically biased high relative to observations (283.5±1.1 DU) by approximately 10 DU.

Figure 8: the 150 ppb ozone tropopause forcings agree quite well between models. Is this expected and/or worth a comment?

A comment to this effect has been added:

There is close agreement between models in the tropospheric forcing, but more model variability when including stratospheric changes as indicated by the large standard deviation across models in the total ozone radiative forcing.

Very minor, editorial things

Line 51: reference after full stop

**Fixed** 

Line 79: write out equation on a new line

**Fixed** 

Line 116: comma after full stop

**Fixed** 

Line 310: Walters citation as author (year).

**Fixed** 

Line 545: Bodeker Scientific doesn't show up in the references.

Fixed

Line 576: superscript -2

Fixed

### Anonymous Reviewer 2

#### **General Comments**

The manuscript compares across different metrics and methods to estimate ozone radiative forcing by first presenting a synthesis of prior work and then calculating the radiative forcing from 2015 to 2050 using the current generation of Earth system models. The manuscript documents inconsistencies in approaches in prior work as well as unique configurations in some models that complicate a straightforward inter-model comparison. This detailed documentation is invaluable to the modeling community and is a critical piece for interpreting some simulations and possibly the next round of multi-model studies. It does, however, lead to a lengthy manuscript in which major conclusions may be missed. At the same time, providing some additional context to the abstract and conclusions may help a reader understand the importance of the work. Two general suggestions:

1. Articulate more clearly the key messages in the abstract/conclusions with some short synthesis statements that provide slightly broader context. For example, the authors may wish to consider the following questions: How much confidence is there in estimates of ozone forcing and the stratosphere versus troposphere contributions as reported here and in prior work? Given the emphasis on this prior work in the introduction, I expected the authors to conclude by comparing their results to that earlier work. Do the conclusions drawn here have implications for the interpretation of historical radiative forcing estimates for ozone? How important is ozone relative to other greenhouse gases? How do the emission trends driving the ozone forcing in this scenario compare with other future scenarios?

We have revised the conclusions to include the context that this calculation is larger than that assumed in the IPCC AR6 and would make ozone the second most important contributor to radiative forcing over the 2015-2050 period in this scenario. We also draw attention to the calculations that the ERF metric indicates much larger climate effects than SARF for stratospheric ozone recovery.

The abstract has been completely rewritten

## Abstract.

We use Earth system models and a chemistry transport model For the first time this study assesses to determine three different measures of radiative forcing (instantaneous: IRF, stratospheric-temperature adjusted: SARF, effective: ERF)the radiative forcing due to for future changes in ozone. These, use a combination of online and offline methods. We separate the effects of changes in ozone precursors and ozone depleting substances (ODS) changes, and—Three different measures of radiative forcing (instantaneous: IRF, stratospheric temperature adjusted: SARF, effective: ERF) are compared using both online and offline calculations for the IRF and SARF, and online calculations for the ERF. To isolate the ozone radiative forcing, we configure the model experiments such that only the ozone changes (including respective consequent changes in water vapour humidity, clouds, surface albedo-ete.) affect the evolution of the model physics and dynamics.

In the shared socioeconomic pathway SSP-3-7.0scenario, wWe find robust increaseschanges in ozone due to future increaseschanges in ozone precursors and decreases in ODSs<sub>2</sub>. These leading to a positive radiative forcing increase from 2015 to 2050 of 0.2687 ±0.0849 Wm<sup>2--2</sup> ERF, 0.244 ± 0.05721 W m<sup>-2</sup> offline-SARF, and 0.2889 ± 0.101 Wm<sup>-2</sup> online-IRF. This increase makes ozone the second largest contributor to future warming by 2050 in this scenario, approximately half of which is due to stratospheric ozone recovery and half due to tropospheric ozone precursors.

Increases in ozone are found to lead to an overall decrease in-cloud fraction, (although there are increases at some levels). This decrease causeings an overall negative adjustment to the radiative forcing (positive in the short-wave (SW), but negative in the long-wave (LW)). Non-cloud adjustments (excluding stratospheric temperature) due to water vapour and albedo changes are positive (both LW and SW). ERF is slightly larger than the offline The opposing signs of the cloud and non-cloud adjustments mean the overall adjustment to the SARF-is slightly positive for the total ozone change, but approximately double the SARF for the ODS-driven change (0.156 ± 0.071 Wm<sup>2-2</sup> ERF, 0.076 ± 0.025 W m<sup>-2</sup> SARF). Hence ERF may be a more appropriate metric for diagnosing the climate effects of stratospheric ozone changes.

We find general agreement between models in the impact of the ozone changes on temperature and cloud fractions and agreement in the signs of the individual adjustment terms when split into SW and LW. However, the overall difference between the ERF and SARF is smaller than the inter-model variability.

The conclusions are largely rewritten

### 6. Conclusions

We have shown that projected increases in tropospheric ozone precursors and decreases in ODSs in the SSP3-7.0 scenario lead to increases in ozone in the troposphere and stratosphere. By restricting the impact of composition changes on the evolution of the physical model, we can isolate the changes solely due to changes in ozone. This contributes an ERF of  $0.2\underline{687} \pm 0.0\underline{849}$  Wm<sup>-2</sup> by from 2015 to 2050. This is larger than the forcing of 0.19 Wm<sup>-2</sup> assessed by IPCC AR6 for this period and scenario (Dentener et al., 2021), and would make ozone the second largest contributor to warming over this period due to the combination of the forcing from stratospheric ozone recovery, and the increases in ozone precursor emissions in this scenario.

A subset of models calculated the ERF excluding ODS changes. These show that ozone precursor increases and ODS decreases contribute approximately equally to the total ERF change. The future contribution of ODSsfor these, the ERF decreased from 0.31 ± 0.08 Wm² in the standard perturbation to 0.15 ± 0.03 Wm² with fixed ODSs. Decreases in ozone between the standard and fODS perturbations were not confined to the stratosphere but also affected the upper troposphere, to ozone ERF is comparable (though of opposite sign) to the contribution of ODSs to the historical ozone forcing as assessed by IPCC AR6. This would make the indirect radiative efficiency of ODSs (in forcing per change in equivalent effective stratospheric chlorine) almost three times that assessed in the IPCC and WMO (2022). (Szopa et al., 2021) This highlights the increase in ozone forcing expected from commitments to reduce ODSs which could therefore offset mosts some of the climate benefits from reducing their direct greenhouse effect.

ERF and SARF diagnosed from the offline kernel agree within the uncertainty range for the combined effect of ozone precursor and ODS changes. However the difference between ERF and SARF becomes more apparent when looking at the ozone precursors and ODSs separately. The SARF with fixed ODSs is higher than the ERF, which could be explained by systematically higher kernel results (since this is not the case for the online-calculated SARF in the EMAC model). The SARF due to ODS changes is significantly lower than the ERF which suggests

the fixed dynamical heating approach used in the SARF overestimates the temperature increase in the stratosphere from stratospheric ozone recovery, and that there is an additional contribution to the ERF from increasing stratospheric water vapour. The radiative adjustment due to reduced surface albedo is significant and positive.

Ozone increases from both ozone precursors and decreasing ODSs consistently reduce cloud cover in the upper troposphere, with most models finding increased cloud cover in the mid-troposphere. This leads to significant radiative adjustments that are negative in the LW and positive in the SW. The net effects of clouds largely cancel giving a net adjustment that is not significantly different from zero (slightly negative from ozone precursor increases, slightly positive from ODS decreases).

Few of the models were able to calculate a SARF online, so we used offline kernel calculations to determine the SARF. We tested the kernel calculations by comparing the IRFs for kernel and online calculations. The kernel calculations give consistently higher IRFs (both LW and SW). The multi model mean ERF for the standard perturbation is 0.27±0.09 Wm<sup>-2</sup> compared to the kernel SARF of 0.24±0.02 W m<sup>-2</sup>, suggesting an overall positive (non stratospheric temperature) adjustment. This also true for the online ERF vs SARF in the EMAC model. The cloud adjustments are negative (0.02±0.04 Wm<sup>-2</sup>) which implies positive non cloud adjustments. This is supported by the finding of positive adjustments in both SW and LW comparing clear sky ERF and SARF. For the fODS perturbation the ERF is 0.15±0.03 Wm<sup>-2</sup>, with a kernel SARF of 0.17±0.01 Wm<sup>-2</sup>. The overall negative adjustment is due to the more negative cloud adjustment in two of the models (mean of -0.04±0.04 Wm<sup>-2</sup>) and a less positive non-cloud LW adjustment.

This study shows that care is needed when interpreting or comparing radiative forcing calculations for ozone. There are discrepancies between the online and offline calculations of the IRF, particularly in the LW. This might indicate differences in the SARF. While Ultimately the radiative forcing calculated as ERFs or SARF is similar for the combined effects of ozone precursors and ODSs, this is not true for the ozone precursors and ODSs separately. Here, we find that the kernel-calculated SARF is a factor of two lower than the ERF for -the ozone radiative forcing from ODS changes highlighting a need to compare offline radiative transfer modelling against a full Earth-system model. This occurs due to a cancellation between a negative cloud adjustment and a positive non-cloud adjustment.

Justify the use of SSP3-7.0. For example, is this the scenario that regional emissions have
followed most closely in the last decade or simply the one that all the models ran? What are NOx,
NMVOC, and CO emission trends as well as ODS and methane in this scenario? Consider
showing a plot with zonal mean 2050-2015 emission changes (and ODS) to highlight regional
differences.

We have now explained that the choice of SSP3-7.0 is a pragmatic choice designed to give the largest ozone signal. The key point of the paper is not the final radiative forcing number, but a comparison of the different methods and metrics to calculate radiative forcing. For this reason we do not discuss the trends in the different precursors or the regional differences.

This scenario is chosen as it has the largest increase in tropospheric ozone (Keeble et al., 2021; Turnock et al., 2020) through increases in methane, NO<sub>x</sub> and other ozone precursors - although note NO<sub>x</sub> emissions decrease in OECD countries (Szopa et al., 2021).

### **Specific Comments**

1. The abstract should include the time frame and scenario over which the forcings are calculated.

We have now clarified that the forcing is over the period 2015 to 2050 and for the SSP3-7.0 scenario.

In the shared socioeconomic pathway SSP-3-7.0scenario, wWe find robust increases changes in ozone due to future increases changes in ozone precursors and decreases in ODSs. These leading to a positive radiative forcing increase from 2015 to 2050 of 0.2687 ±0.0849 Wm<sup>2--2</sup> ERF, 0.244 ± 0.05721 W m<sup>-2</sup> offline SARF, and 0.2889 ± 0.101 Wm<sup>-2</sup> online IRF.

2. Please clarify what appears to be conflicting findings: Checa-Garcia et al. (2018) report that tropospheric ozone cools the lower stratosphere (line 117) but Figure 9g shows warming in the lower stratosphere in response to increasing tropospheric ozone (fODS).

Thank you for pointing this out. We now discuss in section 4.7 that the modelled temperature changes can differ from the FDH calculations, and have added a figure S9 to show this.

Figure S9a shows more detail on the stratospheric temperature profile change for one model (EMAC). Here it can be seen that the increased temperature occurs in the lower and middle stratosphere (roughly from 200 to 20 hPa), and also around 2 hPa. Temperature changes at higher altitudes have less impact on the net TOA radiative forcing as the density of the atmosphere decreases. In the *fODS-perturbation*, however, SARF is enhanced by 0.062 Wm<sup>-2</sup> compared to IRF due to the decreased stratospheric temperatures as diagnosed by the FDH calculations (see Fig. S9). Note in Fig. 9h (and Fig S9a) there is warming in the modelled temperatures in the upper troposphere and lower stratosphere even in the *fODS-perturbation* that is not captured in the <u>(Fig. 9g).FDH calculations.</u>

3. Pages 2-3: Clarify focus here is not only on tropospheric ozone.

We have clarified that changes in tropospheric and stratospheric ozone are quantified.

This study <u>assesses ozone radiative forcing using a combination of definitions and methodologies asis</u> part of the Tropospheric Ozone Assessment Report Phase II (TOAR-II). <u>It quantifies forcing due to changes in both tropospheric and stratospheric ozone.</u>

In section 2 a discussion of past studies of stratospheric ozone forcing is added:

Forcing from stratospheric ozone changes is sensitive to the altitude of the change, with decreases in lower stratospheric ozone contributing a negative forcing and decreases in upper stratospheric ozone contributing a positive forcing (Skeie et al., 2020). Estimates of historical stratospheric ozone forcing have therefore been uncertain even in the sign. IPCC AR6 (Forster et al., 2021) references a forcing from historical stratospheric ozone changes of  $0.02 \pm 0.07$  W m<sup>-2</sup> based on offline kernel SARF calculations in Skeie et al. (2020).

The ozone radiative forcing from historical ODS increases is more robustly negative as it excludes contributions from increasing ozone precursors and includes the impact of ozone depletion on upper tropospheric concentrations. IPCC AR5 (Myhre et al., 2013) quantified an ozone SARF attributed to ODSs of  $-0.15 \pm 0.15$  Wm<sup>-2</sup> (where the uncertainty is 5-95% confidence limit) and AR6 (Szopa et al., 2021) a value of -0.16 Wm<sup>-2</sup> (no confidence limit provided). An ozone ERF of  $-0.04 \pm 0.03$  Wm<sup>-2</sup> due to the historical ODS increase of  $-0.04 \pm 0.03$  Wm<sup>-2</sup> has been calculated from one model (Michou et al., 2019), but it is not clear that this can be directly compared to the SARF calculations.

4. Line 42: Include example of a non-cloud adjustment the first time this term is mentioned.

We now give water vapour and albedo changes as examples.

Non-cloud adjustments (excluding stratospheric temperature) due to water vapour and albedo changes are positive (both LW and SW).

5. Figure 1: Clarify if this is for tropospheric ozone only?

We have clarified that this is for ozone changes up to 0.1 hPa

Figure 1: Radiative efficiencies (SARF) for ozone changes up to 0.1 hPa SARF in mWm<sup>-2</sup> per DU based on calculations in Skeie et al. (2020)

6. Section 2.1: Different information is provided for different models (number of chemical species, reactions given for some but not others). Consider a summary Table with consistent information provided for all models. This table could be main text and detailed text describing unique model aspects could move to supplement to shorten main text.

Thank you for the suggestion. We have now included such a table and moved the more detailed model descriptions into the Supplementary Material.

| Model | Model        | <u>Horizontal</u> | No of       | Chemistry  | No. of  | No. of re  | eactions |               |            | Radiative | Reference       |
|-------|--------------|-------------------|-------------|------------|---------|------------|----------|---------------|------------|-----------|-----------------|
| Name  | <u>Type</u>  | Resolution        | Vertical    | scheme     | species | Gas-       | Aqueous  | <u>Hetero</u> | Photolysis | transfer  |                 |
|       |              |                   | levels      |            |         | phase      | -phase   | geneou        |            | scheme    |                 |
|       |              |                   | (Model lid) |            |         |            |          | <u>s</u>      |            |           |                 |
| CESM2 | <u>Earth</u> | 0.9°x1.25°        | 32 (2.26    | MOZART-    | 221     | <u>405</u> |          | <u>17</u>     | 123        | RRTMG     | Danabaso        |
|       | System       |                   | <u>hPa)</u> | <u>TS1</u> |         |            |          | (strat.)      |            |           | glu et al.      |
|       | Model        |                   |             |            |         |            |          |               |            |           | <u>(2020)</u> , |
|       |              |                   |             |            |         |            |          |               |            |           | <b>Emmons</b>   |
|       |              |                   |             |            |         |            |          |               |            |           | et al.          |
|       |              |                   |             |            |         |            |          |               |            |           | (2020)          |

etc

7. Lines 341-342: Was stabilization achieved in all the runs?

We clarify that the spin ups were long enough for the models to stabilise.

This was found to be sufficient for stabilisation.

8. Section 2.4: mention here that IRF is also calculated (line 797 points to IRF kernels but only SARF kernels are introduced)

We now mention that an IRF kernel is used too.

For comparison with the online IRF calculations the IRF kernel is the sum of SW and LW for either clear sky or all sky.

9. Figure 6: Is the tropopause used for the observations consistent with the definition in the models? Clarify that the TOAR value is from this study.

The comparison is for total column ozone.

### We have added clarification that the TOAR value is from this study.

The <u>TOAR-RF</u> multi-model mean (black diamond), and the inter-model spread expressed as ±1 standard deviation <u>from this study</u> is based on the models: CESM2, EMAC, GEOS-Chem, GFDL-ESM4, GISS-E2.1\_FR, NorESM2, and UKESM1-0-LL.

10. Sections 3.3/3.4/3.5 and 4.3/4.4: Add a final summary sentence or short paragraph to convey the key message(s) emerging from the findings reported in each section?

We have added summary sentences to these sections.

4.3

In summary, the net all-sky multi-model mean ERF is similar to the IRF within the uncertainties due to a reduction in the LW forcing, but an increase in the SW forcing. The ERF is higher than the SARF for the one model that calculated it, particularly in the clear-sky.

4.4

There is close agreement between models in the tropospheric forcing, but more model variability when including stratospheric changes as indicated by the large standard deviation across models in the total ozone radiative forcing.

#### 4.5

Overall the models show a robust decrease in cloud cover in response to increased ozone. This causes a significant increase in SW forcing and a significant decrease in LW forcing, with a small decrease in net forcing that varies across models contributing to the differences in how rapid adjustments enhance or reduce the ERF compared to IRF.

5.3

## 5.3.3 Summary of clear-sky comparison

Both experiments show a decrease in surface albedo, with a consequently larger SW clear-sky ERF than SARF. This has a proportionally greater effect for the fODS perturbation. In the LW clear-sky the ERF is larger than the kernel SARF for the standard perturbation. For the effect of ODSs (standard minus fODS), the LW clear-sky ERF even has a different sign. This sign change between LW clear-sky ERF and SARF is also the case for the EMAC model which diagnoses both online. Since the kernel LW IRF was found to be 40% greater than the models (Section 5.2) the true increase of ERF compared to the SARF could be even larger.

5.4

There is a suggestion that the cloud adjustment is negative for the ozone precursor changes, and positive for the ODS changes, but the uncertainties are largeand no systematic difference between the standard and fODS perturbations. Although uncertain Nevertheless, the cloud adjustment (-0.1 to +0.05 Wm<sup>-2</sup>) is a significant fraction of the total ERF (0.27±0.04-08 Wm<sup>-2</sup>).

This has been clarified to explain that the meaningful finding is that the different cloud fields mean that the fluxes are noisy

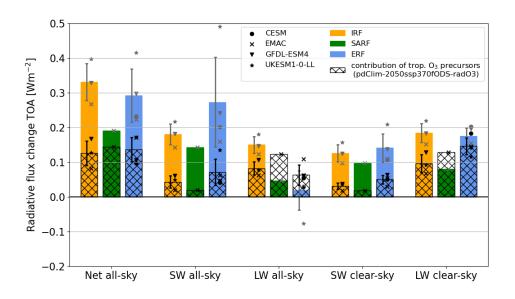
For all-sky ERF, the fluxes show are considerable noise due to cloud changes between the simulationsy as this diagnostic is calculated from two simulations (*EMAC pdClim 2050ssp370 radO3* minus *EMAC pdClim control*), which feature different cloud fields (see Sect. 34.5). This demonstrates that even with nudging there is large variability in cloudiness.

12. Line 653. GEOS-Chem reads in the cloud fields, so rather than a zero response it's the same clouds being used as input.

Agreed, this has been removed from the text and the cell in the table has been changed to "N/A".

13. Lines 720-722. So tropospheric ozone precursors and ODS (&N2O?) contribute equally to ERF in 2050 in this scenario? Figure 12 might better illustrate this by using hatching as in Figure 8. Consider adding this point to the abstract.

Thank you for your suggestion on the bar chart. We have revised this to better illustrate the comparison.



We agree on the importance of the ODS contribution, and have added this point to the abstract.

This increase makes ozone the second largest contributor to future warming by 2050 in this scenario, approximately half of which is due to stratospheric ozone recovery and half due to tropospheric ozone precursors.

An additional simulation varying N2O has been performed by one model (UKESM1). The text describing N2O in section 4.7 has been changed to include these calculation.

An additional effect that we cannot isolate with the *fODS* perturbation simulation is the impact of increasing  $N_2O$  emissions concentrations increase from a year-2015 value of 328 to 362 nmol mol<sup>-1</sup> by 2050 –in the *fODS*-perturbation simulation following SSP3-7.0. The effects on ozone of this  $N_2O$  increase have been tested in one model (UKESM1.0-LL). The increase in  $N_2O$  is in the SSP3-7.0 scenario on ozone. In the stratosphere,  $NO_x$ -is produced from  $N_2O$  and hence  $N_2O$  is foreseen to be an important factor with respect to ozone depletion in the 21<sup>st</sup> century . Prescribing increasing  $N_2O$  surface mixing ratios leads to an ozone reduction in the middle and upper stratosphere, which would counteract the increase of stratospheric ozone related to the expected decrease

in ODSs until 2050 (; results based on a different emission pathway), despite ozone loss due to  $NO_*$  being less efficient when  $CO_2$  and  $CH_4$ -concentrations increase\_is found to have a negligible effect on the total ozone column (-0.2 DU) due mostly to depletion in the upper stratosphere (Ffig. S10?). It does cause a positive radiative forcing (0.034  $\pm$  0.043 Wm<sup>-2</sup>-ERF, 0.029 Wm<sup>-2</sup> SARF) since ozone in the upper stratosphere has a negative radiative efficiency (Fig.

## **Technical Corrections**

1. On Figure 5, is the topography used to determine where to plot as a function of pressure the same in the different panels? Around 40S and 90N there are some differences.

Thank you for pointing this out. There was an error in how topography was used, which has now been corrected.

2. Bodeker Scientific, 2024 is not in the bibliography

**Fixed** 

### Other changes:

#### A table of ozone column differences has been added to section 4.1

| Madala             | <u>2050 - 2015 Di</u> | fference (DU) | 2050 fODS – 201 | 2050 fODS – 2015 Difference (DU) |  |  |
|--------------------|-----------------------|---------------|-----------------|----------------------------------|--|--|
| Models             | <u>TCO</u>            | <u>TrCO</u>   | <u>TCO</u>      | <u>TrCO</u>                      |  |  |
| UKESM1.0           | <u>19.6</u>           | <u>4.7</u>    | <u>5.7</u>      | <u>3.1</u>                       |  |  |
| GFDL-ESM4          | <u>12.2</u>           | <u>4.1</u>    | <u>4.9</u>      | <u>3.3</u>                       |  |  |
| <b>EMAC</b>        | <u>13.6</u>           | <u>4.0</u>    | <u>3.8</u>      | <u>3.5</u>                       |  |  |
| GISS-E2.1_FR       | <u>4.3</u>            | <u>3.0</u>    | <u>N/A</u>      | <u>N/A</u>                       |  |  |
| CESM2              | <u>11.5</u>           | <u>4.4</u>    | <u>4.8</u>      | <u>3.8</u>                       |  |  |
| NorESM2            | <u>9.1</u>            | <u>4.1</u>    | <u>N/A</u>      | <u>N/A</u>                       |  |  |
| GEOS-Chem          | <u>17.2</u>           | <u>5.4</u>    | <u>N/A</u>      | <u>N/A</u>                       |  |  |
| Multi-model mean   |                       |               |                 |                                  |  |  |
| ± 1 std. deviation | $12.2 \pm 5.2$        | $4.3 \pm 1.0$ | <u>N/A</u>      | <u>N/A</u>                       |  |  |
| (7 models)         |                       |               |                 |                                  |  |  |
| Multi-model mean   |                       |               |                 |                                  |  |  |
| ± 1 std. deviation | $14.2 \pm 3.2$        | $4.3 \pm 0.3$ | $4.8 \pm 0.7$   | $3.4 \pm 0.3$                    |  |  |
| (4 models)         |                       |               |                 |                                  |  |  |

Table 4: Differences in global multi-annual mean total column ozone (TCO) and tropospheric column ozone (TrCO) in Dobson Units (DU) between 2015 and 2050 for pdClim-2050ssp370-radO3 relative to pdClim-control from the 7 members of the TOAR-RF ensemble and for pdClim-2050ssp370fODS-radO3 relative to pdClim-control from the 4 models that ran the sensitivity simulation (in bold). Multi-model means and standard deviations are shown for the 7-member and 4-member ensembles.

Specific humidity profiles have been added to figure 9, and text describing them has been added to section 4.5

The vertical profile of relative change in humidity (expressed as a % change in specific humidity, shown in Fig. 9e) shows a maximum increase of 3 to 5% in the stratosphere around 100 hPa. In the troposphere, the change is smaller and increases with height from the surface to the tropopause. The change in the stratosphere humidity is due to meteorological adjustments, such as through higher tropospheric humidity and higher tropical UTLS temperature, rather than chemical adjustments. The experimental set up was designed to have similar chemical water vapour production from methane oxidation in all simulations).

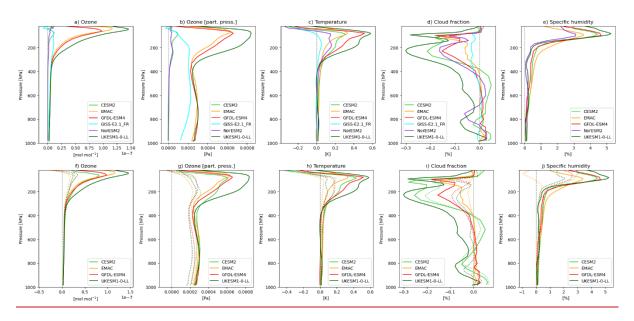


Fig 9: Global mean profile of difference in (a, ef) ozone mole fraction, (b, gf) ozone partial pressure (Pa), (c, hg) temperature (K), and-(d, ih) cloud fractioneover (%), and of relative difference in specific humidity (e, j) between the 2050 and 2015 (pdClim-control) state. The top row panels show the results from the standard experiment pdClim-ssp2050ssp370-radO3 minus pdClim-control, the bottom panels show for the models that ran the fODS-perturbation experiment (CESM2, EMAC, GFDL-ESM4, UKESM1-0-LL) the results from the standard (pdClim-ssp2050ssp370-radO3 minus pdClim-control) experiment using solid lines (which are repeated from panels a-ed) and fODS (pdClim-ssp2050ssp370fODS-radO3 minus pdClim-control) experiments using dotted lines.

## A section 4.6 describing the albedo changes has been added

## 4.6 Albedo changes

The clear-sky SW ERFs include a component from changes in the surface albedo due to changes in snow and ice cover. Surface albedo was diagnosed as the ratio of the upward and downward clear-sky SW fluxes at the surface. The albedo change can be approximately converted to a clear-sky SW forcing using the formula  $\Delta F = -T_a^{cr} \times I_t \times \Delta \alpha_s$  (Qu & Hall, 2006) where  $T_a^{cr}$  is the effective atmospheric clear-sky transmissivity (taken to be 0.7),  $I_t$  is the incoming SW TOA flux and  $\Delta \alpha_s$  is the change in albedo. Results are shown in Table 65. The change in surface albedo is consistent among the models (apart from EMAC) with a decrease of around  $2\times10^{-4}$ . The interannual standard deviation in the surface albedo is large (around  $3\times10^{-4}$ ) so a few decades of simulation are needed to reduce the standard error on the mean. The EMAC model has the smallest change in albedo. It has a large interannual standard deviation (around  $10\times10^{-4}$ ), but the nudging ensures these are correlated between control and perturbations so that the interannual standard deviation in the albedo difference is very small (around  $0.05\times10^{-4}$ ). It appears that nudging the meteorology significantly reduces the albedo response in EMAC.

|                  | Surface albedo change                         | Albedo forcing [W m <sup>-2</sup> ] |
|------------------|---|-------------------------------------|
| CESM2            | $-2.2 \times 10^{-4} \pm 0.8 \times 10^{-4}$  | $0.052 \pm 0.025$                   |
| <u>EMAC</u>      | $-0.4 \times 10^{-4} \pm 0.03 \times 10^{-4}$ | $0.010 \pm 0.001$                   |
| <u>GFDL-ESM4</u> | $-1.6 \times 10^{-4} \pm 0.5 \times 10^{-4}$  | $0.038 \pm 0.011$                   |
| NorESM2          | $-1.8 \times 10^{-4} \pm 0.7 \times 10^{-4}$  | $0.042 \pm 0.017$                   |
| UKESM1-0-LL      | $-2.5 \times 10^{-4} \pm 0.7 \times 10^{-4}$  | $0.060 \pm 0.016$                   |
| Multi-model mean | $-1.7 \times 10^{-4} \pm 0.7 \times 10^{-4}$  | $0.040 \pm 0.017$                   |

Table 6: Global mean absolute surface albedo change and associated forcing from pdClim-2050ssp370-radO3 minus pdClim-control. Uncertainties for individual models are errors on the mean. Uncertainties for the multi-model mean

are standard deviations across the models. Forcings are calculated using the formula in the text. Only models that provided surface SW fluxes are shown.

There is a robust decrease in surface albedo which translates to a positive SW contribution to the ERF.

## The albedo changes are discussed in 5.3.1

By excluding cloud effects, the clear sky ERFs should compare to the kernel SARFs if the tropospheric non-cloud adjustments are small. The modelled clear-sky SW ERF correlates well with the ISARF (equal to SARF in SW) kernel calculations (see Fig. 15). Talthough the ERF calculation is consistently higher than the IRF (see Ttable 76). This is also the case using the double-call clear-sky SW IRF for the models that diagnose it, the ratio depending on whether ODS changes are included (standard) or not (fODS). The clear sky SW ERF is also higher than the online IRF (Tables S1 and S3). This implies a positive clear sky adjustment. The largest clear sky SW adjustment would be expected to come from the surface albedo change such as snow cover, which is expected to be positive Click or tap here to enter text... is expected from the decreases in surface albedo diagnosed in Section 4.6. For the fODS experiments the albedo adjustment explains the difference between the ERF and IRF such that the residual defined as ERF-IRF-adjustment is zero, suggesting that albedo changes explain the difference. In the standard experiments when decreases in ODSs are included the residual is negative, suggesting that there may be further negative adjustments in the SW as a response to stratospheric ozone recovery. The ozone recovery leads to an increase in stratospheric water vapour (Fig. 9(j)), but it is not obvious why this would lead to a negative SW forcing adjustment.

In this study we define the ERF using fixed SSTs. If land temperatures were also fixed it is likely that the albedo adjustment would be substantially reduced. If an ERF defined using fixed SSTs and land temperatures were required, as in IPCC AR6 (Forster et al., 2021), then it may be necessary to subtract the albedo adjustment from the diagnosed fSST ERFs. This would be analogous to subtracting the land surface temperature adjustment as in Tang et al. (2019).

|              | Clear-sky SW                 | Clear-sky SW      | Clear-sky SW                  | Albedo                        | Residual           |
|--------------|------------------------------|-------------------|-------------------------------|-------------------------------|--------------------|
|              | Double-call                  | kernel IRF        | <u>ERF</u>                    | <u>adjustment</u>             |                    |
|              | <u>IRF</u>                   |                   |                               |                               |                    |
| Standard     |                              | $0.115 \pm 0.032$ | $0.131 \pm 0.042$             | $\underline{0.040 \pm 0.017}$ | $-0.024 \pm 0.012$ |
| Standard*    | $\underline{0.125 \pm 0.25}$ |                   | $\underline{0.152 \pm 0.042}$ | $\underline{0.036 \pm 0.020}$ | $-0.010 \pm 0.012$ |
| <u>fODS</u>  |                              | $0.034 \pm 0.007$ | $0.050 \pm 0.012$             | $0.017 \pm 0.10$              | $0.000 \pm 0.004$  |
| <u>fODS*</u> | $0.031 \pm 0.009$            |                   | $\underline{0.050 \pm 0.014}$ | $\underline{0.016 \pm 0.12}$  | $0.003 \pm 0.007$  |

<sup>\*</sup> Only models that included double-call diagnostic of IRF.

Table 7: Comparison of the difference between ERF and IRF with the albedo adjustment in the clear-sky SW. Two definitions of IRF are used, kernel and double-call. Only a subset of models included double-call diagnostics. The residual is defined as ERF - IRF - albedo adjustment.