

Author's response to referee comments on "The Impact of Future Emission Policies on Tropospheric Ozone using a Parameterised Approach"

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We would like to thank all of the reviewers for their helpful and constructive comments. Below we have responded to each comment in turn and made alterations to the manuscript where appropriate (shown enclosed in "speech marks and italic font" and any deletions from the manuscript shown with a strikethrough "~~example~~"). The referee comments are shown first in grey shading and the author's response is shown below in normal font.

10

Response to Referee 1

Overall, I think this will be an important and useful paper detailing the impact of a great many future emission scenarios on surface ozone and radiative forcing. In general, the paper is well written with lots of detailed tables and clear figures. I would recommend publication following the changes detailed below.

15 Major Comments

1. Overall, I found the explanation for the parameterization rather confusing and not straightforward. Please think about how to make the explanation more precise. Some details...P4, L5: "the scale factor, f, is replaced by g". This is a rather confusing way to put it. Why don't you write out the full parameterization from the beginning by defining the various terms in equation (2) dependent on the constituent and not explain the parameterization by first defining f, then replacing f with g? Once could easily expand equation (2) to include the definitions for the various constituents. In addition, equation (2) is written as one might write out a computer code, but does not make sense from a mathematical viewpoint. Where does the factor (2f-g) come from. The factor g is evidently different for both CH₄ and NO_x from that given in equation 2? (equation 3). This should be discussed at the beginning and not mid-way through the paper. Where exactly is the ozone adjustment factor used (page 10)?
20
25 Section 2.1 is titled "Original Ozone Parameterization", but as far as I understand it is also the parameterization used in the present paper.

We would like to thank the reviewer for their useful comments on the initial description of the parameterisation and appreciate some of the confusion in the description. The parameterisation used in this study is fully consistent with that of Wild et al., (2012) but we have updated the notation to make it easier to follow.

30 Section 2.1. has been renamed as "*Parameterisation of Ozone*".

To avoid some of the confusion with equation 2 in the manuscript a new fractional emission change factor (r) is defined as below and becomes equation (1):

$$r_{ij} = \frac{\Delta E_{ij}}{-0.2 * E_{ij}} \quad (1)$$

35 Equation (2) in the original manuscript has been replaced with the following three equations (which now become equations 3, 4 and 5) to clearly identify the different emission scaling factors and when they are used in the parameterisation.

$$f_{ij} = r_{ij} \quad \text{Linear Scaling of } O_3 \text{ response} \quad (3)$$

$$f_{ij} = 0.95r_{ij} + 0.05r_{ij}^2 \quad \text{Scaling accounting for reduced } O_3 \text{ increases from } NO_x \text{ and } CH_4 \quad (4)$$

$$f_{ij} = 1.05r_{ij} - 0.05r_{ij}^2 \quad \text{Scaling for titration regimes where decreasing } NO_x \text{ increases } O_3 \quad (5)$$

In addition, a schematic (shown below) has now been included in the supplementary as Figure S1 to show the various steps to the parameterisation and when to use the different scaling factors.

1. Calculate fractional emission change (r)

$$r_{ij} = \frac{\Delta E_{ij}}{-0.2 \times E_{ij}}$$

2. Generate scaling factors (f)

2.1. Linear scaling of O₃ response

$$f_{ij} = r_{ij}$$

2.2. Non-linear scaling accounting for reduced O₃ increases from NO_x and CH₄

$$f_{ij} = 0.95r_{ij} + 0.05r_{ij}^2$$

2.3. Non-linear scaling for titration regimes where decreasing NO_x increases O₃

$$f_{ij} = 1.05r_{ij} - 0.05r_{ij}^2$$

3. Apply different scaling factors to precursors and source regions under different chemical regimes

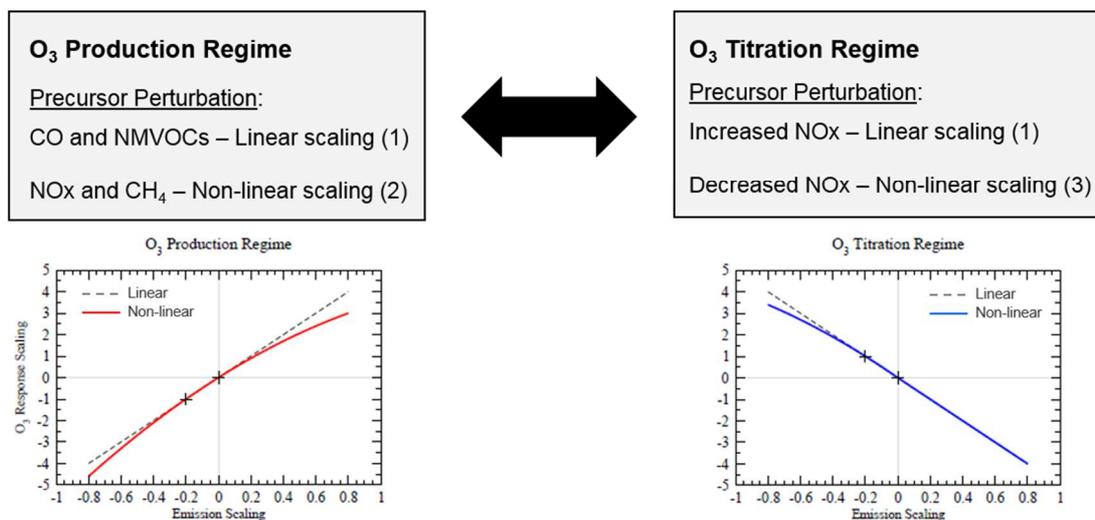


Figure S1 – Schematic showing the different steps used in the parameterisation from calculating a fractional emission change (1), to generating an emission scaling factor (2) and applying this to the appropriate precursor in a particular chemical regime (3). The figures at the bottom illustrate the effect of applying the quadratic function compared to the linear one in the different chemical regimes.

We have amended Section 2.1 to make it clearer and easier to read and reflect the changes above. For the convenience of the reviewer we present the entire revised section 2.1 below.

10 “2.1 Parameterisation of Ozone

The parameterisation developed in this study is based on an earlier version developed from the TF-HTAP1 experiments by Wild et al., (2012). This simple parameterisation enabled the regional response in surface O₃ concentrations to be estimated based on changes in precursor emissions and CH₄ abundance. The input for this parameterisation came from 14 different models that contributed to TF-HTAP1. All the models ran the same emission perturbation experiments (20% reduction in emissions of oxides of nitrogen (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs) individually and all together) over the four major northern hemisphere source regions of Europe, North America, East Asia and South Asia. Additional experiments included global perturbations of emission precursors (E), as well as a 20% reduction in global CH₄ abundance. The multi-model O₃ responses from the 20% emission perturbation experiments (ΔO_{3e} for emissions of NO_x, CO and NMVOCs and ΔO_{3m} for CH₄) are then scaled by the fractional emission changes (r) from a given emission scenario over each source region (Eq. 1).

$$r_{ij} = \frac{\Delta E_{ij}}{-0.2 * E_{ij}} \quad (1)$$

The monthly mean O_3 response (ΔO_3) is calculated as the sum over each receptor region (k) of the scaled O_3 response from each model to the individual precursor species (i - CO , NO_x and $NMVOCs$) in each of the five source regions (j - Europe, North America, East Asia, South Asia and rest of the world), including the response from the change in global CH_4 abundance (Eq. 2, reproduced from Wild et al., 2012).

$$\Delta O_3(k) = \sum_{i=1}^3 \sum_{j=1}^5 f_{ij} \Delta O_{3e}(i, j, k) + f_m \Delta O_{3m}(k) \quad (2)$$

$$f_{ij} = r_{ij} \quad \text{Linear Scaling of } O_3 \text{ response} \quad (3)$$

$$f_{ij} = 0.95r_{ij} + 0.05r_{ij}^2 \quad \text{Scaling accounting for reduced } O_3 \text{ increases from } NO_x \text{ and } CH_4 \quad (4)$$

$$f_{ij} = 1.05r_{ij} - 0.05r_{ij}^2 \quad \text{Scaling for titration regimes where decreasing } NO_x \text{ increases } O_3 \quad (5)$$

A linear emission scale factor (Eq. 3) is used in Eq. 2 for each emission scenario involving the precursor emissions CO and $NMVOCs$ and is defined as the ratio of the fractional emission change to the 20% emission reduction in the TF-HTAP1 simulations. A similar scale factor for methane (f_m) is based on the ratio of the change in the global abundance of CH_4 to that from the 20% reduced CH_4 simulation ($\Delta[CH_4]/-0.2 \times [CH_4]$). Perturbations to emissions of CO , NO_x and $NMVOCs$ induce a long-term (decadal) change in tropospheric O_3 from the change in the oxidising capacity of the atmosphere (OH) and the CH_4 lifetime (Wild and Akimoto, 2001; Collins et al., 2002; Stevenson et al., 2004). The long-term impacts from 20% global emission reductions can reduce the O_3 response by 6-14% from NO_x emission changes and increase the O_3 response by 16-21% from CO changes (West et al., 2007). This long-term response is not accounted for in the simulations used here as CH_4 abundances are fixed.

Wild et al., (2012) found that this simple linear scaling relationship between emissions and surface O_3 was sufficient for small emissions perturbations, but that the relationship started to exhibit larger non-linear behaviour for larger perturbations, particularly for NO_x . The linear scaling factor was found to be sufficient for the surface O_3 response from emission perturbations of CO and $NMVOCs$ as non-linear behaviour from these precursors is small (Wu et al., (2009). To account for non-linear behaviour of surface O_3 to NO_x emission changes, a quadratic scaling factor (Eq. 4) is used, based on additional simulations of surface O_3 response over a larger range of emission perturbations in Wild et al., (2012).

For the special case of source regions that are under titration regimes, where a reduction in NO_x emissions may lead to an increase in O_3 , the curvature of the response is reversed for NO_x emission decreases (Eq. 5), as described in Wild et al., (2012). A linear scale factor is used for emission increases under these conditions (Eq. 3). The spatial extent of ozone titration is assumed constant as the parameterisation is based on differences between two model simulations and is therefore unable to represent any future changes in chemical regime.

The surface O_3 response to changes in global CH_4 abundances shows a similar degree of non-linearity as that from changes in NO_x emissions (Wild et al., 2012). Therefore, the non-linear scale factor (Eq. 4) is also used to represent the O_3 response to changes in CH_4 abundances.

In summary, the surface O_3 response to CO and $NMVOc$ emission perturbations is represented by the linear scale factor (Eq. 3) and to changes in NO_x emissions and CH_4 abundances by the non-linear scale factor (Eq. 4). For source regions under titration regimes, the surface O_3 response to NO_x emissions is limited by Eq. 5 for emission decreases but uses the linear scale factor (Eq. 3) for emission increases. The parameterisation is represented schematically in Figure S1 of the supplementary material."

Equation 3 was generated from TF-HTAP2 models to test whether the non-linear representation from Wild et al., (2012) was still valid. Since the response in this equation is comparable to that in Wild et al., (2012) it was decided

for consistency to retain the same non-linear scaling factor and as such is presented in Section 3.3 as a comparison to TF-HTAP1. The adjustment factor on page 10 was only used to compare the O₃ response from CH₄ between TF-HTAP1 and TF-HTAP2 models. The incorporation of O₃ response to CH₄ within the parameterisation are described within Section 2.3.3 (Additions from TF-HTAP2) on Page 7 Lines 1 to 4.

5 2. I think the paper could do a better job of emphasizing which results should be believed and which should be treated with skepticism. There are a number of emission scenarios where the emission change is over 50% (either with a positive or a negative change). To what extent should the results from these scenarios be believed? Results that should be treated with caution could be clearly indicated in the tables.

10 The radiative forcing calculation does not seem particularly accurate. The authors claim that the parameterization reproduces the ACCMIP changes fairly well (p13, l23), but if I understand correctly the parameterized radiative forcing should be 20 to 30 MW m⁻² larger than the ACCMIP results (as it does not account for the climate feedbacks which represent a large part of the ACCMIP signal). Thus, it looks like in most cases the future mean radiative forcing is dramatically underestimated (although, perhaps with the extremely large error bars in ACCMIP it is difficult to really say anything meaningful). Unless I missed it, the authors compared the change in the ozone burden between the parameterization and HadGEM2-ES but not the overall radiative forcing. Without some more evidence, and in a context that does not assume changes in climate, it is somewhat difficult to see what the parameterized radiative forcing calculation adds. I am willing to be convinced otherwise, but do need some convincing.

20 The paper often makes somewhat vague statements about the comparison of the parameterization to explicit results (e.g., it states that the parameterization is valid, or compares well : :). It would be nice to see in the conclusion a somewhat more explicit discussion of when and under what scenarios the parameterization should be believed: e.g., should it be believed under scenarios with large changes (e.g., +/- 50%), over regions where decreasing NO_x increases ozone, over southeast Asia even with small emission changes, for the radiative forcing in 2100 given the strong climate influence etc? In other words, the certainty bounds should be discussed and

25 quantified in more detail with an overall summary given in the conclusions.

In section 3.1 we evaluate the parameterisation for emission reductions of 20%, 50% and 75% over Europe, a region where deviation from linear behaviour can be large. Detailed testing and evaluation of the limitations of the parameterisation was carried out in Wild et al., (2012), where emission perturbations ranging from a doubling of emissions to a complete removal were undertaken. Wild et al., (2012) found that errors remained below 1 ppbv for emission changes of less than +/- 60% and we find very similar results, with regional monthly mean errors below this level even with a 75% emission reduction (Fig. 2). We do not expect the parameterisation to work as well for large emission perturbations of greater than 75% or for source regions under a titration regime. The limitations of the parameterisation are further discussed in Section 3.2 and summarised on P9 Lines 13 to 16 where the parameterisation is compared and evaluated against global model simulations. But based on the reviewers comments we have attempted to make the limitations of the parameterisation clearer in the

35 manuscript by including the following changes.

P7 line 30 has been amended to:

40 *“This small internal error between the parameterisation based on HadGEM2-ES and HadGEM2-ES simulations indicates that the parameterisation of O₃ is working well for emission changes at least as great as 50%. This is similar to the results of Wild et al., (2012) where more detailed testing found that that the parameterisation resulted in errors of < 1 ppbv for emission perturbations of up to +/- 60%. Here, monthly mean errors are < 1 ppbv for a 75% emission reduction (Fig. 2). The parameterisation is not expected to perform as well for emission perturbations of larger than +/- 60% and in source regions under titration regimes.”*

Discussion of the limitations of the parameterisation have also been added to the conclusion section as described below:

P14 Lines 19 to 20 have been removed

~~*“The parameterisation is shown to perform well under most conditions, although there are larger uncertainties for future surface O₃ responses over South Asia where changes in emissions are particularly large.”*~~

New text has been inserted at P14 Line 18 of the conclusions to provide a discussion of the limitations:

“However, larger errors may occur when using emission changes of greater than +/- 60% and when considering long term future scenarios where there may be a significant influence from climate change. In addition, the parameterisation may not perform well over regions where chemical titration is expected to become dominant in the future under large emission increases e.g. South Asia, as it is based on the ozone responses in 2010.”

Thank you to the reviewer for their useful comments on the comparison to the ACCMIP models. We have completely revisited this section in the manuscript. Results in the APCD manuscript for the comparison to ACCMIP were based on an adjustment to O₃ responses from emissions perturbations over the period 2010 to 2030 to correct them to the baseline year of 2000. To provide a more direct comparison with the ACCMIP models and the results from Wild et al., (2012) we have re-calculated the responses using the baseline year from the TF-HTAP1 models (2001) and the fractional changes in total emissions based on the period 2000 to 2030.

The results from this more consistent comparison are presented below as an amendment to Table 6, Table 9 and Figure 5 in the original manuscript. The global annual mean surface O₃ response for the CMIP5 scenarios in 2030 and 2050 now shows a very close agreement between the parameterisation in this study and that of Wild et al., (2012). Similarly, the prediction of the change in global surface O₃ response by the parameterisation is within the spread of the ACCMIP multi-model mean response for the CMIP5 RCPs in 2030. The calculated change in global O₃ burden and O₃ radiative forcing for each of the CMIP5 RCPs is now within 1 standard deviation of the ACCMIP multi-model mean response. Differences between the parameterisation and the ACCMIP models are expected as the latter contain the influence of climate change and stratospheric sources on tropospheric ozone. The climate change signal for the ACCMIP models reported in Stevenson et al., (2013) was -24 +/- 27 mW m⁻² over the period 1850 to 2000 and -25 +/- 25 mW m⁻² for RCP8.5 for the period 1850 to 2030. Therefore, as expected, there is only a relatively small influence of climate change over the period of 2000 to 2030s.

An additional test was performed for emission perturbations under the ECLIPSEv5a current legislation scenario in 2030 comparing the parameterisation based only on HadGEM2-ES input with HadGEM2-ES simulations. The results (new Table 5 below) show that the parameterisation is able to reproduce the changes seen in the HadGEM2-ES model simulations based only on emission perturbations.

The following amendments have been made to the manuscript figures, tables and text to reflect the above results.

Table 6 has been replaced in the manuscript by the following, now identified as Table 7:

Table 7. Annual mean surface O₃ change (ppbv plus one standard deviation) in 2030 and 2050 (relative to 2000) for each RCP scenario derived from the parameterisation in this study and that of Wild et al., (2012).

CMIP5 RCP	Global Surface O ₃ response from 2000 to 2050 (ppbv)			
	This Study		Wild et al., (2012)	
	2030	2050	2030	2050
RCP2.6	-1.1 +/- 0.1	-1.9 +/- 0.3	-1.1 +/- 0.3	-2.0 +/- 0.5
RCP4.5	-0.1 +/- 0.1	-0.8 +/- 0.2	-0.2 +/- 0.2	-0.8 +/- 0.4
RCP6.0	-0.4 +/- 0.1	-0.4 +/- 0.1	-0.4 +/- 0.1	-0.4 +/- 0.2
RCP8.5	+1.0 +/- 0.2	+1.5 +/- 0.5	+1.0 +/- 0.2	+1.5 +/- 0.5

Table 9 has been replaced in the manuscript by the following, now identified as Table 10:

- 5 **Table 10. Multi-mean parameterised responses in annual mean surface ozone, global ozone burden and ozone radiative forcing in 1980 and for the CMIP5 emission scenarios in 2030, with changes calculated relative to the year 2000 for comparison with values from ACCMIP (+/- 1 standard deviation of multi-model responses).**

Year	Surface Ozone (ppbv)		Ozone Burden (Tg)		Ozone Radiative Forcing (mW m ⁻²)	
	Param	ACCMIP*	Param	ACCMIP*	Param	ACCMIP*
1980	-1.3	-1.3 +/- 0.4	-17	-15 +/- 6	-67	-59 +/- 21
2000	0	0	0	0	0	0
RCP2.6 2030	-1.1	-1.5 +/- 0.6	-12	-12 +/- 8	-45	-45 +/- 30
RCP4.5 2030	-0.1	+0.2 +/- 0.5	+4	+7 +/- 5	+14	+24 +/- 19
RCP6.0 2030	-0.4	-0.8 +/- 1.0	+0.3	-2 +/- 11	+2	-13 +/- 39
RCP8.5 2030	+1.0	+1.5 +/- 0.7	+20	+23 +/- 7	+80	+81 +/- 26

* - Mean change in the tropospheric Ozone burden and radiative forcing between 2030 and 2000s from the ACCMIP models that provided results for each year of each scenario, as presented in Table 5 of Young et al., (2013) and Table 12 of Stevenson et al., (2013).

- 10 The numbers from Wild et al., (2012) on Figure 5 have been slightly amended as shown below.

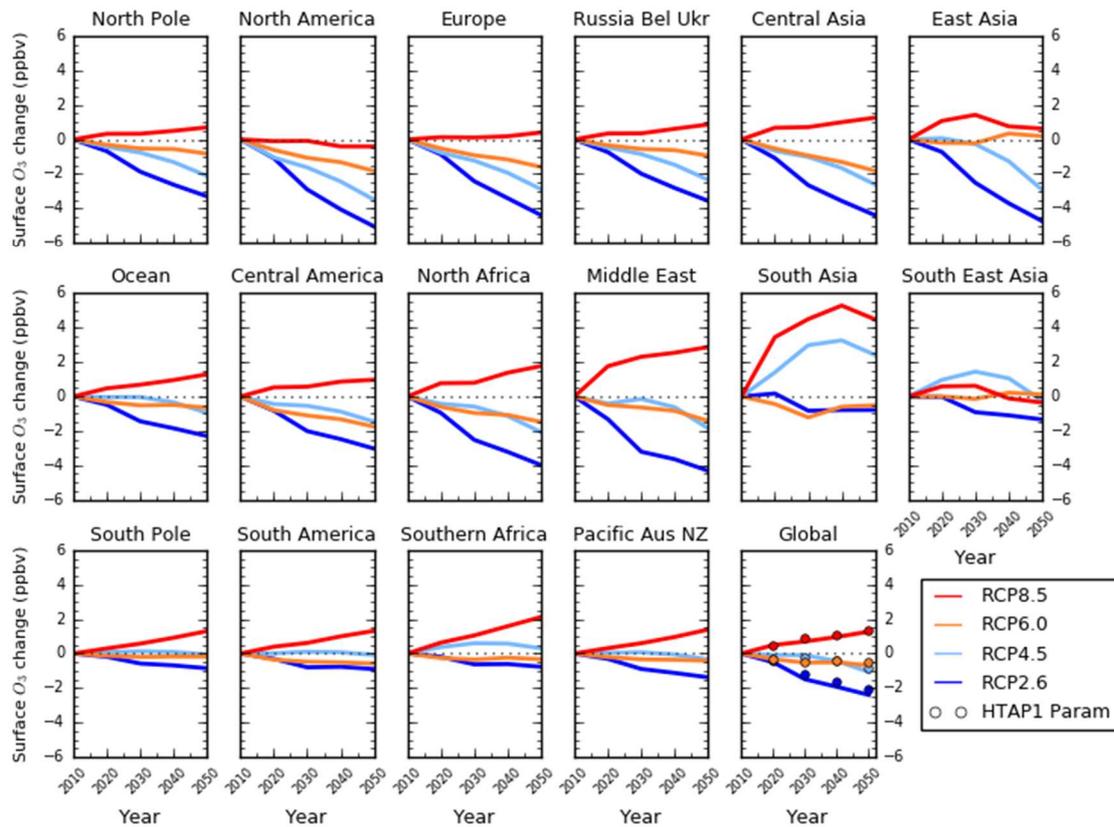


Figure 5: Annual mean regional surface O₃ changes between 2010 and 2050 from the parameterisation for the CMIP5 emissions scenarios of RCP8.5 (red), RCP6.0 (orange), RCP4.5 (light blue) and RCP2.6 (blue). The global surface O₃ response from the parameterisation of Wild et al., (2012) for each scenario is represented as circles, but due to differences in regional definitions a straightforward comparison with TF-HTAP1 regions (Europe, North America, South Asia and East Asia) is not possible.

The following new table (Table 5) has been included in the manuscript now to show the comparison of the parameterisation against HadGEM2-ES simulations for the ECLIPSE CLE scenario in 2030, neglecting any influence from climate change occurs.

Table 5. Parameterised responses based only on HadGEM2-ES input for annual mean surface ozone, global ozone burden and ozone radiative forcing using the ECLIPSE CLE emission scenarios in 2030, with changes calculated relative to the year 2010.

Scenario	Surface Ozone (ppbv)		Ozone Burden (Tg)		Ozone Radiative Forcing (mW m ⁻²)	
	Param ¹	HadGEM2-ES	Param ¹	HadGEM2-ES	Param ¹	HadGEM2-ES*
ECL 2030	-0.21	-0.20	-0.93	-0.95	-0.6	-0.9

¹ Parameterisation based only on HadGEM2-ES input

* Ozone radiative forcing is calculated by applying the same methodology as in the parameterisation (using the relationship between radiative forcing and tropospheric column O₃ change based on multi-model ensemble mean results from ACCMIP)

P8, Line 20 to 22 the sentence is amended to:

“Monthly (Fig. 3) and annual (Table 5) surface O₃ changes between 2010 and 2030 over the TF-HTAP2 regions for the ECLIPSE V5a CLE scenario from the HadGEM2-ES simulation are compared to that from the parameterisation (based solely on HadGEM2-ES model responses and based on responses from all models).”

P13, Line 19 to 31 has been amended to:

“The change in tropospheric O₃ burden and O₃ radiative forcing for the ECLIPSE CLE scenario in 2030 from the parameterisation was evaluated against the change from the equivalent HadGEM2-ES simulation (Table 5), a self-

consistent test based only on emission perturbations with no influence from climate change. The parameterisation is able to reproduce the change in global tropospheric O₃ burden (-0.93 Tg) and O₃ radiative forcing (-0.6 mW m⁻²) simulated by HadGEM2-ES (-0.95 Tg and -0.9 mW m⁻²), with any slight differences due to the discrepancies identified over South Asia (Figure 3)."

5 P13 Lines 22 to 30 have been amended to:

"In comparison to the ACCMIP multi-model mean, the predicted changes between 2000 to 2030 in both global annual mean surface O₃ and global O₃ burden from the parameterisation are within the range of the ACCMIP multi model responses (+/- 1 standard deviation) for all the CMIP5 RCPs (Table 10). The predictions of O₃ radiative forcing in 2030 from the parameterisation across all the RCPs, when the influence of climate change is anticipated to be small, are also consistent with those from ACCMIP. The sign and magnitude of change in global O₃ burden and O₃ radiative forcing with the parameterisation for RCP6.0 is different from the ACCMIP results but is still within the range of model responses, which is the largest for this scenario. The comparison with ACCMIP results shows that the parameterisation is able to reproduce changes in global O₃ burden and O₃ radiative forcing on near-term timescales, when the influence of climate change is small."

10

15 P14 Lines 24 to 27 have been amended as follows:

"Tropospheric O₃ burden and O₃ radiative forcing calculated using the parameterisation are within the spread of the response from the ACCMIP models for all of the CMIP5 RCPs in 2030, where the influence from climate change is anticipated to be small."

Minor Comments:

20 Page 1:

L26 "are valid". This is really rather strong language as the accuracy of the parameterization differs depending on the region. It would be better to quantify this a bit more, saying instead something like "are reasonably accurate for most regions" or "are within the model spread for most regions". Once you say they are valid, it is difficult to quantify how valid are they?

25 Line 26 has been changed to the following:

"Tests against model simulations using HadGEM2-ES confirm that the approaches used within the parameterisation perform well for most regions"

L24. The neglect of climate change is mentioned in regards to radiative forcing but not to changes in surface ozone concentration. It would be important to emphasize that changes in climate and associated changes in climate dependent precursor emissions are neglected at the outset (e.g., in L24).

30

The following sentence has been amended at line 23 to 24:

"Surface and tropospheric O₃ changes are calculated globally and across 16 regions from perturbations in precursor emissions (NO_x, CO, VOCs) and methane (CH₄) abundance only, neglecting any impact from climate change."

35 L32 "across different regions". This is a bit confusing. It might be better to say: "will regionally increase by 1 to 8 ppbv".

Line 32 has been changed to the following:

“Emission changes for the future ECLIPSE scenarios and a subset of preliminary Shared Socio-economic Pathways (SSPs) indicate that surface O₃ concentrations will increase regionally by 1 to 8 ppbv in 2050”

L33 I wonder if it would be clearer to say "change in radiative forcing from 2010 to 2050"?

The sentence on Line 33 has been amended to:

5 *“A change in the global tropospheric O₃ radiative forcing of +0.07 W m⁻² from 2010 to 2050 ...”*

Page 4

L31: which regions are not sources?

The Polar regions are not considered as sources as they were not included in the TF-HTAP2 experiments. Line 31 has been altered to include mention of these regions:

10 *“The source regions were updated in TF-HTAP2 to represent 14 new regions (excluding the North and South Poles), aligned on geo-political and land/sea boundaries (Figure 1).”*

Page 5:

L3. "Models covered". Rather awkward English usage. Models don't cover ...

The word covered on Line 3 has been replaced with “conducted”.

15 L15-16. It would be worthwhile to add a line that the paper discusses in detail how the results TF-HTAP2 are incorporated below.

The following has been added after line 16.

“The following sections discuss in detail how the results from TF-HTAP2 have been incorporated into the parameterisation.”

20 L25. It is not clear how the parameterization has been improved.

The sentence on Line 25 has been changed to:

“The parameterisation of Wild et al., (2012) was used to calculate new baseline O₃ concentrations in 2010 for use in this version of the parameterisation and for comparison to the TF-HTAP2 multi-model mean.”

25 L24-34. In Table 1 the emission differences are averaged for both the MACCity and EDGAR inventories? It is not clear why the authors did not average the emission differences as used in the TF-HTAP2 models versus those in the TF-HTAP1 models. It is unclear how or why internal consistency (whatever that means) should be relevant here. The actual difference in emissions would seem to be more relevant in comparing the change in O₃.

30 The TF-HTAP1 models each used their own emissions as input for experiments (Fiore et al., 2009) and therefore there is no consistent baseline to compare to the prescribed TF-HTAP2 emissions. This is mentioned in the manuscript on P5 Line 19 and Line 29. Therefore the EDGAR and MACCity emission inventories were chosen to provide an emission change between year 2000 and 2010 using consistent datasets in both time periods. The change in precursor emissions between 2000 and 2010 was then used to provide new baseline ozone concentrations in 2010 for use in the parameterisation (see point below). The following text on P5 Line 26 to 30 has been amended to make this clearer.

35 *“The mean fractional change in NO_x, CO and NMVOC emissions between 2000 and 2010 across the TF-HTAP1 source regions from two different emission inventories, MACCity (Granier et al., 2011) and EDGARv4.3.1 (Crippa*

et al., 2016), was used (Table 1), as the use of a specific emission inventory was not prescribed for TF-HTAP1 experiments (See – Fiore et al., 2009)."

"The MACCity and EDGAR inventories provide a consistent set of emissions in 2000 and 2010, enabling the change in emissions between future and historical time periods to be explored."

5 L35 - how many models contributed to the parameterized ozone response? -In general it is not really clear what was done here. Were the emission differences in table 1 used to compute the parameterized change in ozone in table 2? Was the parameterized response from each model computed separately to give the standard deviation in the parameterized response?

10 The parameterisation of Wild et al., (2012) was used to calculate new baseline O₃ concentrations in 2010 (Table 2) using the emission differences in Table 1 (see lines 24 to 34.) This version of the parameterisation used 14 models from the TF-HTAP1 experiments and computed responses individually which were then averaged to give the multi-model mean response and standard deviation from the parameterisation. Line 35 has been amended to make these points clearer.

15 "The parameterised surface ozone response in 2010 was calculated using the method of Wild et al., (2012), based on the individual response of 14 TF-HTAP1 models using the fractional emission changes in Table 1. The parameterised ozone response across the"

L37-38. "similar to the TF-HTAP2 multi-model mean values". This seems a little misleading as the parameterized responses are also similar to the TF-HTAP1 values. It would be more insightful to quantify the extent to which the parameterization quantifies the changes between TF-HTAP1 and TF-HTAP2.

20 The reviewer makes a valid point that the TF-HTAP1 and TF-HTAP2 results are similar and the remaining part of this paragraph goes onto make the point that the larger range in ozone responses for both TF-HTAP1 and TF-HTAP2 dominates the uncertainty in ozone concentrations and is much larger than any response from changing emissions between 2000 and 2010. Therefore any predictions using the parameterisation for ozone response between 2000 and 2010 will still have an uncertainty associated with it mainly due to the large spread in model
25 responses.

Changed Line 27 to 38 to the following:

"Table 2 shows that the O₃ concentrations from the parameterisation (H-P) are within the spread of the individual model values from TF-HTAP2 (H-2), represented by one standard deviation, over most of the receptor regions."

30 Page 6

L5. "adjusted". I assume the authors explain how the emissions are adjusted below.

The emissions were not adjusted here. The O₃ response fields from the European, North American, East Asian and South Asian source regions of TF-HTAP1 models were reapportioned so that a larger number of models (14 in total) than available from TF-HTAP2 are able to represent the equivalent TF-HTAP2 source regions. P6 Lines 8
35 to 10 specifically mentions the adjustment of O₃ response fields with the rest of section 2.3.2 explaining in more detail how this source region adjustment of TF-HTAP1 models was performed.

L11-21. Both the source and receptor regions are changed between HTAP1 and HTAP2. It is unclear from the description here how you discretize the response in the HTAP1 models into both smaller source regions and smaller receptor regions. The discussion in 2.3.2 seems to only concern the source region adjustment. Sections
40 2.3.2 and 2.3.3 should be clarified as to the exact procedure used.

The source region adjustment has been described here as this is the only part of the parameterisation that requires adjusting. Additional simulations were performed with HadGEM2-ES (which participated in TF-HTAP2) using the TF-HTAP1 source regions (Europe, North America, East Asia, South Asia) to inform and evaluate this source region adjustment. The methodology for this is described in section 2.3.2.

- 5 No specific adjustment is required for the receptor regions, as these can be defined arbitrarily based on the global distribution of ozone responses generated in both TF-HTAP1 and TF-HTAP2 studies. Here we have defined the receptor regions to match the source regions used in TF-HTAP2 for consistency. This is referred to on P7 Line 19 which notes 'Output is provided on a standard grid to facilitate the calculation of O₃ responses over any selected receptor regions.' An additional comment referring to the definition of receptor regions has been included on P6
10 Line 25.

"Here, receptor regions are defined in accordance with those in TF-HTAP2 (16 in total), although it is possible to define any required receptor regions using the global distribution of O₃ responses."

Page 7

-L16-23. This seems to be largely a repeat of what is said above.

- 15 See changes made to relevant text in next response.

-L17 "significant improvements". In what way? Are more models are used, or are the source-receptor regions are better defined, or do the authors feel the parameterization itself has been improved in some fundamental way? The improved parameterization is again mentioned on page 9, line 19 (and probably elsewhere). Please be explicit on how exactly the parameterization is improved.

- 20 The underlying approach used in the parameterisation remains the same as Wild et al., (2012) but improvements have been made to the input and output. These improvements are described in the remaining lines of the paragraph. The text on P7 lines 16 to 22 have been replaced with the following to try and make the improvements clearer.

- "The original parameterisation developed by Wild et al., (2012) was based on the surface O₃ response to 20% continental-scale emission perturbations from TF-HTAP1 for 2001. We have adopted the same approach but have made a number of major improvements: updating the base year to 2010, included additional models from TF-HTAP2, extending the number of source regions to 14, and generating three-dimensional O₃ responses to permit calculation of tropospheric O₃ burden and O₃ radiative forcing for any scenario. To test and verify the improved parameterisation, additional simulations have been conducted with HadGEM2-ES, which are discussed in the following sections."*
- 25
30

-L31 "is working well". This is a little hard to tell from the figures. It would be valuable to show the percentage error as a function of month for the two responses.

- We have chosen to present absolute changes as this is the measure that matters most, and is also easiest for the reader to interpret. Percentage errors exaggerate differences where the underlying O₃ responses are very small.
35 Therefore we think it is best to focus on absolute errors (ppb) as that is discussed throughout the manuscript. For the reviewers convenience we have also computed the relative errors in the table below from the data used to plot Figure 2. The relative errors are largest, in transitioning from the winter titration regime to the spring/summer ozone production season. Generally errors are below 20% apart from a few select months (March, April, September and October).

Emission Reduction		Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
20%	Param	0.700	0.598	0.279	-0.192	-0.571	-0.743	-0.771	-0.600	-0.211	0.190	0.519	0.635
	HG	0.597	0.642	0.234	-0.071	-0.542	-0.734	-0.724	-0.570	-0.265	0.225	0.453	0.584
	Error	-0.103	0.045	-0.046	0.121	0.029	0.009	0.047	0.030	-0.054	0.035	-0.067	-0.051
	% err	-17%	7%	-20%	-169%	-5%	-1%	-7%	-5%	20%	16%	-15%	-9%
50%	Param	1.574	1.342	0.608	-0.553	-1.520	-1.964	-2.038	-1.601	-0.611	0.382	1.158	1.427
	HG	1.563	1.617	0.451	-0.396	-1.653	-2.124	-2.096	-1.689	-0.952	0.415	1.114	1.515
	Error	-0.011	0.275	-0.156	0.157	-0.133	-0.160	-0.058	-0.088	-0.341	0.033	-0.043	0.089
	% err	-1%	17%	-35%	-40%	8%	8%	3%	5%	36%	8%	-4%	6%
75%	Param	2.141	1.824	0.798	-0.922	-2.395	-3.080	-3.195	-2.528	-1.021	0.457	1.561	1.940
	HG	2.402	2.351	0.388	-1.011	-2.996	-3.673	-3.632	-2.987	-1.998	0.278	1.569	2.309
	Error	0.261	0.526	-0.410	-0.090	-0.601	-0.594	-0.438	-0.459	-0.977	-0.179	0.008	0.369
	% err	11%	22%	-106%	9%	20%	16%	12%	15%	49%	-64%	1%	16%

Page 8.

-The results over South Asia are really quite strange as the parameterization based on HadGEM2 fits the multi-model parameterization. The authors seem to be arguing on page 8 and 9 that this is a difficult region to simulate and that perhaps it is not surprising that the parameterization based on HadGEM with the large titration might not be able to simulate this region accurately. However, this seems to be only half of the story.... Why does the parameterization based specifically on HadGEM match the multi-model parameterization? And does the multi-model parameterization capture the multi-model response in this region?

The reviewer is correct to point out that the results over South Asia are different from that over other regions and highlights a particular limitation in the parameterised approach. The parameterisation is based on ozone response fields from 20% emission reduction experiments, but in the ECLIPSE CLE scenario there is a ~70% increase in NOx emissions over South Asia. The ozone responses generated by a 20% emission reduction show ozone reductions over clean, marine regions, but are close to zero over the continent (see Figure R1 below). Table 5 shows the parameterisation matches the TF-HTAP2 multi-model mean response well over South Asia for a 20% emission reduction.

For the 70% increase in NOx emissions over South Asia the parameterisation generates an increase in ozone over South Asia in January (Figure R2), scaled from the 20% emission change (Figure R1), whereas the HadGEM2-ES modelled response is a decrease in ozone (Figure R3). The simple scaling within the parameterisation is not able to simulate the shift in chemical environment over South Asia from production to loss that is associated with this large increase in emissions. This is because it is based on ozone distributions from only two model runs, and is thus not able to represent the strong non-linearities that may arise from large emission changes over regions which are close to maximum ozone production in current conditions. We note that the errors would be greater if we used a linear assumption. Future developments could attempt to address this problem by including additional simulations that would allow a full characterisation of the ozone response over a much wider range of emission changes.

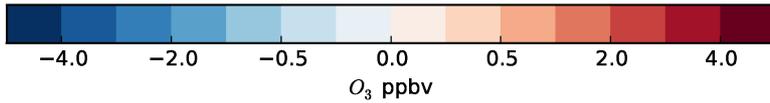
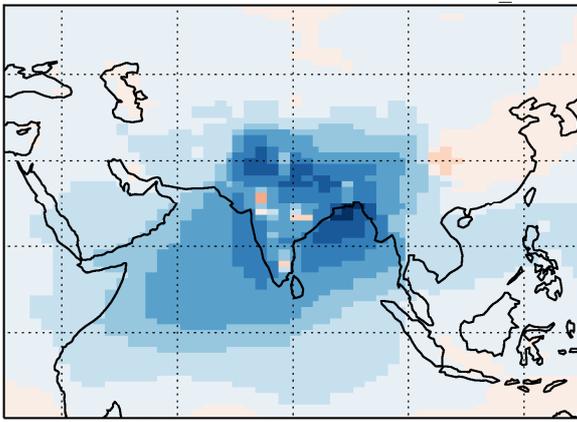
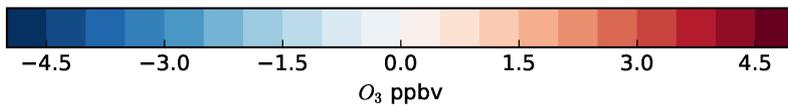
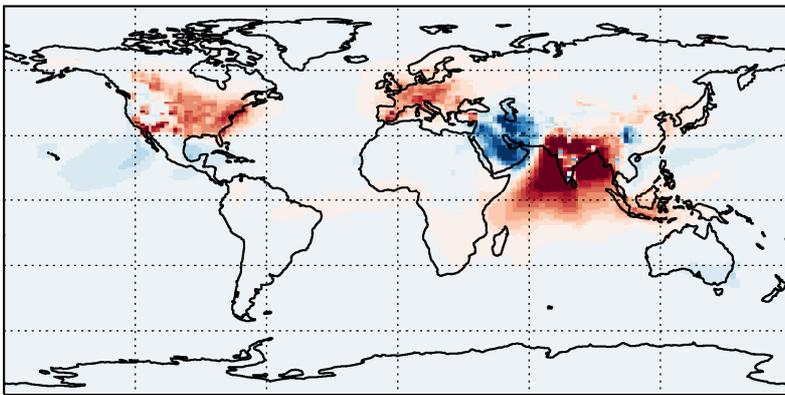


Figure R1 – January ozone response in HadGEM2-ES to a 20% emission reduction over South Asia of all anthropogenic precursor emissions (NO_x, CO, NMVOCs)



5 **Figure R2** – January ozone response from the parameterisation to the emission changes in the ECLIPSE CLE 2030 scenario, relative to 2010

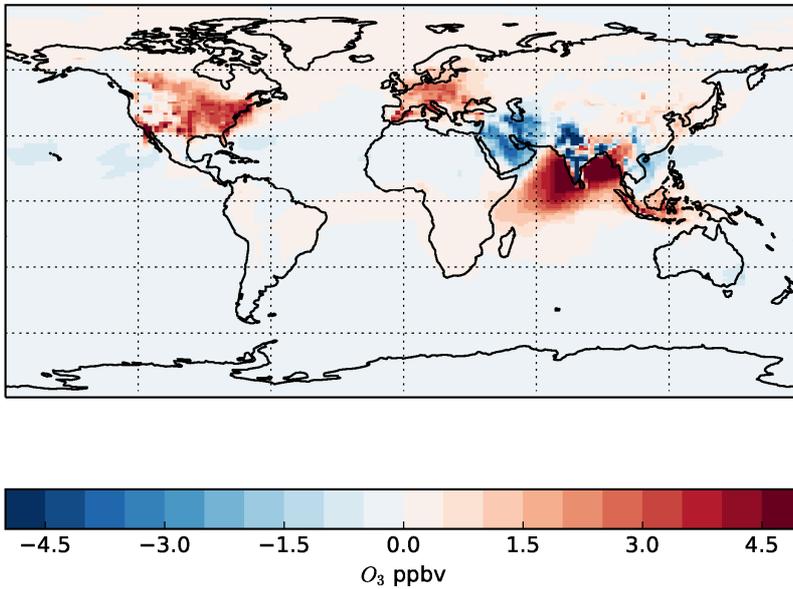


Figure R3 – January ozone response from HadGEM2-ES to the emission changes within the ECLIPSE CLE 2030 scenario, relative to 2010

The HadGEM2-ES simulation using emission changes from the ECLIPSE CLE scenario in 2030 was conducted to evaluate the parameterisation against a global emission perturbation and did not form part of the TF-HTAP2 set of experiments. No other model results are available using this scenario to identify if the results over South Asia are specific to HadGEM2-ES. Discussion of this occurs on Page 9, line 10.

The following sections of the text on P8 Lines 31 to 40 have been amended to improve the description of the result over South Asia, with the above figures included in the supplementary material:

10 *“The large increase in emissions causes the chemical environment in HadGEM2-ES in January to shift from O₃ production to that of titration (Figure S3). The parameterisation is not able to represent this shift (Figure S4) as it is based on a single ozone response to a 20% emission reduction (Figure S5) and is unable to capture the strongly non-linear transition into a net ozone titration regime. This is a smaller problem over North America, Europe or East Asia, as wintertime titration regimes are already present over these regions. This effect seen over South Asia*
 15 *highlights a weakness in the parameterised approach in representing strongly non-linear chemical regimes where there are large emission changes, although we note that the errors would be worse if a linear scaling was used.”*

Page 14

-L25-26: "compare well with ACCMIP multi-model means for intermediate emission scenarios..." What about RCP4.5? Isn't this a intermediate emission scenario? The comparison from RCP4.5 does not look that good.

20 See response to Point 2 above where a revised comparison with ACCMIP models is presented.

Response to Referee 2

The manuscript by Turnock et al. presents an updated version of earlier work done by Wild et al. (2012), who constructed a parameterisation for calculating the response of tropospheric ozone to changes in precursor emissions (and methane abundances). The parameterisation reduces the need to run an ensemble of computationally expensive global models of atmospheric chemistry in order to explore the effects of different emission scenarios on the abundance of tropospheric ozone, and the need to run multiple model experiments to determine the influences of emissions from multiple source regions on individual receptor regions. As such, this is a tool for rapid assessment of alternative emission control policies, but does not replace global atmospheric

chemistry models, since it does not represent the nonlinearities of tropospheric ozone chemistry, or the influence of future climate changes on tropospheric ozone. As an update to Wild et al. (2012), I expect that this paper will be widely used.

5 Unfortunately I found much of the description of the method to be vague and confusing. This should be improved before the paper is published. Also, the authors could do more to compare the predictions of their parameterised ozone with actual simulations from global atmospheric chemistry models. More details are given below:

The description of parameterisation has been improved by changes made to Section 2.1. Please see the response to point 1 for reviewer 1 above for the full text with the relevant sections included again in response to the specific points below.

10 Page 3, lines 5-7: Ignoring changes in future ozone due to climate change is an important limitation of this study. Here it would be appropriate to give a short summary of the expected changes in surface ozone due to climate change, to provide the reader with more information about this limitation.

The following text on the impact of climate change on surface ozone has been added on page 3 to the end of line 7:

15 *“Future climate change is expected to alter surface concentrations of ozone through changes to meteorological variables such as temperature, precipitation, water vapour, clouds, advection and mixing processes (Doherty et al., 2017).”*

Page 3, line 13: There appears to be a typo in the last sentence of this paragraph.

Removed extra O₃ from sentence so it becomes the following:

20 *“Section 5 uses the same future emission scenarios to predict future tropospheric O₃ burden and radiative forcing.”*

Page 4, line 5: Why is *f* replaced with *g*? Is this just a difference in terminology between Wild et al. (2012) and this study? Or something else?

25 As the parameterisation is based on that in Wild et al., (2012) the same terminology was used for consistency purposes. Wild et al., (2012) defined the linear scale factor as *f* for CO and NMVOC emission perturbations whereas the non-linear scale factor is represented by *g* for perturbations to NO_x emissions and CH₄ abundances. Equation 2 has been replaced with Equations 1 to 5, shown in the response to point 1 of reviewer 1, to make the notation clearer. The revised equation 4 is shown below (for more details see the response to point 1 in reviewer).

$$f_{ij} = 0.95r_{ij} + 0.05r_{ij}^2 \quad \text{Scaling accounting for reduced } O_3 \text{ increases from } NO_x \text{ and } CH_4 \quad (4)$$

P4, Line 4 - 6 has been replaced with the following:

30 *“To account for non-linear behaviour of surface O₃ to NO_x emission changes, a quadratic scaling factor (Eq. 4) is used, based on additional simulations of surface O₃ response over a larger range of emission perturbations in Wild et al., (2012).”*

Page 3, line 9: Where does "2f -g" come from? This appears to come out of nowhere.

35 This expression represents a reversal in curvature for NO_x emission reductions in titration regimes (where increased emissions lead to reduced ozone) and is described in more detail in Wild et al., 2012. However, we have revised the notation used here in response to the comments from both reviewers (see new Equations 1 to 5 in response to point 1 of reviewer 1). The revised equation 5 is shown below.

$$f_{ij} = 1.05r_{ij} - 0.05r_{ij}^2 \quad \text{Scaling for titration regimes where decreasing } NO_x \text{ increases } O_3 \quad (5)$$

P4 Lines 8 to 11 have been amended as follows:

“For the special case of source regions that are under titration regimes, where a reduction in NO_x emissions may lead to an increase in O₃, the curvature of the response is reversed for NO_x emission decreases (Eq. 5), as described in Wild et al., (2012). A linear scale factor is used for emission increases under these conditions (Eq. 3).”

- 5 Page 3, line 10: Much more explanation is needed here. Which model simulations? Which year? Why is the spatial extent of the titration regimes important? How is the magnitude defined? A lot of very important information appears to have simply been left out.

P4 Line 10 has been altered to reflect the fact that the parameterisation is based on the difference between two model simulations and is unable to calculate future changes to chemical regimes.

- 10 *“The spatial extent of ozone titration is assumed constant as the parameterisation is based on differences between two model simulations and is therefore unable to represent any future changes in chemical regime.”*

Page 3, line 14: It’s fascinating to read that the response of surface ozone to methane, a well-mixed gas with a lifetime measured in years, could be similar to the response to NO_x, which has a lifetime on the order of hours. In what way is the response "similar"? More explanation is needed here.

- 15 As we state in the manuscript, the non-linear behaviour of the ozone response to CH₄ is similar to that of NO_x emissions but not the magnitude of the response. The same quadratic function is therefore used within the parameterisation to scale O₃ responses from changes in CH₄ abundances and NO_x emissions. However, we appreciate the confusion here so have re-written line 14 to make this clearer.

- 20 *“The surface O₃ response to changes in global CH₄ abundance shows a similar degree of non-linearity as that from changes in NO_x emissions (Wild et al., 2012). Therefore, the non-linear scale factor (Eq. 4) is also used to represent the O₃ response to changes in CH₄ abundances.”*

Equation 2: How are the coefficients in the calculation of g determined?

The coefficients for g are those previously determined by additional simulations in (Wild et al., 2012). Page 4 Line 4 has been amended to the following (see point 1 for reviewer 1):

- 25 *“To account for non-linear behaviour of surface O₃ to NO_x emission changes, a quadratic scaling factor (Eq. 4) is used, based on additional simulations of surface O₃ response over a larger range of emission perturbations in Wild et al., (2012).”*

Page 8, line 40: There is no need to mention titration a second time.

- 30 We state here that the ozone response over South Asia from HadGEM2-ES simulations could be due to a shift to an ozone titration regime in the model which is not represented in the parameterisation, and thus explains some of the discrepancy between them. The important part here is the shift in regime, which has not been discussed previously and we feel is important to keep in the manuscript.

- 35 Section 3.3.1: Can you compare the predictions of the parameterisation using the RCP scenarios with the actual global model runs done in the ACCMIP exercise? See Young et al. (2013) for some examples. This comparison would really help the reader to understand more about how well the parameterisation is doing in comparison with the global models. Perhaps this comparison could be added to Fig. 5. Also, why isn’t this part of Section 4? ("Future Surface Ozone Predictions")

The comparison with the RCPs provides an evaluation of future predictions of the parameterisation as ozone projections for these scenarios have already been made previously using the parameterisation of Wild et al.,

(2012). The parameterised predictions of global surface ozone, global tropospheric burden and ozone radiative forcing in 2030 from each of the RCPs have now been compared to that from the ACCMIP models in the new version of Table 9 (see response to point 2 of reviewer 1). However, it should be noted that changes to ozone concentrations in the ACCMIP models will have additional contributions from climate change and stratospheric ozone recovery, which are not represented in the parameterisation.

Table 9 has been amended as described in the response to point 2 for reviewer 1 above (Table 10 in revised manuscript) and now includes a comparison to surface ozone from the ACCMIP models.

The following text has been included to discuss the comparison to ACCMIP models (see response to point 2 for reviewer 1 for more details).

10 P13 Lines 22 to 30 have been amended to:

15 *“In comparison to the ACCMIP multi-model mean, the predicted changes between 2000 to 2030 in both global annual mean surface O₃ and global O₃ burden from the parameterisation are within the range of the ACCMIP multi model responses (+/- 1 standard deviation) for all the CMIP5 RCPs (Table 10). The predictions of O₃ radiative forcing in 2030 from the parameterisation across all the RCPs, when the influence of climate change is anticipated to be small, are also consistent with those from ACCMIP. The sign and magnitude of change in global O₃ burden and O₃ radiative forcing with the parameterisation for RCP6.0 is different from the ACCMIP results but is still within the range of model responses, which is the largest for this scenario. The comparison with ACCMIP results shows that the parameterisation is able to reproduce changes in global O₃ burden and O₃ radiative forcing on near-term timescales, when the influence of climate change is small.”*

20 Page 10, line 8: "successive emission increases" of what magnitude? In what succession?

Each additional increment in emissions (an amount corresponding to 20% of current emissions) gives a 10% smaller ozone response than the previous one. This reflects the nonlinearity described by the quadratic expression that is given. Line 8 has been re-worded to the following:

25 *“For simplicity the parameterisation used the same non-linear scaling factor as for NO_x emissions (Eq. 4 i.e. $g = 0.95f + 0.05f^2$), which represents a 10% smaller response for successive 20% emission increases.”*

Equation 3: The coefficients appear to have changed since Equation 2. Why? How are they calculated? This seems like something for Section 2.

30 This section uses results from the models in TF-HTAP2 that conducted methane perturbation experiments to see if the coefficients derived for the quadratic function in Wild et al., (2012) are still valid (P10, line 8). This used the same method as in Wild et al., (2012) to derive the coefficients for the TF-HTAP2 models, shown in equation 3. Since equation 3 and 2 are similar and within the level of uncertainty it was decided for consistency purposes to retain the same coefficients of the quadratic function used in Wild et al., (2012) (stated on P10 Lines 11 to 12). As this was a comparison of TF-HTAP2 results to TF-HTAP1 and evaluation of the existing parameterisation it was decided to put this discussion into Section 3 – Testing and Validation. To avoid further confusion we have removed equation 3 from the manuscript.

35 ~~$$g = 0.937f + 0.063f^2 \quad (3)$$~~

P10 Line 11 has been amended as follows to reflect this change:

“We find a slightly larger sensitivity, with both models yielding a 12.6% smaller surface O₃ response for an increase in CH₄ than a decrease (Eq. 3).”

40 Reference to equation 3 is also removed from P10 L17-19:

5 “To enable a direct comparison with TF-HTAP1 results, the O₃ response from the CH4DEC and CH4INC experiments in TF-HTAP2 are scaled to represent the response from a 20% reduction in CH₄ abundances, using Eq. 3. An adjustment factor is calculated based on the global mean difference between the TF-HTAP2 O₃ response in each experiment and that of an equivalent 20% reduction in CH₄ abundance (calculated using Eq. 3), resulting in a factor of 1.557 for CH4DEC and -1.256 for CH4INC.”

Page 10, line 33: What are the "appropriate fields"?

P10, line 33 have been modified to include the appropriate fields.

“Table 8 summarises the calculated CH₄ lifetime and feedback factors for the two TF-HTAP2 models that provided CH₄ chemical loss rates.”

10 Section 4.1: Can you compare the predictions of the parameterisation using the ECLIPSE scenarios with the actual global model runs done in the ECLIPSE exercise? See Stohl et al. (2015, DOI: 10.5194/acp-15-10529-2015) and work citing that paper for examples.

15 A different version of the ECLIPSE emissions inventory has been used as input to the models for the ECLIPSE project (Version 4 and 5) than was used with the parameterisation here (Version 5a). Different future emissions pathways exist in these versions of the emission inventory and this hinders a direct comparison between the models and the parameterisation. Therefore, we think it is not feasible to provide a direct comparison between the ECLIPSE models and the parameterisation here. Whereas output from the ACCMIP models was available for the same set of CMIP5 future RCPs, making a direct comparison between these models and the parameterisation more appropriate.

20 However, the percentage change difference in surface ozone concentrations between the mitigation and current legislation scenario in 2050 is presented in Table 4 of Stohl et al., (2015). This shows a multi-model reduction of surface ozone concentrations over Europe, China, India and the United States of between 13 to 20% for the mitigation scenario. Whilst not directly comparable to our study in terms of scenarios used or receptor regions we calculate a similar reduction in future surface ozone concentrations of approximately 20% over Europe, North
25 America and East Asia for the ECLIPSE MTRF scenario in 2050 (relative to CLE) and ~30% for South Asia.

Response to Short Comment by R. Van Dingenen

In general this is a well-written and very useful paper that addresses relevant policy issues.

30 As a possible user of the ozone precursor source-receptor relations, I would like to make some suggestions that would improve the readability of the paper and create the possibility for the scientific community to replicate the results.

Eq. 1: the same variable symbol (ΔO_3) is used at left and right-hand side of the equation, while they have different meanings. The same observation can be made for Eq. 2 where e.g is written $f_{ij} = 2f_{ij} - g_{ij}$; suggest to use a different symbol at the left hand side.

35 We appreciate the possible confusion here, and have adjusted equation 1 by adding the subscripts e (for emissions) and m (for methane) to the ΔO_3 terms on the right-hand side to distinguish the ΔO_3 terms from each other. We have left the overall form of the equation as it is, for consistency with Wild et al., (2012). We have revised the notation in Eq.2 to clarify the expressions in response to the comments of the reviewers (see the response to point 1 for reviewer 1).

40 Eq. 1 expresses ΔO_3 as response to the sum of an emission change (for NO_x, CO and NMVOCs), and an abundance change in CH₄. For the user, using emission changes for all precursors would make more sense. Isn't

it possible, from the box model mentioned in section 3.2, and using a feedback factor, to relate a change in abundance to a change in emissions? Why not normalise the source-receptor responses by the emission strength? It would be useful to emphasise the time scale of the CH₄ responses and how to deal with this in such a parametrised approach.

5 The parameterisation is based on simulations from HTAP1 and HTAP2 that used a change in global methane abundance to simulate an O₃ response and so the parameterisation is based on methane abundance. It would be possible to extract a CH₄ emission response based on the prescribed CH₄ abundance change (e.g. Meinshausen et al., 2011; Holmes et al., 2013), but this would be different for each model, and including this adds an additional layer of complexity to the parameterisation.

10 Future developments could include some sort of emissions to abundance conversion as a post processing step.

It's not clear why paragraph 3.1 is named 'Scaling Factors'

Change section title to "*Limits of Linear Scaling*".

Page 10, line 12: 'the same scaling factor', is not clear if 'same' refers to using the same as in HTAP1, or using the same (new) factor for CH₄ and NO_x. So, Eq. 3: is this now the scaling factor replacing the $0.95f+0.05f^2$ from HTAP1 both for NO_x and CH₄?

Changed Line 12 to:

"... same representation of non-linearity for both NO_x and CH₄ (Eq. 4), as used in Wild et al., (2012)"

Figures 7 and 8 (and similar in SI): does the ozone trend from CH₄ include the transient effect of the 12y perturbation response time? How can Eq. 3 be applied (for CH₄) to obtain this trend? The figures show the change in ozone relative to year 2010; does it include the time-lagged impact of CH₄ emissions before that date? I would appreciate having the box model for CH₄ better documented.

The contribution to the ozone trend from changes in methane reflects the effects of the change in methane abundance alone, and is the equilibrium response (short and long term). No transient effects are considered in the parameterisation, and this is another argument for the simplicity of basing it on abundance rather than emissions. The abundance is calculated using the methods and expressions given in Meinshausen et al., (2011) and Holmes et al., (2013) and no new aspects have been introduced for the purposes of this study.

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The Impact of Future Emission Policies on Tropospheric Ozone using a Parameterised Approach

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20 **Abstract.**

This study quantifies future changes in tropospheric ozone (O₃) using a simple parameterisation of source-receptor relationships based on simulations from a range of models participating in the Task Force on Hemispheric Transport of Air Pollutants (TF-HTAP) experiments. Surface and tropospheric O₃ changes are calculated globally and across 16 regions from perturbations in precursor emissions (NO_x, CO, VOCs) and methane (CH₄) abundance only, neglecting any impact from

25 climate change. A source attribution is provided for each source region along with an estimate of uncertainty based on the spread of the results from the models. Tests against model simulations using HadGEM2-ES confirm that the approaches used within the parameterisation are valid, perform well for most regions. The O₃ response to changes in CH₄ abundance is slightly larger in TF-HTAP Phase 2 than in the TF-HTAP Phase 1 assessment (2010) and provides further evidence that controlling CH₄ is important for limiting future O₃ concentrations. Different treatments of chemistry and meteorology in models remains

30 one of the largest uncertainties in calculating the O₃ response to perturbations in CH₄ abundance and precursor emissions, particularly over the Middle East and South Asian regions. Emission changes for the future ECLIPSE scenarios and a subset of preliminary Shared Socio-economic Pathways (SSPs) indicate that surface O₃ concentrations will increase regionally by 1 to 8 ppbv in 2050 across different regions. Source attribution analysis highlights the growing importance of CH₄ in the future under current legislation. A change in the global tropospheric O₃ radiative forcing of +0.07 W m⁻² from 2010 to 2050 is

35 predicted using the ECLIPSE scenarios and SSPs, based solely on changes in CH₄ abundance and tropospheric O₃ precursor emissions and neglecting any influence of climate change. Current legislation is shown to be inadequate in limiting the future degradation of surface ozone air quality and enhancement of near-term climate warming. More stringent future emission controls provide a large reduction in both surface O₃ concentrations and O₃ radiative forcing. The parameterisation provides a simple tool to highlight the different impacts and associated uncertainties of local and hemispheric emission control strategies

40 on both surface air quality and the near-term climate forcing by tropospheric O₃.

1 Introduction

Tropospheric ozone (O₃) is an air pollutant at both regional and global scales. It is harmful to human health (Brunekreef and Holgate, 2002; Jerrett et al., 2009; Turner et al., 2016; Malley et al., 2017), whilst also affecting climate (Myhre et al., 2013) and causing damage to natural and managed ecosystems (Fowler et al., 2009; United Nations Economic Commission for Europe (UNECE), 2016). Long-range transport of air pollutants and their precursors can degrade air quality at locations remote from their source region (Fiore et al., 2009). Predicting source-receptor relationships for O₃ is complex due to large natural background sources, formation of O₃ from local emissions, non-linear chemistry and inter-continental transport processes (TF-HTAP, 2010). In particular, it is uncertain how the interaction of local and regional emission controls with global changes (e.g. of methane and climate) could affect O₃ concentrations in the near-term future (2050s) (Jacob and Winner, 2009; Fiore et al., 2012; von Schneidemesser et al., 2015). This is evident from the wide range of modelled O₃ responses in future emission and climate scenarios (Kawase et al., 2011; Young et al., 2013; Kim et al., 2015). The setting and achieving of effective future emission control policies is therefore difficult, as a substantial proportion of O₃ comes from outside individual countries and regions.

Phase 1 of the Task Force on Hemispheric Transport of Air Pollutants (TF-HTAP1) (TF-HTAP, 2010) coordinated several sets of experiments using multiple models to study the source-receptor relationships from the intercontinental transport of O₃ and its precursors. It found that at least 30% of the total change in surface ozone concentration within a particular source region can be attributed to emission changes of similar magnitude that are external to the source region (TF-HTAP, 2010). This highlights the importance of source contributions outside the control of local/regional air pollutant policies, including those of stratospheric origin, natural sources and intercontinental transport. Changes in global methane (CH₄) concentrations are also an important contributor to baseline O₃ concentrations and are shown to be as important as changes in local source region emissions (TF-HTAP, 2010). Improving our understanding of the impact of anthropogenic emission changes on the source-receptor relationships arising from the intercontinental transport of tropospheric O₃ and its precursors will ultimately reduce the uncertainty in the impact of O₃ on air quality and climate, improving future predictions.

To predict how O₃ concentrations might respond to future changes in emissions, a simple parameterisation was developed based upon the surface O₃ response in different chemistry models contributing to TF-HTAP1 Wild et al., (2012). The surface O₃ response in these models was calculated from simulations with reductions in tropospheric O₃ precursor emissions across the four major northern hemisphere emission regions (Europe, North America, East Asia, and South Asia). The parameterisation using these results provided a fast and simple tool to predict future surface O₃ concentrations for the Intergovernmental Panel on Climate Change (IPCC) Representative Concentration Pathways (RCPs), highlighting the importance of future changes in emissions and CH₄ abundance for surface O₃ concentrations and quantifying the associated uncertainty.

A second phase of model experiments, TF-HTAP2, was initiated to extend the work from TF-HTAP1 and further consider the source-receptor relationships between regional emission reductions and air pollutants. Major advances in TF-HTAP2 include more policy-relevant source-receptor regions aligned to geo-political borders, a larger variety of idealised 20% emission reduction experiments, more recent (2008-2010) emission inventories that are consistent across all models and the use of new and updated models (Galmarini et al., 2017).

Here we improve and extend the parameterisation of Wild et al., (2012) by including additional information from TF-HTAP2 to refine the source-receptor relationships arising from emission changes, long range transport and surface O₃ formation. The parameterisation provides the contribution from local, remote and methane sources to the total surface O₃ response in each

emission scenario. The range of responses from the models contributing to the parameterisation provides an estimate of the uncertainty involved. The parameterisation is extended to estimate changes in tropospheric O₃ burden and its impact on O₃ radiative forcing. It is then used with the latest emission scenarios from ECLIPSE V5a (Klimont *et al.*, 2017; Klimont *et al.*, *in prep.*) and the 6th Coupled Model Intercomparison Project (CMIP6) (Rao *et al.*, 2017) to explore how source-receptor relationships change in the future, informing the future direction of emission control policies. These predictions of changes in surface and tropospheric O₃ are based solely on changes in precursor emissions, as the parameterisation does not represent any impact from future changes in climate. Future climate change is expected to alter surface concentrations of ozone through changes to meteorological variables such as temperature, precipitation, water vapour, clouds, advection and mixing processes (Doherty *et al.*, 2017).

Section 2 of this paper describes the parameterisation and the updates from TF-HTAP1 to TF-HTAP2, including the extension from surface O₃ to global tropospheric O₃ and its radiative forcing. Section 3 outlines the testing and validation of the parameterisation. A comparison is made to results from TF-HTAP1, highlighting changes in the O₃ response to changes in methane abundance. In section 4, the parameterisation is applied to the ECLIPSE V5a and CMIP6 emission scenarios to predict future surface O₃ concentrations over the period 2010 to 2050. Section 5-O₃ uses the same future emission scenarios to predict future tropospheric O₃ burden and radiative forcing. We conclude by suggesting how this approach could be used to inform future emission policy in relation to O₃ concentrations.

2. Methods

2.1 ~~Original Ozone~~ Parameterisation of Ozone

The parameterisation developed in this study is based on an earlier version developed ~~for~~ the TF-HTAP1 experiments by Wild *et al.*, (2012). This simple parameterisation enabled the regional response in surface O₃ concentrations to be estimated based on changes in precursor emissions and CH₄ abundance. The input for this parameterisation came from 14 different models that contributed to TF-HTAP1. All the models ran the same emission perturbation experiments (20% reduction in emissions of oxides of nitrogen (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs) individually and all together) over the four major northern hemisphere source regions of Europe, North America, East Asia and South Asia. Additional experiments included global perturbations of emission precursors (*E*), as well as a 20% reduction in global CH₄ abundance. The multi-model O₃ responses from the 20% emission perturbation experiments (ΔO_{3e} for emissions of NO_x, CO and NMVOCs and ΔO_{3m} for CH₄) are then scaled by the fractional emission changes (*r*) from a given emission scenario over each source region (Eq. 1).

$$r_{ij} = \frac{\Delta E_{ij}}{-0.2 \times E_{ij}} \quad (1)$$

The monthly mean O₃ response (ΔO₃) is calculated as the sum over each receptor region (*k*) of the scaled O₃ response from each model to the individual precursor species (*i* - CO, NO_x and NMVOCs) in each of the five source regions (*j* - Europe, North America, East Asia, South Asia and rest of the world), including the response from the change in global CH₄ abundance (Eq. ~~42~~, reproduced from Wild *et al.*, 2012):

$$\Delta O_3(k) = \sum_{i=1}^3 \sum_{j=1}^5 f_{ij} \Delta O_{3e}(i, j, k) + f_m \Delta O_{3m}(k) \quad (42)$$

$$f_{ij} = r_{ij} \quad \text{Linear Scaling of O}_3 \text{ response} \quad (3)$$

$$f_{ij} = 0.95r_{ij} + 0.05r_{ij}^2 \quad \text{Scaling accounting for reduced O}_3 \text{ increases from NO}_x \text{ and CH}_4 \quad (4)$$

$$f_{ij} = 1.05r_{ij} - 0.05r_{ij}^2 \quad \text{Scaling for titration regimes where decreasing } NO_x \text{ increases } O_3 \text{ (5)}$$

A linear ~~The~~ emission scale factor (Eq. 3) ~~f_{ij}~~ is used in Eq. 2 for each emission scenario involving the precursor emissions CO and NMVOCs and is defined as the ratio of the fractional emission change ($\Delta E_{ij}/0.2 \times E_{ij}$) to the 20% emission reduction in the TF-HTAP1 simulations (Eq. 2). A similar scale factor for methane (f_m) is based on the ratio of the change in the global abundance of CH₄ to that from the 20% reduced CH₄ simulation ($\Delta[CH_4]/-0.2 \times [CH_4]$). Perturbations to emissions of CO, NO_x and NMVOCs induce a long-term (decadal) change in tropospheric O₃ from the change in the oxidising capacity of the atmosphere (OH) and the CH₄ lifetime (Wild and Akimoto, 2001; Collins et al., 2002; Stevenson et al., 2004). The long-term impacts from 20% global emission reductions can reduce the O₃ response by 6-14% from NO_x emission changes and increase the O₃ response by 16-21% from CO changes (West et al., 2007). This long-term response is not accounted for in the simulations used here as CH₄ abundances are fixed.

Wild et al., (2012) found that this simple linear scaling relationship between emissions and surface O₃ was sufficient for small emissions perturbations, but that the relationship started to exhibit larger non-linear behaviour for larger perturbations, particularly for NO_x. The linear scaling factor ~~was found to be~~ sufficient for the surface O₃ response from emission perturbations of CO and NMVOCs as non-linear behaviour from these precursors is small (Wu et al., (2009). To account for non-linear behaviour of surface O₃ to NO_x emission changes, ~~the a quadratic scaling factor, f , is replaced with g (Eq. 4) is used, which has a quadratic dependency on f (Eq. 2), and is~~ based on additional simulations of surface O₃ response over a larger range of emission perturbations undertaken in Wild et al., (2012).

For the special case of source regions that are under titration regimes, where a reduction in NO_x emissions may lead to an increase in O₃, ~~the surface O₃ the curvature of the response is reversed for NO_x emission decreases (Eq. 5), as described in Wild et al., (2012). A linear scale factor is used for emission increases under these conditions (Eq. 3) is limited (by a factor of $2f - g$ for emission reductions and for emission increases the linear scaling factor is used (Eq. 2). The spatial extent and magnitude of titration regimes is assumed constant as it is based on model simulations from a single meteorological year. The spatial extent of ozone titration is assumed constant as the parameterisation is based on differences between two model simulations and is therefore unable to represent any future changes in chemical regime.~~

The surface O₃ response to changes in global CH₄ abundances ~~is also shows a similar degree of non-linearity and showed similar behaviour to~~ as that ~~due to~~ from changes in NO_x emissions (Wild et al., 2012). Therefore, the non-linear scale factor (g in Eq. 4) is also used to represent the O₃ response to ~~increases and decreases~~ changes in CH₄ abundances.

In summary, the surface O₃ response to CO and NMVOC emission perturbations is represented by the linear scale factor (Eq. 3) f_{ij} and to changes in NO_x emissions and CH₄ abundances by the non-linear scale factor (Eq. 4) g_{ij} (Eq. 2). For source regions under titration regimes, the surface O₃ response to NO_x emissions is limited by Eq. 5 for emission decreases but uses the linear scale factor (Eq. 3) for emission increases. The parameterisation is represented schematically in Figure S1 of the supplementary material. ~~The representation of the surface O₃ response to NO_x emissions is determined by the conditions specified in Eq. 2.~~

$$f_{ij} = \begin{cases} f_{ij} = \Delta E_{ij}/0.2 \times E_{ij} & \text{if } \Delta O_3(i, j, k) > 0 \text{ and } \Delta E_{ij} > 0 \\ \frac{2f_{ij} - g_{ij}}{g_{ij}} & \text{if } \Delta O_3(i, j, k) > 0 \text{ and } \Delta E_{ij} < 0 \\ g_{ij} & \text{otherwise (where } g = 0.95f + 0.05f^2) \end{cases} \quad (2)$$

2.2 Phase 2 of TF-HTAP

A second phase of simulations has been undertaken as part of TF-HTAP to further study the transport of air pollutants and their impacts and to assess potential mitigation options (Galmarini et al., 2017). Phase 2 (TF-HTAP2) involved experiments using new and/or updated models that conducted idealised 20% perturbation simulations of O₃ emission precursors for different source regions and source sectors over the years 2008 to 2010. A 20% emission perturbation was chosen to generate a sizeable response, whilst still being small enough to minimise non-linear chemistry effects. To determine the O₃ response to CH₄ changes, simulations increasing methane to 2121 ppbv (18%) and decreasing to 1562 ppbv (-13%) from a baseline of 1798 ppbv were undertaken in TF-HTAP-2. This range in CH₄ abundances was selected to encompass the uncertainty in CH₄ changes in 2030 from the 5th Coupled Model Intercomparison Project (CMIP5) scenarios of RCP8.5 and RCP2.6 (Galmarini et al., 2017).

The source ~~and receptor~~ regions were updated in TF-HTAP2 to represent 164 new ~~receptor~~ regions (14 of which excluding the North and South Poles are also sources), aligned on geo-political and land/sea boundaries (Figure 1). Emission inventories (consistent across all models (Janssens-Maenhout et al., 2015)) and meteorology (driving data specific to individual models) were updated to consider the years 2008 to 2010 (the focus of TF-HTAP1 was 2001). The Global Fire Emission Database version 3 (GFED3 – <http://globalfiredata.org/>) biomass burning (grassland and forest fires) emissions were recommended for TF-HTAP2 experiments, although some models selected other inventories. Individual modelling groups used their own information for other natural emission sources (e.g. biogenic VOCs, lightning NO_x), as many of these are based on internal model calculations and not externally prescribed datasets.

Priority in TF-HTAP-2 was placed on conducting a baseline simulation, a simulation with increased CH₄ concentrations and seven regional simulations involving 20% reductions of all precursor emissions across the globe, North America, Europe, East Asia, South Asia, Russia Belarus and Ukraine and the Middle East in the year 2010. A lower priority was given to emission perturbation experiments across the remaining source regions and experiments with individual emission perturbations and perturbations to individual source sectors. Models ~~conducted~~ ~~vered~~ a consistent core set of 10-20 simulations and then ~~undertook~~ ~~conducted~~ other experiments of their own choosing (see Galmarini et al., (2017) for a full list of models and experiments), resulting in sparse coverage for many of the experiments. This contrasts with TF-HTAP1 where all models conducted the same set of 20% emission perturbation experiments covering all precursor emissions (individually and combined) and CH₄ across four source regions.

30 2.3 Improvements to the surface Ozone Parametric Model for TF-HTAP2

Differences in the experimental setup in TF-HTAP1 and TF-HTAP2 means that it is not straight forward to replace the simulations underpinning the parameterisation of Wild et al., (2012) with those from TF-HTAP2. The larger number of simulations and fewer models involved preclude the development of a robust parameterisation based solely on TF-HTAP2 simulations. We therefore extend the existing parameterisation by including additional information from the new simulations in TF-HTAP2. To maintain a robust response over the major source regions of Europe, North America, East Asia and South Asia, results from the 14 models contributing to TF-HTAP1 over these regions were retained in the parameterisation. Results from the models contributing to TF-HTAP2 were then incorporated, accounting for the different baseline year for emissions (2010 rather than 2001) and the change in size and number of source ~~receptor~~ regions. The following sections discuss in detail how the results from TF-HTAP2 have been incorporated into the parameterisation.

2.3.1 New Baseline Year

The baseline year used in the parameterisation was first adjusted from 2001 (TF-HTAP1) to 2010 (TF-HTAP2), to reflect changes in anthropogenic emissions between these years. It should be noted that the emission inventories used in TF-HTAP1 were not consistent between models, particularly for NMVOCs, and this partially contributed to the different O₃ responses (Fiore et al., 2009). In TF-HTAP2, the same anthropogenic emission inventory was used in all models to prevent uncertainty in anthropogenic emissions dominating the variability across models.

The parameterisation of Wild et al., (2012) was used to calculate new baseline O₃ concentrations in 2010 for use in this version of the improved parameterisation and for comparison to the TF-HTAP2 multi-model mean. To account for different CH₄ abundances the change between TF-HTAP1 and TF-HTAP2 was used. The mean fractional change in NO_x, CO and NMVOC emissions between 2000 and 2010 across the TF-HTAP1 source regions from two different emission inventories, (MACCity (Granier et al., 2011) and EDGARv4.3.1 (Crippa et al., 2016)) was used (Table 1) due to the inconsistencies, as the use of the specific emissions inventories was not prescribed for the TF-HTAP1 experiments (see Fiore et al., 2009) and TF-HTAP2 (Table 4). The MACCity and EDGAR inventories are internally provide a consistent set of emissions in 2000 and 2010, enabling the changes in emissions for both between future and historical time periods to be explored. Table 1 shows that the emissions of NO_x, CO and NMVOC increased in Asia and decreased across Europe and North America over the period 2000 to 2010. Year 2000 was used as a consistent starting point for both emission inventories and can be considered equivalent of 2001 in representing changes to 2010.

The parameterised surface ozone response in 2010 was calculated using the method of Wild et al., (2012), based on the individual response of 14 TF-HTAP1 models using the fractional emission changes in Table 1. The parameterised ozone response across ~~The parameterised surface ozone response in 2010 across~~ the original TF-HTAP1 source/receptor regions was compared to a multi-model ozone concentration in 2010 from the baseline simulations of seven TF-HTAP2 models that use the TF-HTAP2 emissions (Janssens-Maenhout et al., 2015). Table 2 shows that the O₃ concentrations from the parameterisation (H-P) are similar to the within the spread of the individual model values from TF-HTAP2 multi-model mean values (H-2), represented by one standard deviation, over most of the receptor regions, ~~and within the spread of individual model values (represented by one standard deviation).~~ The large range and standard deviation in Table 2 highlights the large spread in O₃ concentrations over the models in both sets of experiments (H-1 and H-2). The range in O₃ concentrations is much larger than the differences between the parameterised values and the TF-HTAP2 multi-model mean in 2010. This indicates that the range of responses over the models dominates the uncertainty in O₃ concentrations and is much greater than differences due to the subset of models contributing to each study or from changing emissions over the period 2000 to 2010.

2.3.2 Source Region Adjustment

The original parameterisation was based on the continental-scale emission source regions defined in TF-HTAP1. To continue using these results in an improved parameterisation, the O₃ response fields were adjusted to represent the equivalent source regions in TF-HTAP2. The different regional definitions used within TF-HTAP1 and TF-HTAP2 experiments are shown in Figure 1 and are particularly large for Europe, where the TF-HTAP1 source region covers parts of five TF-HTAP2 source regions (Europe, Ocean, North Africa, Middle East and Russia Belarus and Ukraine). O₃ response fields from TF-HTAP1 models that formed the basis of the original parameterisation were adjusted to be more representative of the equivalent TF-HTAP2 source region.

No single model contributed experiments in both TF-HTAP1 and TF-HTAP2 to inform the adjustment of source regions. Therefore, 20% emission perturbation simulations were conducted with HadGEM2-ES (Collins et al., 2011; Martin et al.,

2011), which contributed to TF-HTAP2 experiments, for the TF-HTAP1 source regions of Europe, North America, East Asia and South Asia. The ratio of the O₃ responses between the simulations using TF-HTAP1 and TF-HTAP2 source regions was then applied to the O₃ response fields from each of the TF-HTAP1 models used in the parameterisation of Wild et al., (2012). We assume that each model behaves in a similar way as HadGEM2-ES when the source regions are adjusted in this way. This generates an O₃ response field from emission perturbations within the equivalent TF-HTAP2 source regions of Europe, North America, East Asia and South Asia. The resulting O₃ parameterisation is based on a larger number of models (14 adjusted TF-HTAP1 models) than would have been available from using TF-HTAP2 simulations alone (7 TF-HTAP2 models), allowing for a larger diversity of model responses to represent the four major emission source regions.

2.3.3 Additions from TF-HTAP2

The O₃ responses from emission perturbations for the other ten TF-HTAP2 source regions were then used to augment the source region adjusted O₃ response fields from TF-HTAP-1. This extends the parameterisation to cover a much larger range of source regions (14 in total) than was previously possible. Here, receptor regions are defined in accordance with those in TF-HTAP2 (16 in total), although it is possible to define any required receptor regions using the global distribution of O₃ responses. Table 3 lists the number of model simulations available for the TF-HTAP2 source regions over and above the four main source regions of Europe, North America, South Asia and East Asia, highlighting the sparseness of results for some of the TF-HTAP2 regions.

The monthly O₃ response fields from the additional ten TF-HTAP2 emission source regions were converted onto the same standard grid (1° x 1° in the horizontal, with 21 vertical levels based on regular pressure intervals from the surface at 1000 hPa to an upper level of 10 hPa) as used for the four source regions from the adjusted TF-HTAP1 models. In addition, the fields from the TF-HTAP1 models are based on the O₃ response to the individual emissions perturbations of NO_x, CO and NMVOCs, whereas the regional emission perturbation simulations for TF-HTAP2 are based on all emission precursors together (due to the limited availability of results from regional individual precursor emission simulations in TF-HTAP2). To maintain consistency with the TF-HTAP1 parameterisation, the O₃ response for each TF-HTAP2 emission perturbation simulation is divided up to represent the response from individual emission precursors, as Wild et al., (2012) and Fiore et al., (2009) previously showed that O₃ responses from individual emission perturbations matched closely to that from combined emissions changes (within 2-7%). Therefore, the fractional contribution from individual emissions to the total O₃ response in the multi-model mean of TF-HTAP1 models is used to apportion the contribution from individual emissions in TF-HTAP2 simulations to the total O₃ response.

The CH₄ perturbation experiments in TF-HTAP2 were based on global changes of -13% and +18% to reflect the expected atmospheric abundance in RCP2.6 and RCP8.5, respectively. These were adjusted to the 20% reduction used in TF-HTAP1 using the parameterisation, allowing O₃ responses to CH₄ from the original 14 TF-HTAP1 models and the five TF-HTAP2 models that provided sufficient results to be combined.

2.3.4 Extension to Tropospheric Ozone

The parameterisation has been extended from the surface through the depth of the troposphere, enabling the calculation of the tropospheric O₃ burden. The three-dimensional monthly O₃ fields from the model simulations are interpolated onto 21 vertical levels at regularly spaced mid-level pressure intervals from 1000 hPa to 10 hPa. These O₃ fields were then used with the parameterisation to generate global and regional tropospheric O₃ burdens for each scenario, with the tropopause defined as an O₃ concentration of 150 ppbv (Prather et al., 2001). An O₃ radiative forcing is derived by using the tropospheric O₃ burden from the parameterisation and the relationship between radiative forcing and tropospheric column O₃ change based on multi-

model ensemble mean results from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Stevenson et al., 2013). This relationship is provided as a two-dimensional global map, enabling regional and global O₃ radiative forcing to be calculated from the parameterisation.

3.0 Testing and Validation

5 The original parameterisation developed by Wild et al., (2012) was based on the surface O₃ response to 20% ~~continental-~~
~~scaleregional~~ emission perturbations from TF-HTAP1 for 2001. ~~We have updated this to reflect conditions in 2010, and have~~
~~made significant improvements based on results from TF-HTAP2. The parameterisation has been extended to include the 14~~
~~sources regions in TF-HTAP2. Output is provided on a standard grid to facilitate the calculation of O₃ responses over any~~
~~selected receptor regions. The parameterisation has been extended to generate three dimensional O₃ fields that permit the~~
10 ~~calculation of tropospheric O₃ burden and O₃ radiative forcing for any scenario. We have adopted the same approach but have~~
~~made a number of major improvements: updating the base year to 2010, included additional models from TF-HTAP2,~~
~~extending the number of source regions to 14, and generating three-dimensional O₃ responses to permit calculation of~~
~~tropospheric O₃ burden and O₃ radiative forcing for any scenario.~~ To test and verify the improved parameterisation, additional
simulations have been conducted with HadGEM2-ES, which are discussed in the following sections.

15 3.1 Sealing Factors Limits of Linear Scaling

We conducted experiments with HadGEM2-ES where all O₃ anthropogenic precursor emissions were reduced by 50% and
75% over Europe, to complement the existing 20% emission reduction scenarios performed as part of TF-HTAP2. Figure 2
shows a comparison of the annual and monthly surface O₃ response from the 20%, 50% and 75% European emission reduction
simulations across Europe (a local receptor) and North America (remote receptor), using HadGEM2-ES and the
20 parameterisation based on the O₃ response fields from HadGEM2-ES alone (a self-consistent test of the parameterisation). The
largest errors of <1 ppbv occur over the source region (Fig. 2c), with smaller errors of <0.1 ppbv for the remote receptor region
(Fig 2d). This small internal error between the parameterisation based on HadGEM2-ES and HadGEM2-ES simulations
indicates that the parameterisation of O₃ is working well for emission changes at least as great as 50%. This is similar to the
results of Wild et al., (2012) ~~where more detailed testing found that that the parameterisation resulted in errors of < 1 ppbv for~~
25 ~~emission perturbations of up to 60% and indicates that the parameterisation performs well. Here, monthly mean errors are < 1~~
~~ppbv even for a 75% emission reduction (Fig. 2). The parameterisation is not expected to perform as well for emission~~
~~perturbations of larger than +/- 60% and in source regions under titration regimes.~~ Figure S24 compares the output of
HadGEM2-ES simulations with the parameterisation based on O₃ response fields from multiple models. The magnitude of
error is larger at ~2.0 ppbv over Europe and ~0.3 ppbv over North America for a 75% reduction. This highlights that the
30 uncertainty in the parameterised O₃ response is dominated by the large spread in O₃ responses over the different models rather
than by errors in the parameterisation itself.

3.2 Global Emission Perturbation

To further test the parameterisation, we compare the surface O₃ response from the parameterisation to a HadGEM2-ES model
simulation using the ECLIPSE V5a current legislation scenario (CLE) in 2030 (see
35 http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html, Klimont *et al.*, (2017) and Klimont
et al., (*in prep.*)). The ECLIPSE V5a emission scenarios provide future greenhouse gas and air pollutant emissions based on
assumptions of energy use, economic growth and emission control policies for different anthropogenic emission sectors from
the International Energy Authority (IEA). Three scenarios from ECLIPSE V5a are used in this study: Current Legislation
(CLE) assumes future implementation of existing environmental legislation, Current Legislation with Climate policies (CLIM)

is an energy and climate scenario targeting 2°C of climate warming in which air pollutants and CH₄ are reduced and Maximum Technical Feasible Reduction (MTFR) is the introduction of maximum feasible available technology assuming no economic or technological constraints. Emissions of O₃ precursor species and CH₄ are available at decadal increments over the period 2010 to 2050 for each ECLIPSE scenario (MTFR is only available for 2030 and 2050). The CH₄ abundance was derived from the CH₄ emissions at decadal increments by using a simple box model that accounts for the sources and sinks of CH₄ and the feedbacks on its chemical lifetime following Holmes et al., (2013). Table 4 shows changes in annual CH₄ abundances and NO_x emissions from the ECLIPSE scenario, with changes in CO and NMVOCs shown in Table S1 and S2 respectively. An ECLIPSE CLE 2030 scenario was generated by scaling the anthropogenic emissions in the TF-HTAP2 BASE scenario by the fractional emission changes in NO_x, CO and NMVOCs in CLE. A HadGEM2-ES simulation was performed using the change in emissions based on CLE for comparison to the parameterisation.

~~Fig 3. presents a comparison of M~~monthly (Fig. 3) and annual (Table 5) surface O₃ changes between 2010 and 2030 over the TF-HTAP2 regions for the ECLIPSE V5a CLE scenario from the HadGEM2-ES simulation ~~and are compared to that~~ from the parameterisation (based solely on HadGEM2-ES model responses and based on responses from all models). This shows that the parameterisation is able to reproduce the magnitude and seasonality of surface O₃ changes over different regions when compared to the responses from a full global emission perturbation simulation. In particular, the parameterisation is able to reproduce the seasonality in O₃ across Europe and North America, indicating that the adjustment to represent the new TF-HTAP2 sources regions is valid. Differences between the parameterisations highlight regions where HadGEM2-ES model responses differ from those of the multi-model mean. This is particularly evident for the Middle East, where there are differences of as much as 2 ppb. However, the parameterisation based on results of the HadGEM2-ES model alone agrees relatively well with the model simulation, as expected.

For South Asia, the parameterisation based on HadGEM2-ES and on the multi-model responses agrees well but differs substantially (in sign and magnitude) from the HadGEM2-ES simulated O₃ changes. The largest difference in surface O₃ concentrations of 5 ppbv between the model and the parameterisation occurs in the winter months (December, January, February), with differences in summer being much smaller (0.5 to 1 ppbv). Over the South Asian region the ECLIPSE CLE emission scenario predicts a ~70% increase in NO_x emissions by 2030 (Table 4). ~~This large increase could lead to errors in the parameterised response due to the transition from O₃ production to titration, which the parameterisation is unable to represent well. The large increase in emissions causes the chemical environment in HadGEM2-ES in January to shift from O₃ production to that of titration (Figure S3). The parameterisation is not able to represent this shift (Figure S4) as it is based on a single ozone response to a 20% emission reduction (Figure S5) and is unable to capture the strongly non-linear transition into a net ozone titration regime. This is a smaller problem over North America, Europe or East Asia, as wintertime titration regimes are already present over these regions. This effect seen over South Asia highlights a weakness in the parameterised approach in representing strongly non-linear chemical regimes where there are large emission changes, although we note that the errors would be worse if a linear scaling was used. However, it is able to represent the O₃ responses in the TF-HTAP2 models for a smaller emission change of 20% over South Asia (Table 5).~~ The boundary layer mixing in HadGEM2-ES over South Asia (a region with challenging topography) has been shown to be insufficient, particularly in winter (Hayman et al., 2014; O'Connor et al., 2014), and a large increase in NO_x emissions could lead to a transition to O₃ titration over this region, accounting for some of the discrepancy in surface O₃ responses. However, it is able to represent the O₃ responses in the TF-HTAP2 models for a smaller emission change of 20% over South Asia (Table 65).

As the parameterisation of Wild et al., (2012) did not show a similar discrepancy over South Asia for large emission perturbations, a comparison has been made between the monthly surface O₃ response from HadGEM2-ES and the

parameterisation across both the TF-HTAP1 and TF-HTAP2 definitions of South Asia in January and July (Figure 4). This shows that continental O₃ titration in January is less evident in the HadGEM2-ES simulation over the larger TF-HTAP1 South Asia region, as it includes a large area of ocean. The TF-HTAP2 South Asia region is only continental and HadGEM2-ES shows the larger impact of O₃ titration over the continental region in January. The parameterisation and HadGEM2-ES O₃ responses agree much better over South Asia in July when there is less evidence of O₃ titration effects. The parameterisation, using only HadGEM2-ES as input, is not able to represent the O₃ response in HadGEM2-ES over TF-HTAP2 South Asia as it is based on a 20% emission reduction simulation of HadGEM-ES, where the extent of O₃ titration over the continental area is small. Additional model simulations conducted with large emission increases over South Asia would be ~~able~~ valuable to further explore this issue, although none are currently available.

These results highlight that caution is needed when applying the parameterisation with emission changes larger than 50-60%, as noted previously in Wild et al., (2012). In particular, the shift into O₃ chemical titration regimes cannot be represented easily in a simple parameterisation. For smaller emission changes, the parameterisation is shown to be relatively robust at representing monthly surface O₃ changes.

3.3 Comparison to HTAP-I

3.3.1 CMIP5 Scenarios

We now use the improved parameterisation described above to explore how future predictions of regional surface O₃ for the RCPs used in CMIP5 have changed since TF-HTAP1. The four RCPs assume different amounts of climate mitigation to reach a target anthropogenic radiative forcing in 2100: RCP2.6, RCP4.5, RCP6.0 and RCP8.5 (van Vuuren et al., 2011). Emissions of O₃ precursor species and CH₄ are available at decadal increments over the period 2010 to 2050 for each RCP. CH₄ emissions are converted to CH₄ abundances in each RCP using the MAGICC model which takes into account feedbacks on the CH₄ lifetime (Meinshausen et al., 2011). The parameterisation only accounts for the impact from changes in anthropogenic emissions over the period 2010 to 2050 and does not account for changes in climate, but on this near-term timescale changes in O₃ are dominated by emission changes rather than climate effects (Fiore et al., 2012). There are large differences in global CH₄ abundances in the four scenarios, and this strongly influences the O₃ responses.

Figure 5 shows the change in surface O₃ across TF-HTAP2 regions for each of the RCPs. Surface O₃ decreases across most regions in the majority of the scenarios as O₃ precursor emissions are reduced. The largest increases in surface O₃ occur over South Asia in RCP8.5 due to the expected increases in O₃ precursor emissions from 2010 to 2050, although we note that this effect may be exaggerated by the large increase in NO_x emissions here in RCP4.5 and RCP8.5. Surface O₃ concentrations are predicted to increase over most regions in RCP8.5 with increases of 2 ppbv by 2050 over the Middle East and Southern Africa (Table S3). The results in Fig 5. across Europe, North America, South Asia, East Asia and globally are similar to those based on TF-HTAP1 in Wild et al., (2012) (Fig. 5 and Table 7~~6~~) but differ slightly in magnitude due to the change in the spatial extent of the individual source regions from TF-HTAP1 to TF-HTAP2. Additionally, the ~~improved~~ parameterisation [here](#) provides O₃ changes for other regions that were not previously available, including the Middle East and Africa. This provides useful additional information on surface O₃ over these important regions under future emission change.

3.3.2 Sensitivity of Ozone to Methane

The importance of controlling CH₄ to achieve future reductions in O₃ has been highlighted in earlier studies, along with the large uncertainty in the response of O₃ to CH₄ changes (Fiore et al., 2009; Wild et al., 2012). The inclusion of new models provides an opportunity to assess whether the sensitivity of O₃ to CH₄ identified in TF-HTAP1 remain the same. Experiments with both increased (CH4INC) and decreased (CH4DEC) global abundance of CH₄ were conducted in TF-HTAP2. However,

these experiments used an increase of 18% and a reduction of 13% to align with 2010 to 2030 changes in global CH₄ abundance under RCP8.5 and RCP2.6, in contrast to the 20% reduction used in TF-HTAP1.

Wild et al., (2012) found that a 20% increase in CH₄ abundance yielded an 11.4% smaller surface O₃ response than that from a 20% decrease in CH₄. For simplicity the parameterisation used the same non-linear scaling factor as for NO_x emissions (Eq. 42 i.e. $g = 0.95f + 0.05f^2$), which represents a 10% smaller response for successive 20% emission increases. The two TF-HTAP2 models that contributed results to both CH4DEC and CH4INC simulations allow us to check the expression used here. We find a slightly larger sensitivity, with both models yielding a 12.6% smaller surface O₃ response for an increase in CH₄ than a decrease (Eq. 3). Since this O₃ response to CH₄ in TF-HTAP2 is comparable to that from TF-HTAP1, for simplicity and consistency we chose to retain the same scaling factor representation of non-linearity for both NO_x and CH₄ (Eq. 42), as used in Wild et al., (2012).

$$g = 0.937f + 0.063f^2 \tag{3}$$

To enable a direct comparison with TF-HTAP1 results, the O₃ response from the CH4DEC and CH4INC experiments in TF-HTAP2 are scaled to represent the response from a 20% reduction in CH₄ abundances, using Eq. 3. An adjustment factor is calculated based on the global mean difference between the TF-HTAP2 O₃ response in each experiment and that of an equivalent 20% reduction in CH₄ abundance (calculated using Eq. 3), resulting in a factor of 1.557 for CH4DEC and -1.256 for CH4INC. The global mean O₃ responses from CH4DEC (-0.69 ± 0.01 ppbv, 2 models) and CH4INC (0.81 ± 0.14 ppbv, 7 models) are adjusted to generate the equivalent O₃ responses to a 20% reduction in CH₄ abundance, which are used in the parameterisation (-1.05 ± 0.12 ppbv). This response is ~14% larger globally than that in TF-HTAP1 (-0.90 ± 0.14 ppbv, 14 models), highlighting a slightly increased sensitivity of O₃ to CH₄.

To explore the differences between TF-HTAP1 and TF-HTAP2 models the CH₄ lifetime and feedback factor for each TF-HTAP2 model (where data is available) can be calculated in accordance with Fiore et al., (2009). The feedback factor is the ratio of the atmospheric response (or perturbation) time to global atmospheric lifetime and describes how the atmospheric CH₄ abundance responds to a perturbation in CH₄ emissions e.g. a feedback factor of 1.25 means that a 1% increase in emissions would ultimately generate a 1.25% increase in CH₄ concentrations (Fiore et al., 2009). The feedback factors can be used in conjunction with CH₄ emission changes for a region, to relate the O₃ response from the reduction in CH₄ abundance in TF-HTAP scenarios to that equivalent from emissions, taking into account both the long-term and short response of emissions on O₃ (Fiore et al., 2009). Table 87 summarises the calculated CH₄ lifetime and feedback factors for the two TF-HTAP2 models that have provided the appropriate fields CH₄ chemical loss rates. These two models show slightly shorter methane lifetimes and a higher feedback factor (F) than the TF-HTAP1 mean values. This suggests that the sensitivity of O₃ to changes in CH₄ in the two TF-HTAP2 models is slightly larger than the TF-HTAP1 multi-model mean. The increased feedback factor also indicates that a slightly larger reduction in methane emissions is required to achieve a comparable reduction in O₃ concentrations.

Overall, the sensitivity of O₃ to a change in CH₄ abundance is slightly larger in the two TF-HTAP2 models considered here than in TF-HTAP1 models, but still within the range of the TF-HTAP1 multi-model ensemble. The results from TF-HTAP2 will not significantly change any conclusions from TF-HTAP1 but suggests that the previous O₃ changes estimated from TF-HTAP1 are conservative. The O₃ response to CH₄ remains one of the most important processes to understand for controlling future O₃ concentrations.

4. Future Surface Ozone Predictions

4.1 Surface Ozone under ECLIPSEv5a Emissions

The parameterised approach is used with the ECLIPSE v5a emission scenarios described above to determine regional changes in future surface O₃ concentrations. Surface O₃ concentrations for the CLE (current legislation) scenario are predicted to increase from 2010 to 2050 across all regions (Figure 6). Annual mean surface O₃ concentrations increase by 4 to 8 ppbv across the South Asia and Middle East regions due to the large increases expected in NO_x emissions (Table 4), although there is substantial uncertainty in the parameterisation over these regions. Surface O₃ concentrations over Europe and North America in 2050 are similar to those in 2010, even though their regional NO_x emissions decrease by ~50%. The contributions of different sources to the total surface O₃ change has been analysed for each source region (Figures S26 to S173). Results for Europe (Figure. 7) and South Asia (Figure. 8) are shown here, as these regions experience contrasting changes in surface O₃. Across Europe, surface O₃ from local and remote (mainly North American) sources is reduced in response to emission decreases, and the contribution from CH₄ increases by 1.6 ppbv in 2050 (Fig. 7). The increase in global CH₄ abundance in the CLE scenario increases surface O₃ over Europe, offsetting the reduction in O₃ from local and remote sources. This contrasts strongly with South Asia where local sources dominate the total O₃ response. This demonstrates how different local and hemispheric emission control strategies are needed in different regions.

For the CLIM (climate policies on current legislation) scenario, annual mean surface O₃ concentrations in 2050 decrease slightly or stay at 2010 concentrations due to reductions in anthropogenic emissions and control of CH₄ emissions leading to a decrease in its abundance (Table 4). The source contribution analysis for Europe (Fig. 7) and South Asia (Fig. 8) shows that CH₄ contributes much less to the total surface O₃ change under this scenario than CLE. For South Asia, there is also a reduction in the contribution from local sources to surface O₃. Under CLIM, remote sources start to dominate the contribution to European surface O₃ changes in 2050, increasing to -1.3 ppbv. However, across South Asia the contribution from local sources (+3.2 ppbv) is greater than from remote sources (-1.4 ppbv) in 2050, reflecting the importance of local emissions in this region. The contribution of CH₄ sources to the total surface O₃ response is smaller in CLIM due to the targeting of CH₄ for climate mitigation purposes. The implementation of these climate policy measures shifts the dominant factor driving future O₃ changes within a receptor region towards extra-regional sources.

The MTR scenario (maximum technically feasible reduction) considers large reductions in emissions (Table 4) and consequently predicts reductions in surface O₃ concentrations of up to 9 ppbv by 2050. Reductions of surface O₃ in Europe (Fig. 7) are dominated by changes in remote sources, although changes in CH₄ become increasingly important by 2050. For South Asia, the surface O₃ response is dominated by changes to local and remote emission sources. This highlights that achieving decreases in surface O₃ concentrations from the maximum feasible emissions reductions depends not only on local emission policies but on reducing emissions across other regions too.

4.2 Surface Ozone under CMIP6 Emissions

We provide an initial assessment of surface O₃ changes from a subset of the preliminary emission scenarios developed for the CMIP6 project (<https://tntcat.iiasa.ac.at/SspDb>; Rao *et al.*, 2017) based on shared socio-economic pathways (SSPs). Five baseline SSPs are defined (SSP1-5) based on different combinations of future social, economic and environmental development trends over centennial timescales (O'Neill *et al.*, 2014). Different climate targets, defined in terms of anthropogenic radiative forcing by 2100, are combined with the baseline SSPs to develop future scenarios for climate mitigation, including additional assumptions on international co-operation, timing of mitigation and extent of fragmentation between low and high income economies (van Vuuren *et al.*, 2014; Riahi *et al.*, 2017). Scenarios of strong, medium and weak

future air pollutant emission pathways are mapped onto the SSPs and represent differing targets for pollution control, the speed at which developing countries implement strict controls and the pathways to control technologies (Rao et al., 2017). Increasingly stringent air pollutant emission controls are assumed to occur with rising income levels because of the increased focus on human health effects and the declining costs of control technology. SSP2 is a medium pollution control scenario that follows current trajectories of increasing levels of regulation. SSP1 and SSP5 are strong control scenarios where pollution targets become increasingly strict. A weak pollution control scenario is adopted in SSP3 and SSP4 where the implementation of future controls are delayed (Rao et al., 2017).

We select three preliminary SSPs to represent scenarios of business as usual (SSP3 BASE), middle of the road (SSP2 60) and enhanced mitigation (SSP1 26). The SSP2 60 and SSP1 26 scenarios have climate mitigation targets of 6.0 and 2.6 W m⁻² in 2100 applied to them. Currently, air pollutant emissions for each SSP are available globally and across five world regions from <https://tntcat.iiasa.ac.at/SspDb/>. The air pollutant emissions for each region have been mapped onto the equivalent TF-HTAP2 source regions and the grouping of regions is shown in Table 98 along with the percentage change in global CH₄ abundance and NO_x emissions over the period 2010 to 2050. The relative changes in CO and NMVOCs emissions are shown in Table S4 and S5. Gridded versions of these emission scenarios will be made available in due course (K. Riahi, personal communication, 2017), which will allow a more accurate evaluation of the impacts arising from these scenarios.

Surface O₃ concentrations increase across all regions in 2050 for the SSP3 BASE scenario (Figure 9). Europe, North America and East Asia show an increase in surface O₃ of 1 to 3 ppbv, a larger response than in the ECLIPSE CLE scenario. Smaller increases in surface O₃ are predicted over the Middle East (~3 ppbv) and South Asia (~5 ppbv) compared to CLE. Methane dominates the total surface O₃ response over Europe in SSP3 BASE, with small contributions from local and remote emission sources over the period 2010 to 2050 (Fig. 10). Local emissions are the main contribution to O₃ changes over South Asia, with a slightly larger influence from CH₄ than in CLE (Fig. 11).

For SSP2 60 (middle of the road scenario), surface O₃ concentrations reduce slightly by 2050 and to a greater extent than in ECLIPSE CLIM due to the larger reductions in NO_x emissions and global CH₄ abundances (Table 4 and 9). Over South Asia, NO_x emissions in SSP2 60 decrease by 22% from 2010 to 2050, with a corresponding O₃ change of -3 ppbv, compared to CLIM where NO_x emissions increase by 66% and the corresponding O₃ change is +3 ppbv. However, this difference could arise from using preliminary SSP emissions based on five large world regions, where emission changes in South Asia and nearby regions such as East Asia are combined together. For Europe and South Asia the source contributions for each region (Fig. 10 and 11) are similar to those in CLIM. Remote sources are more important under this intermediate climate mitigation scenario, with local emissions sources becoming more important by 2050 over South Asia.

Large reductions in surface O₃ concentrations are predicted across all regions in the strong mitigation scenario (SSP1 26) (Fig. 9). The improvements in O₃ concentrations are less than predicted under the ECLIPSE MTRF due to a smaller reduction in NO_x emissions. Northern mid-latitude regions show reductions in surface O₃ concentrations of up to 6 ppbv under SSP1 26, similar to MTRF. Over South Asia, surface O₃ is predicted to be reduced by up to 7 ppbv, which is less than under MTRF. The source contributions for both Europe (Fig. 10) and South Asia (Fig. 11) are similar to MTRF with the importance of remote sources and the increasing importance of CH₄ by 2050 evident over Europe. Over South Asia, the increasing importance of local and CH₄ sources is clear by 2050.

This analysis of preliminary CMIP6 emission scenarios highlights the large range of future regional surface O₃ responses that are possible depending on the climate and air pollutant policies applied. The assumptions within each of the future SSPs,

particularly for CH₄, results in different sources dominating the contribution to the total surface O₃ response. Uncertainties in the assumed growth rate of CH₄ under the two current legislation scenarios (CLE and SSP3 BASE) result in a 1 ppbv difference in surface O₃ over Europe and North America, highlighting the importance for future air quality of reducing CH₄ on a global scale. The CMIP6 scenarios allow a larger range of pathways to be explored than were available in ECLIPSE or the CMIP5 RCPs, including those of strong, medium and weak policies on air pollutants and climate change. The parameterisation can be used to provide a rapid assessment of the impact of differing policy measures on surface O₃ concentrations across different regions, along with a clear source attribution. This can ultimately inform selection of policies that are most beneficial to future air quality.

5. Future Tropospheric Ozone Burden and Radiative Forcing

As discussed in section 2.3, the parameterisation has been extended to generate three-dimensional O₃ distributions throughout the troposphere, using a tropopause defined as a O₃ concentration of 150 ppbv (Prather et al., 2001). Tropospheric O₃ column burdens are calculated in each grid cell for each emission scenario. These are used to infer changes in O₃ radiative forcing by using the relationship between radiative forcing and tropospheric column O₃ ($\text{W m}^{-2} \text{DU}^{-1}$) and its spatial variation with latitude and longitude from the ACCMIP multi-model ensemble (Stevenson et al., 2013). Tropospheric O₃ burdens and O₃ radiative forcings are calculated for the CMIP5 RCPs to evaluate the parameterisation against values from the ACCMIP multi-model study (Stevenson et al., 2013). Additionally, future projections of O₃ radiative forcing are made for the ECLIPSE and CMIP6 SSPs.

The change in tropospheric O₃ burden and O₃ radiative forcing for the ECLIPSE CLE scenario in 2030 from the parameterisation was evaluated against the change from the equivalent HadGEM2-ES simulation (Table 5), a self-consistent test based only on emission perturbations with no influence from climate change. The parameterisation is able to reproduce the a change in global tropospheric O₃ burden (-0.93 Tg) and O₃ radiative forcing (-0.6 mW m⁻²) of -0.9 Tg which compares well with the -1.0 Tg change simulated by from HadGEM2-ES simulations(-0.95 Tg and -0.9 mW m⁻²), with any slight differences due to the discrepancies identified over South Asia (Figure 3).

In comparison to the ACCMIP multi-model mean (Stevenson et al., 2013), the parameterisation produces changes in the global O₃ burden and O₃ radiative forcing from 2000 to 2030 for RCP2.6 and RCP6.0 (Table 9) relatively well but underestimates the magnitude of change in RCP4.5 and RCP8.5 (although it remains within the range of ACCMIP model responses). The parameterisation also underestimates the changes in global O₃ burden and radiative forcing between 1980 and 2000 compared to the ACCMIP multi-model mean. The parameterised response compares reasonably well for smaller emission changes but is unable to reproduce fully the larger changes in the high emission (low mitigation) scenario (RCP8.5), as it does not include natural emissions or represent impacts on O₃ from changes in climate that are contained within the ACCMIP models. The net impact of climate change on global tropospheric O₃ radiative forcing was estimated from the ACCMIP multi-model ensemble to be between -20 to -30 mW m⁻² (a negative feedback) (Stevenson et al., 2013). In comparison to the ACCMIP multi-model mean, the predicted changes between 2000 to 2030 in both global annual mean surface O₃ and global O₃ burden from the parameterisation are within the range of the ACCMIP multi model responses (+/- 1 standard deviation) for all the CMIP5 RCPs (Table 10). The predictions of O₃ radiative forcing in 2030 from the parameterisation across all the RCPs, when the influence of climate change is anticipated to be small, are also consistent with those from ACCMIP. The sign and magnitude of change in global O₃ burden and O₃ radiative forcing with the parameterisation for RCP6.0 is different from the ACCMIP results but is still within the range of model responses, which is the largest for this scenario. The comparison with ACCMIP

results shows that the parameterisation is able to reproduce changes in global O₃ burden and O₃ radiative forcing on near-term timescales, when the influence of climate change is small.

A global O₃ radiative forcing of +0.05 to +0.08 W m⁻² in 2050, relative to 2010, is estimated under the low mitigation scenarios (RCP8.5, CLE and SSP3 BASE) (Figure 12). The intermediate mitigation scenarios of RCP4.5, RCP6.0 and SSP2 60 show an O₃ radiative forcing of 0 to -0.04 W m⁻² in 2050, with almost no change under CLIM. The more stringent mitigation scenarios (RCP2.6, MTFR and SSP1 26) exhibit an O₃ radiative forcing of between -0.07 and -0.15 W m⁻² by 2050. The parameterisation is able to predict the wide range of impacts that climate and air quality policies could have on short-term climate forcing from O₃. It can be used as a rapid screening tool to select the most appropriate climate scenarios to explore further in full model simulations that can provide more detailed predictions. The current business-as-usual scenarios for CMIP5, ECLIPSE and CMIP6 increase the climate forcing of O₃ by approximately 0.06 W m⁻² in 2050, whereas the strong mitigation scenarios have a larger effect in reducing the near-term climate forcing of O₃ by about 0.10 W m⁻².

The parameterisation generates gridded changes in the tropospheric O₃ column burden and radiative forcing, which can be used to calculate changes over different regions. Figures 13 and 14 show that the largest relative changes in O₃ burden for the ECLIPSE scenarios occur over the Middle East, South Asia and South East Asia (> 10%), with a corresponding larger impact on O₃ radiative forcing (-0.3 W m⁻² in MTFR). Smaller relative changes in the tropospheric O₃ burden are found for CLE over Europe and North America. For MTFR a 15% reduction in O₃ burden is predicted over Europe and North America, similar to that over South Asia, but the change in O₃ radiative forcing is not as large (-0.2 W m⁻² compared to -0.3 W m⁻² over South Asia). The parameterisation allows the regional near-term climate implications (in terms of O₃ radiative forcing) from future emissions changes to be explored under different air quality and climate policy scenarios. It also highlights the wide range of near-term climate forcing that is possible over particular regions from future emission policies.

6. Conclusions

In this study, we describe improvements and extensions to a simple parameterisation of regional surface O₃ responses to changes in precursor emissions and CH₄ abundances based on multiple models. We incorporate results from phase 2 of the Hemispheric Transport of Air Pollutants project to create an enhanced parameterisation that includes new models from TF-HTAP2, a greater number of source regions (14 in total), a new base yearline of 2010 and an extension to three dimensions to represent O₃ changes throughout the troposphere. These improvements allow impacts on surface O₃ concentrations and the near-term O₃ radiative forcing to be calculated from different emission scenarios. Model simulations using HadGEM2-ES confirm the validity of the parameterisation and adjustments made here. However, larger errors may occur when using emission changes of greater than +/- 60% and when considering long term future scenarios where there may be a significant influence from climate change. In addition, the parameterisation may not perform well over regions where chemical titration is expected to become dominant in the future under large emission increases e.g. South Asia, as it is based on the ozone responses in 2010. There is a slight increase in the response of O₃ to CH₄ for the TF-HTAP2 models, resulting in a slightly higher sensitivity of O₃ to CH₄ changes. The extent of the difference varies on a regional basis, but is within the range of model responses in TF-HTAP1.

~~The parameterisation is shown to perform well under most conditions, although there are larger uncertainties for future surface O₃ responses over South Asia where changes in emissions are particularly large.~~ Emission changes from the RCPs are used with the parameterisation and it predicts similar changes in surface O₃ concentrations to those from the original parameterisation (Wild et al., 2012), although now across a larger number of source/~~receptor~~ regions. Tropospheric O₃ burden

and O₃ radiative forcing calculated using the parameterisation ~~compare well with the~~ are within the spread of the response from the ACCMIP ~~multi-model mean values~~ models for all of the intermediate CMIP5 RCPs ~~emission scenarios (RCP2.6 and RCP6.0)~~ in 2030, ~~where the influence from climate change is anticipated to be small-but underestimate the large changes in RCP8.5 due to the neglect of climate induced changes within the parameterisation.~~ The parameterised approach permits rapid

5 assessment of the impact of future emission changes over 14 source regions and associated uncertainties on both surface and tropospheric O₃ concentrations, and allows identification of the differing contributions of local, remote and CH₄ sources to the O₃ response. This enables quantification of the impacts of future air quality and climate emission policies on surface air quality and near-term climate forcing by O₃.

10 Applying future emissions from ECLIPSEv5 and the preliminary SSPs, we show that annual mean surface O₃ concentrations are likely to increase across most world regions by 2050 under current legislation scenarios, with large increases of 4 to 8 ppbv over the Middle East and South Asia. These changes in O₃ concentrations are driven mainly by local emissions and changes in global CH₄ abundance. This demonstrates that current legislation is inadequate in preventing future increases in surface O₃ concentrations across the world. Implementing energy related climate policies on top of current legislation maintains future

15 surface O₃ concentrations at or slightly below 2010 concentrations, counteracting the increases that occur under current legislation. This is achieved mainly through reductions in CH₄, highlighting the importance of controlling CH₄ in limiting future changes in O₃ concentrations, as shown in Wild et al., (2012). Policies that have stringent emission controls lead to substantial reductions in surface O₃ concentrations across all world regions of up to 8 ppbv and could potentially provide large beneficial impacts.

20 A global O₃ radiative forcing of +0.07 W m⁻² is predicted by 2050 (relative to 2010) under the current legislation scenarios of the SSPs and ECLIPSE. There is a large and diverse regional response in O₃ radiative forcing with some regions e.g. Middle East and South Asia more sensitive to changes in emissions than others, and these show a large positive O₃ radiative forcing under current legislation. However, application of aggressive emission mitigation measures leads to large reductions in O₃

25 radiative forcing (-0.10 W m⁻²), lessening the near-term impact on climate.

The new parameterisation provides a valuable assessment tool to evaluate the impact of future emission policies on both surface air quality and near-term climate forcing from O₃. It also provides a full source attribution along with a simple measure of uncertainty, given by the spread of the multi-model responses that reflect different transport and chemistry processes in

30 models. Whilst not replacing full chemistry simulations it provides a quick way of assessing where to target future modelling efforts. However, these O₃ responses are based on changes to anthropogenic emissions only, with no account taken of the impact on O₃ and/or its natural precursor emissions due to future changes in chemistry or climate. The parameterisation could be extended further by including a feedback factor to take some account of the impact of future climate change on O₃. Additional improvements could include coupling the output to an offline radiation model to enable improved calculation of O₃

35 radiative forcing, using O₃ fields from the parameterisation within a land surface model to assess the impacts of O₃ on vegetation and the carbon cycle or with O₃ dose-response functions to calculate impacts on human health.

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Competing Interests. The authors declare that they have no conflict of interest.

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Table 1. Summary of annual mean NO_x, CO and NMVOC emissions changes (%) between 2000 and 2010 over TF-HTAP-1 source regions (values are mean differences from the MACCity and EDGARv4.3.1 emission inventories)

Annual total relative (%) emission change between 2000 and 2010						
	Global	Europe	North America	South Asia	East Asia	Rest of World
NO _x	9.5	-8.4	-25.0	49.8	42.1	13.0
CO	-1.2	-27.1	-47.1	18.8	15.6	9.0
NMVOCs	5.2	-9.7	-31.2	32.1	24.8	10.0

5

Table 2. Summary of multi-model annual mean surface ozone values from all TF-HTAP1 models in 2001 (H-1), the parameterisation of TF-HTAP1 models scaled for emissions in 2010 (H-P) and all TF-HTAP2 models in 2010 (H-2) (seven models contributing).

	Ozone Concentrations (ppbv)														
	Global			Europe			North America			South Asia			East Asia		
	H-1	H-P	H-2	H-1	H-P	H-2	H-1	H-P	H-2	H-1	H-P	H-2	H-1	H-P	H-2
Min	21.2	21.5	23.0	30.2	30.1	29.9	29.4	28.3	29.7	35.2	37.6	35.3	28.9	30.8	31.7
Mean	27.4	27.2	26.4	37.4	36.9	35.8	35.8	34.9	35.1	40.1	42.4	40.7	35.5	37.2	35.5
Max	32.0	30.0	32.3	42.8	42.4	42.0	40.8	39.7	41.2	44.8	48.0	50.7	38.9	40.7	41.3
Standard Deviation	2.94	2.71	3.33	3.84	3.79	4.45	3.56	3.54	3.85	3.73	3.80	3.35	2.92	2.91	5.27

5 NB - The TF-HTAP2 models used to provide the 2010 ozone concentrations are CAMchem, Chaser_re1, Chaser_t106, C-IFS, GEOS-Chem adjoint, HadGEM2-ES, Oslo-CTM

Table 3. Models contributing to each of the TF-HTAP2 emission perturbation experiments, in addition to those for the source regions of Europe, North America, South Asia and East Asia

TF-HTAP2 Model	TF-HTAP2 Experiment											
	CH4INC	CH4DEC	MDE	RBU	NAF	SAF	MCA	SAM	SEA	CAS	PAN	OCN
GFDL-AM3 (Lin et al., 2012)	X											
C-IFS (Flemming et al., 2015)	X											
CAM-Chem (Tilmes et al., 2016)			X	X								
CHASER_re1 (Sudo et al., 2002)	X		X	X								
CHASER_t106 (Sudo et al., 2002)	X											
EMEP_rv4.8 (Simpson et al., 2012)	X		X	X	X							X
GEOS-Chem (Henze et al., 2007).			X	X								
HadGEM2-ES (Collins et al., 2011)	X	X	X	X	X	X	X	X	X	X	X	X
OsloCTM3_v2 (Søvde et al., 2012)	X	X	X	X			X	X				
Total Number of Models	7	2	6	6	2	1	2	2	1	1	1	1

MDE – Middle East, RBU – Russia Belarus Ukraine, NAF – North Africa, SAF – Southern (Sub-Saharan/Sahel) Africa, MCA – Mexico and Central America, SAM – South America, SEA – South East Asia, CAS – Central Asia, PAN – Pacific Australia and New Zealand, OCN – Ocean (for region definitions see Koffi et al., 2016)

Table 4. Percentage change in global CH₄ abundance and global and regional annual NO_x emissions relative to 2010 over each TF-HTAP2 region for the different ECLIPSE V5a emission scenarios (CLE, CLIM and MTRF). MTRF scenarios are only available for 2030 and 2050.

TF-HTAP2 Region	Annual total emission change (%) from 2010									
	CLE				CLIM			MTRF		
	2020	2030	2040	2050	2020	2030	2040	2050	2030	2050
Global CH ₄ Abundance	4	12	21	32	3	8	11	13	-9	-21
Global NO _x	-7	-6	6	19	-17	-27	-26	-24	-88	-86
	<i>Regional NO_x Emissions</i>									
Central America	13	11	21	30	1	-16	-16	-11	-46	-79
Central Asia	10	15	18	26	-5	-16	-26	-32	-57	-80
East Asia	-14	-16	-8	-3	-16	-27	-25	-24	-50	-61
Europe	-31	-46	-50	-50	-33	-51	-57	-58	-67	-72
Middle East	18	31	51	72	-16	-19	-20	-23	-37	-76
North Africa	-9	3	24	53	-24	-25	-16	-2	-56	-71
North America	-28	-51	-51	-51	-31	-55	-59	-64	-73	-78
North Pole	1	-1	-15	-13	-15	-22	-19	-23	-61	-78
Ocean	-6	-0.2	11	25	-14	-22	-29	-27	-51	-64
Pacific Aus NZ	-20	-31	-32	-33	-28	-53	-58	-63	-72	-84
Russia Bel Ukr	-1	-4	-9	-8	-18	-28	-29	-35	-62	-74
Southern Africa	10	13	30	49	-12	-21	-18	-12	-41	-50
South America	-6	1	15	28	-9	-11	-6	-2	-46	-66
South Asia	19	67	139	199	-1	12	41	66	-29	-48
South East Asia	24	45	71	101	-1	-7	-5	1	-35	-59

Table 5. Parameterised responses based only on HadGEM2-ES input for annual mean surface ozone, global ozone burden and ozone radiative forcing using the ECLIPSE CLE emission scenarios in 2030, with changes calculated relative to the year 2010.

<u>Scenario</u>	<u>Surface Ozone (ppbv)</u>		<u>Ozone Burden (Tg)</u>		<u>Ozone Radiative Forcing (mW m⁻²)</u>	
	<u>Param¹</u>	<u>HadGEM2-ES</u>	<u>Param¹</u>	<u>HadGEM2-ES</u>	<u>Param¹</u>	<u>HadGEM2-ES*</u>
<u>ECL 2030</u>	<u>-0.21</u>	<u>-0.20</u>	<u>-0.93</u>	<u>-0.95</u>	<u>-0.6</u>	<u>-0.9</u>

¹ Parameterisation based only on HadGEM2-ES input

- 5 * Ozone radiative forcing is calculated by applying the same methodology as in the parameterisation (using the relationship between radiative forcing and tropospheric column O₃ change based on multi-model ensemble mean results from ACCMIP

Table 65. Monthly and annual mean surface O₃ changes (ppbv) from the TF-HTAP2 multi-model mean and the parameterisation over South Asia due to a 20% reduction in anthropogenic precursor emissions over this region. Multi-model mean values are shown with +/- 1 standard deviation for the available TF-HTAP2 models and the parameterised approach is based on multiple models.

TF-HTAP2 South Asia Experiment	Surface O ₃ response (ppbv +/- one standard deviation)				
	January	April	July	October	Annual Mean
TF-HTAP2 Multi-model Model^{NB}	-1.67 ± 0.73	-1.48 ± 0.29	-1.22 ± 0.21	-1.72 ± 0.44	-1.51 ± 0.35
Parameterisation mean (multi-models)	-1.58 ± 0.54	-1.48 ± 0.39	-1.09 ± 0.33	-1.89 ± 0.55	-1.50 ± 0.29

^{NB} – Models contributing to the multi-model mean are C-IFSv2, CAMchem, CHASER_re1, CHASER_t106, GEOSCHEM-adjoint, HadGEM2-ES, OsloCTM3.v2.

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Table 76. Annual mean surface O₃ change (ppbv plus one standard deviation) in 2030 and 2050 (relative to 2010) for each RCP scenario derived from the parameterisation in this study and that of Wild et al., (2012).

CMIP5 RCP	Global Surface O ₃ response from 2010 to 2050 (ppbv)			
	This Study		Wild et al., (2012)	
	2030	2050	2030	2050
RCP2.6	-1.51 +/- 0.1	-21.94 +/- 0.3	-1.21 +/- 0.3	-2.0 +/- 0.5
RCP4.5	-0.1 +/- 0.1	-10.81 +/- 0.2	-0.2 +/- 0.2	-0.8 +/- 0.4
RCP6.0	-0.54 +/- 0.1	-0.47 +/- 0.1	-0.4 +/- 0.1	-0.4 +/- 0.2
RCP8.5	+01.70 +/- 0.2	+1.50 +/- 0.5	+1.0 +/- 0.2	+1.5 +/- 0.5

Table 87. Methane lifetime (τ) and feedback factor in TF-HTAP2 models that provided appropriate data (OH and CH₄ concentrations).

Model	TF-HTAP2 Experiment	τ_{OH}^1	T_{total}^2	F^3
CHASER_re1	BASE	7.19	6.51	
	CH4INC	7.62	6.86	1.46
HadGEM2-ES	BASE	8.8	7.8	
	CH4INC	9.29	8.17	1.40
	CH4DEC	8.43	7.51	1.37
TF-HTAP1 Mean		10.19 +/- 1.72	8.84 +/- 1.33	1.33 +/- 0.06

¹ CH₄ lifetime for loss by tropospheric OH (years) defined as atmospheric burden in each experiment divided by the tropospheric CH₄ loss rate with OH with a tropopause of 150 ppb of O₃ used.

² Total atmospheric CH₄ lifetime (years) defined as the reciprocal mean of τ_{OH} and assuming a lifetime in the stratosphere and soils of 120 years and 160 years respectively (Prather et al., 2001).

³ The feedback factor is the ratio of the atmospheric response (or perturbation) time to the global atmospheric lifetime. It is defined as $f = 1/(1 - S)$ where S is determined from the BASE and CH₄ perturbation simulations and defined as $S = (\delta \ln(\tau))/(\delta \ln[CH_4])$ and CH₄ abundances for TF-HTAP2 are 1798 ppbv in BASE, 1562 ppbv in CH4DEC and 2121 ppbv for CH4INC (Prather et al., 2001).

Table 98. Percentage change in global CH₄ abundance and global and regional NO_x emissions relative to 2010 over each TF-HTAP2 world region for the different CMIP6 emission scenarios (SSP1 26, SSP2 60 and SSP3 BASE)

TF-HTAP2 Region	Annual total emission change (%) from 2010											
	SSP1 26				SSP2 60				SSP3 BASE			
	2020	2030	2040	2050	2020	2030	2040	2050	2020	2030	2040	2050
Global CH ₄	1	-7	-16	-23	4	6	7	5	8	18	28	37
Global NO _x	-8	-25	-35	-48	-7	-9	-16	-21	10	14	15	16
<i>Regional NO_x Emissions</i>												
Central America, South America	-2	-22	-27	-34	-10	-11	-15	-24	13	22	30	36
Central Asia, Rus Bel Ukr	-14	-32	-40	-49	1	2	-5	-14	-1	-5	-5	-12
East Asia, South Asia, South East Asia	4	-8	-22	-35	-3	-1	-12	-22	26	45	54	54
Europe, North America, Pacific Aus NZ	-31	-62	-68	-74	-31	-43	-51	-57	-8	-22	-30	-32
Middle East, North Africa, Southern Africa	4	-3	-4	-2	3	11	13	12	7	14	26	33

Table 109. Multi-mean parameterised responses in annual mean surface, global ozone burden and ozone radiative forcing in 1980 and for the CMIP5 emission scenarios in 2030, with changes calculated relative to the year 2000 for comparison with values from ACCMIP (+/- 1 standard deviation of multi-model responses).

Year	Ozone Burden (Tg)				Ozone Radiative Forcing relative to 2000 (mW m ⁻²)	
	Total	Change from 2000				
	Parameterisation	ACCMIP*	Parameterisation	ACCMIP*	Parameterisation	ACCMIP*
1980	322	322 +/- 22	-10	-15	-42	-59 +/- 59
2000	332	337 +/- 24	0	0	0	0
RCP2.6 2030	318	319 +/- 22	-14	-18	-49	-39 +/- 52
RCP4.5 2030	331	344 +/- 26	-1	+7	+4	+37 +/- 45
RCP6.0 2030	327	336 +/- 31	-5	-1	-17	-16 +/- 66
RCP8.5 2030	340	357 +/- 26	+8	+20	+35	+83 +/- 61

5 * - Mean tropospheric Ozone burden and radiative forcing between future year and 2000s from the ACCMIP multi-model ensemble as presented in Young et al., (2013) and Stevenson et al., (2013)

Year	Surface Ozone (ppbv)		Ozone Burden (Tg)		Ozone Radiative Forcing (mW m ⁻²)	
	Param	ACCMIP*	Param	ACCMIP*	Param	ACCMIP*
1980	-1.3	-1.3 +/- 0.4	-17	-15 +/- 6	-67	-59 +/- 21
2000	0	0	0	0	0	0
RCP2.6 2030	-1.1	-1.5 +/- 0.6	-12	-12 +/- 8	-45	-45 +/- 30
RCP4.5 2030	-0.1	+0.2 +/- 0.5	+4	+7 +/- 5	+14	+24 +/- 19
RCP6.0 2030	-0.4	-0.8 +/- 1.0	+0.3	-2 +/- 11	+2	-13 +/- 39
RCP8.5 2030	+1.0	+1.5 +/- 0.7	+20	+23 +/- 7	+80	+81 +/- 26

* - Mean change in the tropospheric Ozone burden and radiative forcing between 2030 and 2000s from the ACCMIP models that provided results for each year of each scenario, as presented in Table 5 of Young et al., (2013) and Table 12 of Stevenson et al., (2013).

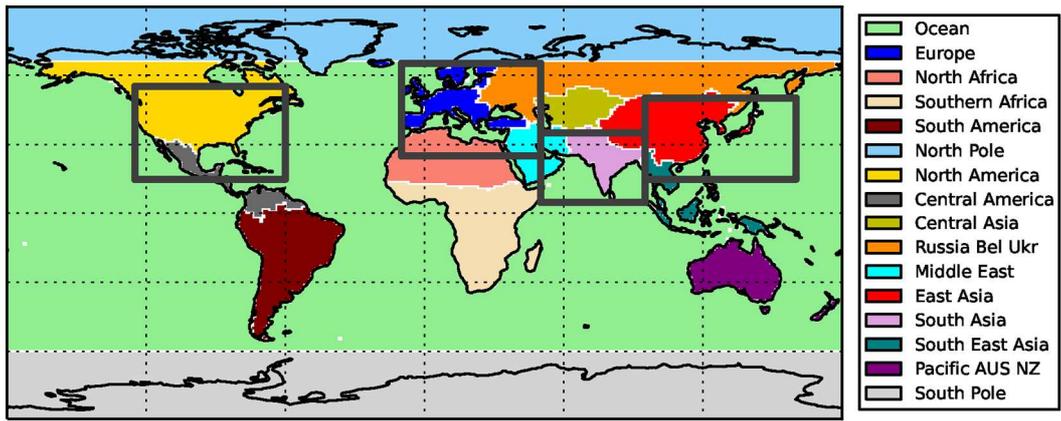
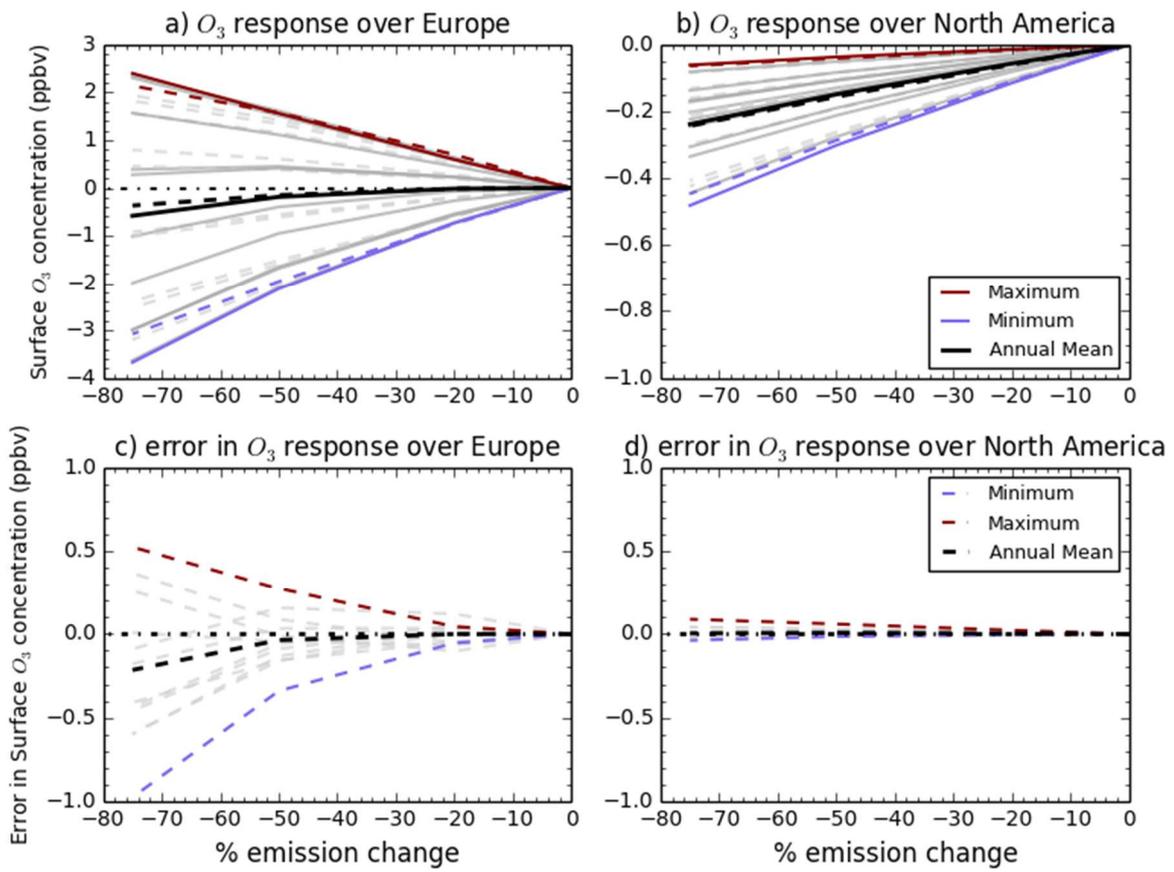


Figure 1: Source/Receptor regions used in TF-HTAP2 (coloured regions) and TF-HTAP1 (solid grey line boxes) experiments.

5



5 **Figure 2: Sensitivity of monthly surface O_3 changes in HadGEM2-ES (solid lines) and that of the parameterised response using solely HadGEM2-ES as input (dashed lines) to 20%, 50% and 75% reduction in all precursor emissions over the European source region (a) and the remote receptor region of North America (b). The difference between HadGEM2-ES and the parameterised response is shown over Europe (c) and North America (d). Annual mean values are in black with monthly responses in grey and the highest and lowest months are highlighted in red and blue.**

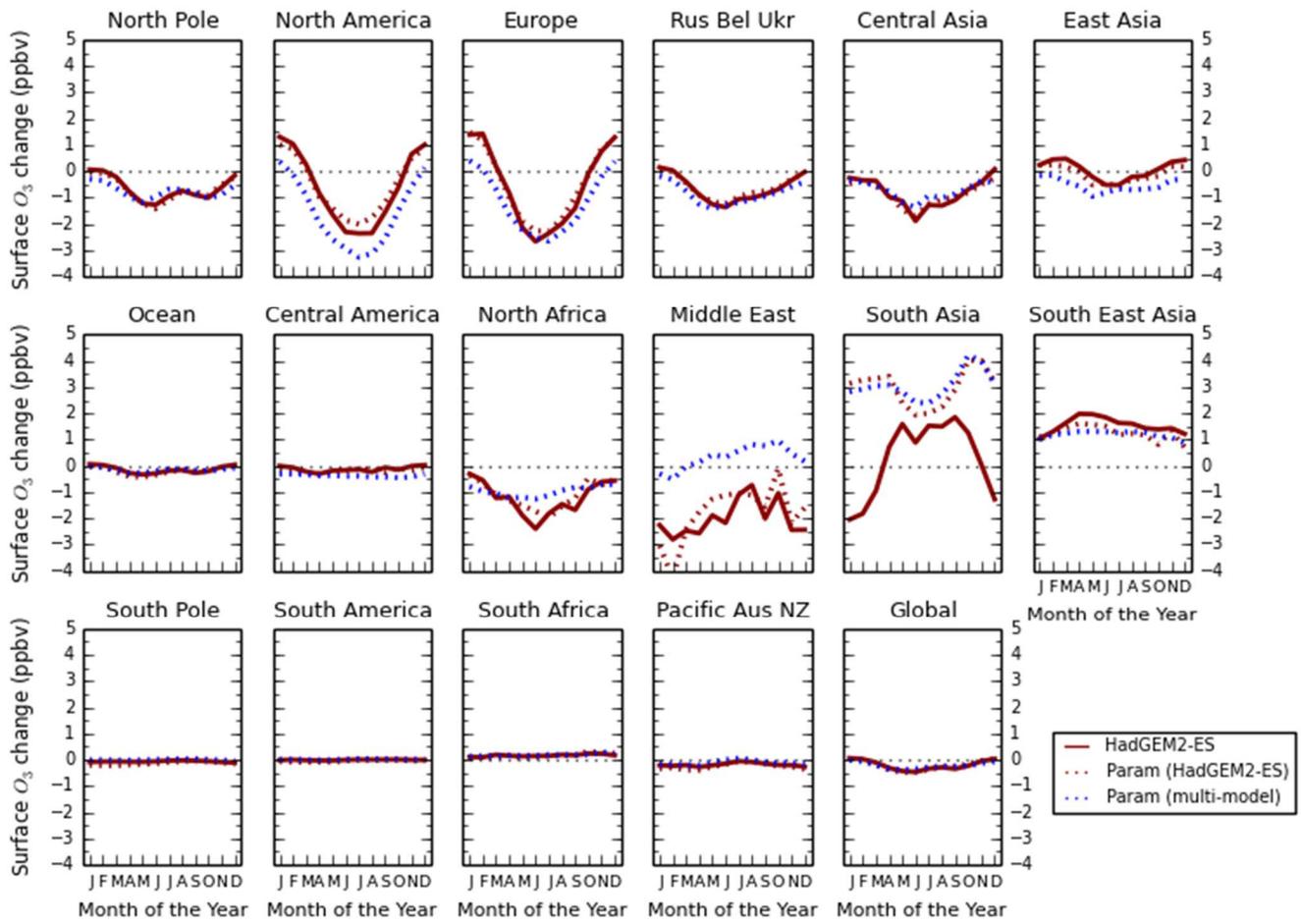


Figure 3: Monthly mean regional surface O₃ changes between 2010 and 2030 for the ECLIPSE V5a CLE scenario in HadGEM2-ES (red) and the parameterised response based on only HadGEM2-ES inputs (red dashed) and multiple model inputs (blue dashed).

5

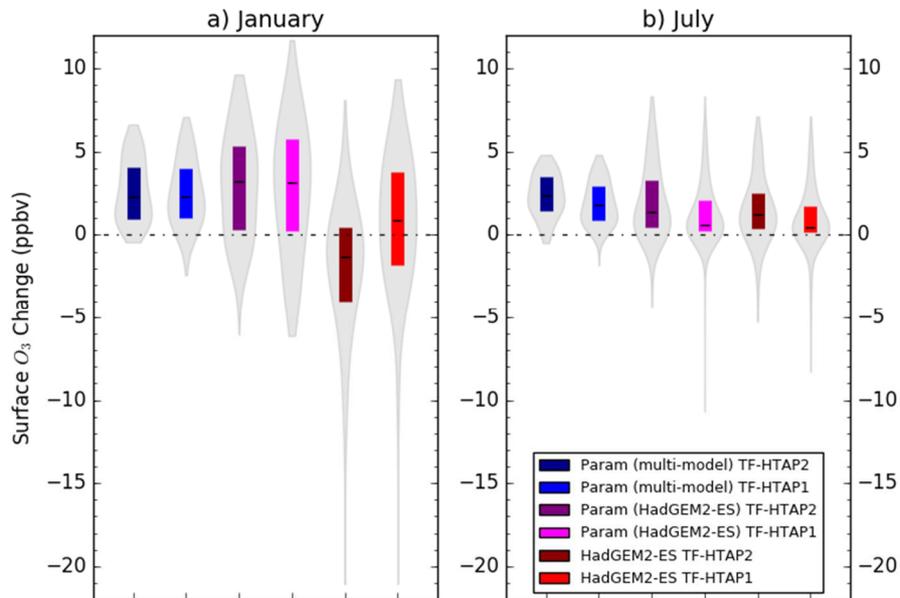
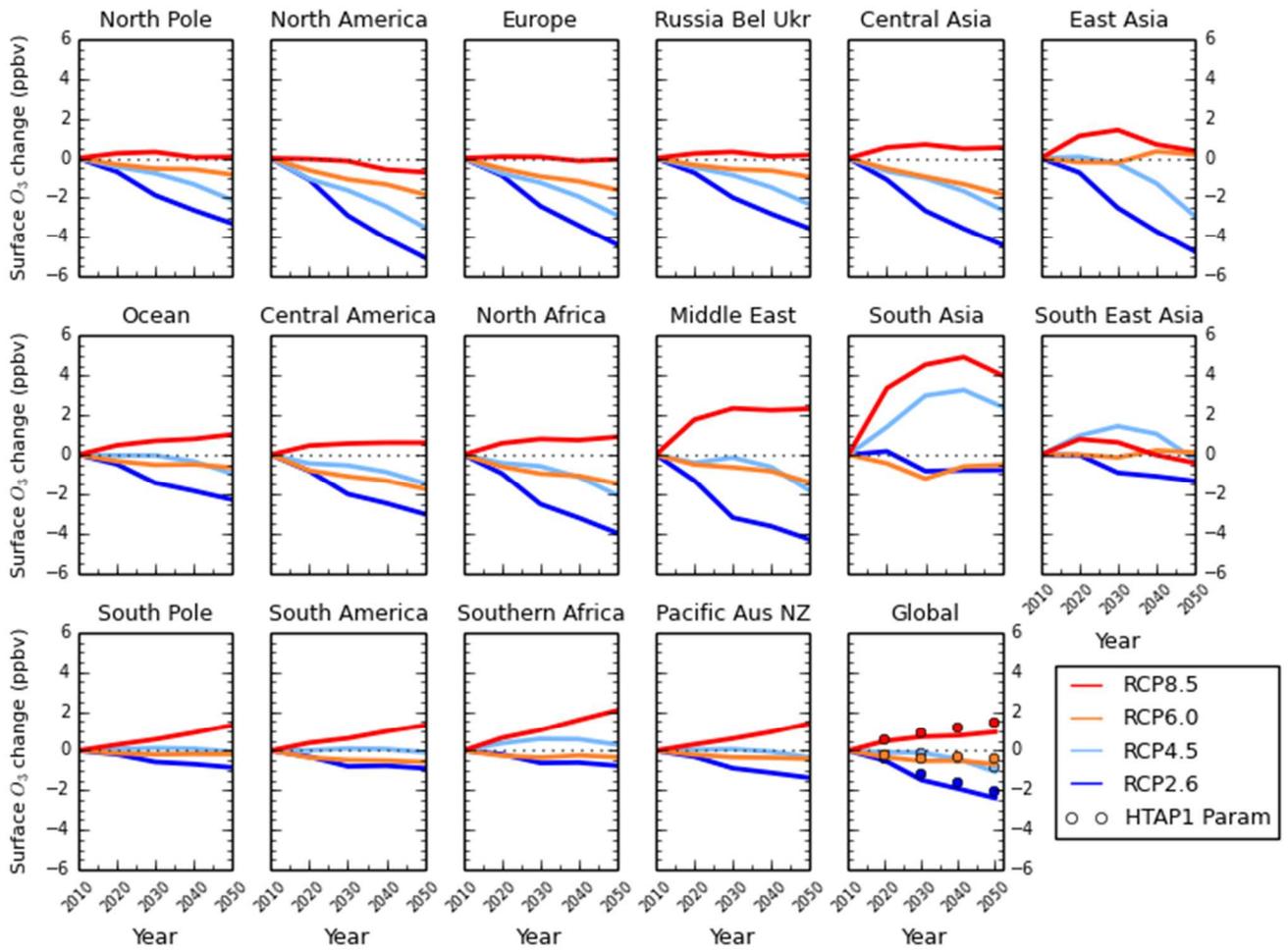


Figure 4: January (a) and July (b) monthly mean surface O_3 changes over South Asia between 2010 and the ECLIPSE V5a CLE emission scenario in 2030 for HadGEM2-ES and the parameterised response based only on HadGEM2-ES and on multiple models. O_3 responses are calculated over the South Asian region as defined in both TF-HTAP1 and TF-HTAP2 (Fig. 1). Grey shading represents the spatial distribution of O_3 changes across all grid boxes, coloured boxes show the range of the 25th to 75th percentile values and the solid line shows the median value over the South Asian region



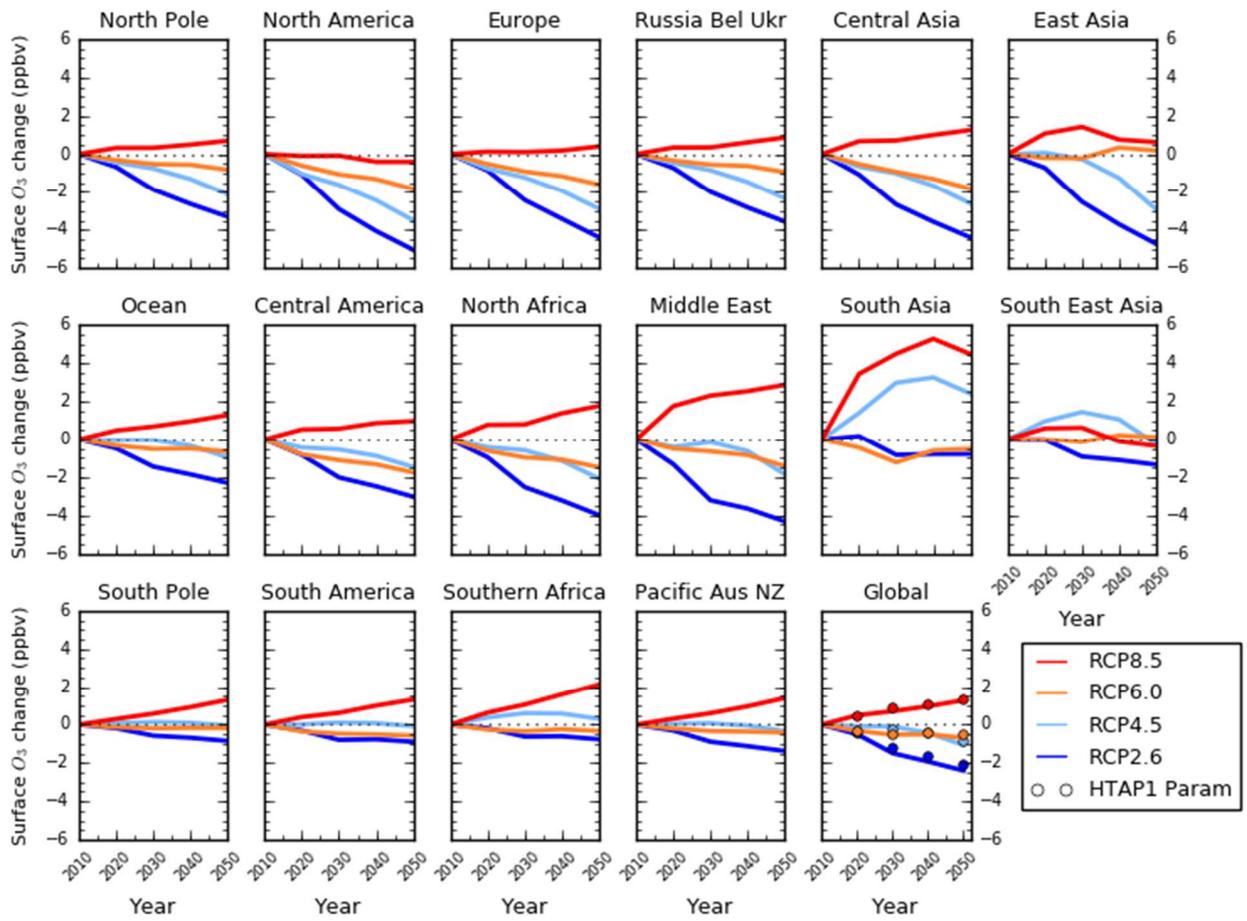


Figure 5: Annual mean regional surface O₃ changes between 2010 and 2050 from the parameterisation for the CMIP5 emissions scenarios of RCP8.5 (red), RCP6.0 (orange), RCP4.5 (light blue) and RCP2.6 (blue). The global surface O₃ response from the parameterisation of Wild et al., (2012) for each scenario is represented as circles, but due to differences in regional definitions a straightforward comparison with TF-HTAP1 regions (Europe, North America, South Asia and East Asia) is not possible.

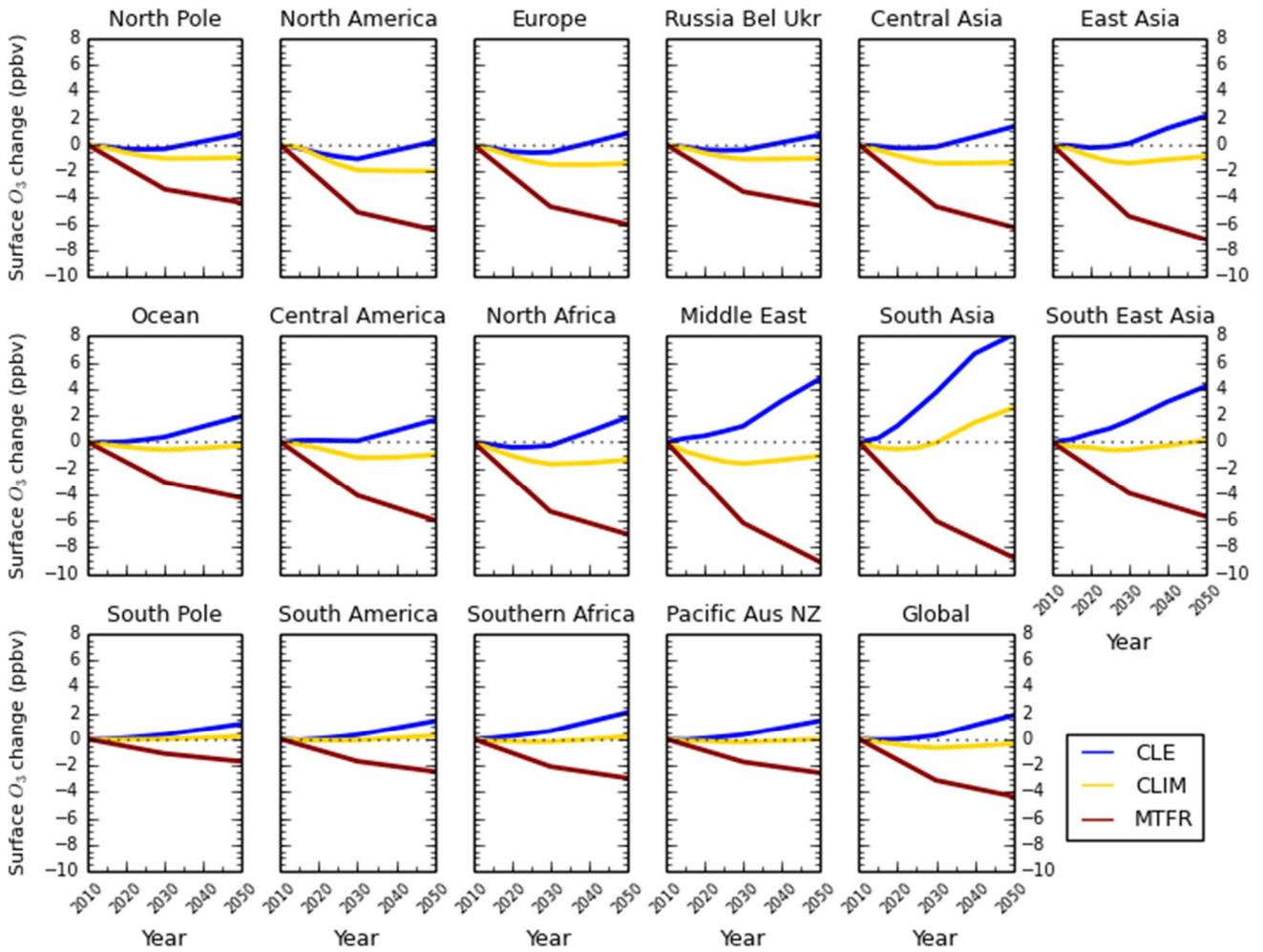
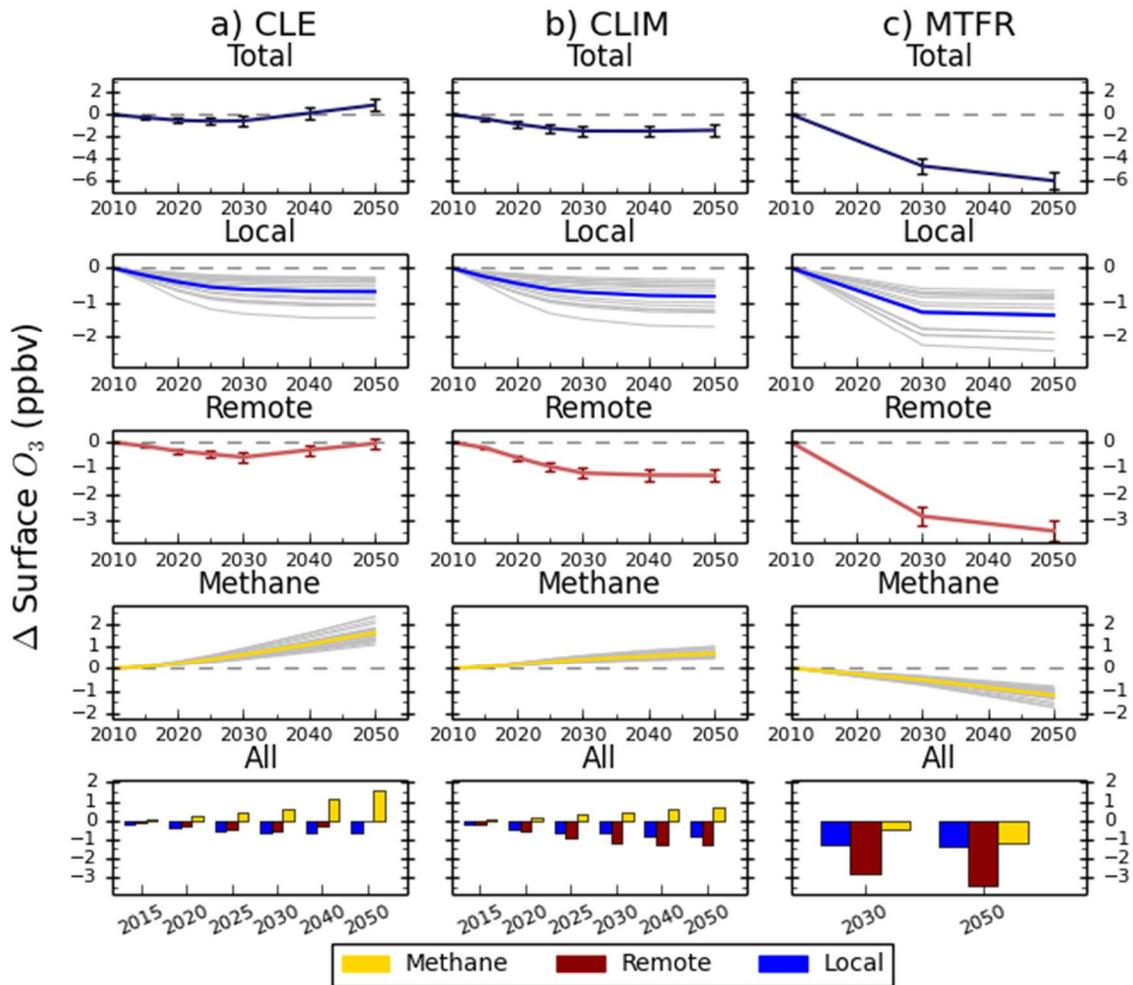


Figure 6: Annual mean change in regional surface O_3 concentrations between 2010 and 2050 from the parameterisation for the ECLIPSEv5a emissions under the CLE (blue), CLIM (gold) and MTRF (red) scenarios.



5 Figure 7: Total annual mean change in regional surface O_3 concentrations over Europe and the contribution of local (blue), remote (red) and methane (gold) sources between 2010 and 2050 from the parameterisation for the ECLIPSEv5a emissions under the CLE (a), CLIM (b) and MTRF (c) scenarios. Grey lines on the local and methane panels represent individual model estimates of O_3 changes, showing the spread in model responses; Solid lines show the multi-model mean. Error bars represent one standard deviation over the model range. The last row of panels shows the O_3 response from individual sources plotted together for each year.

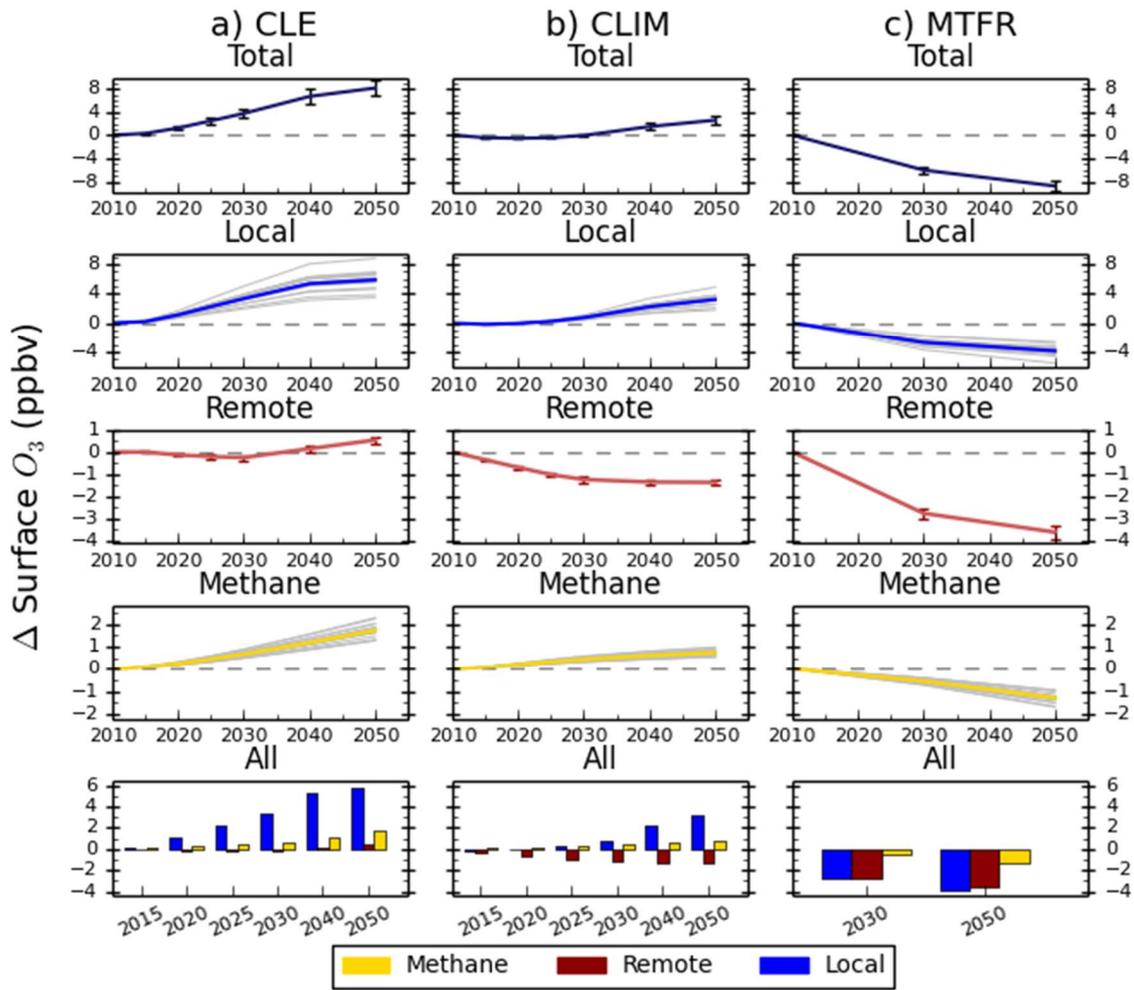
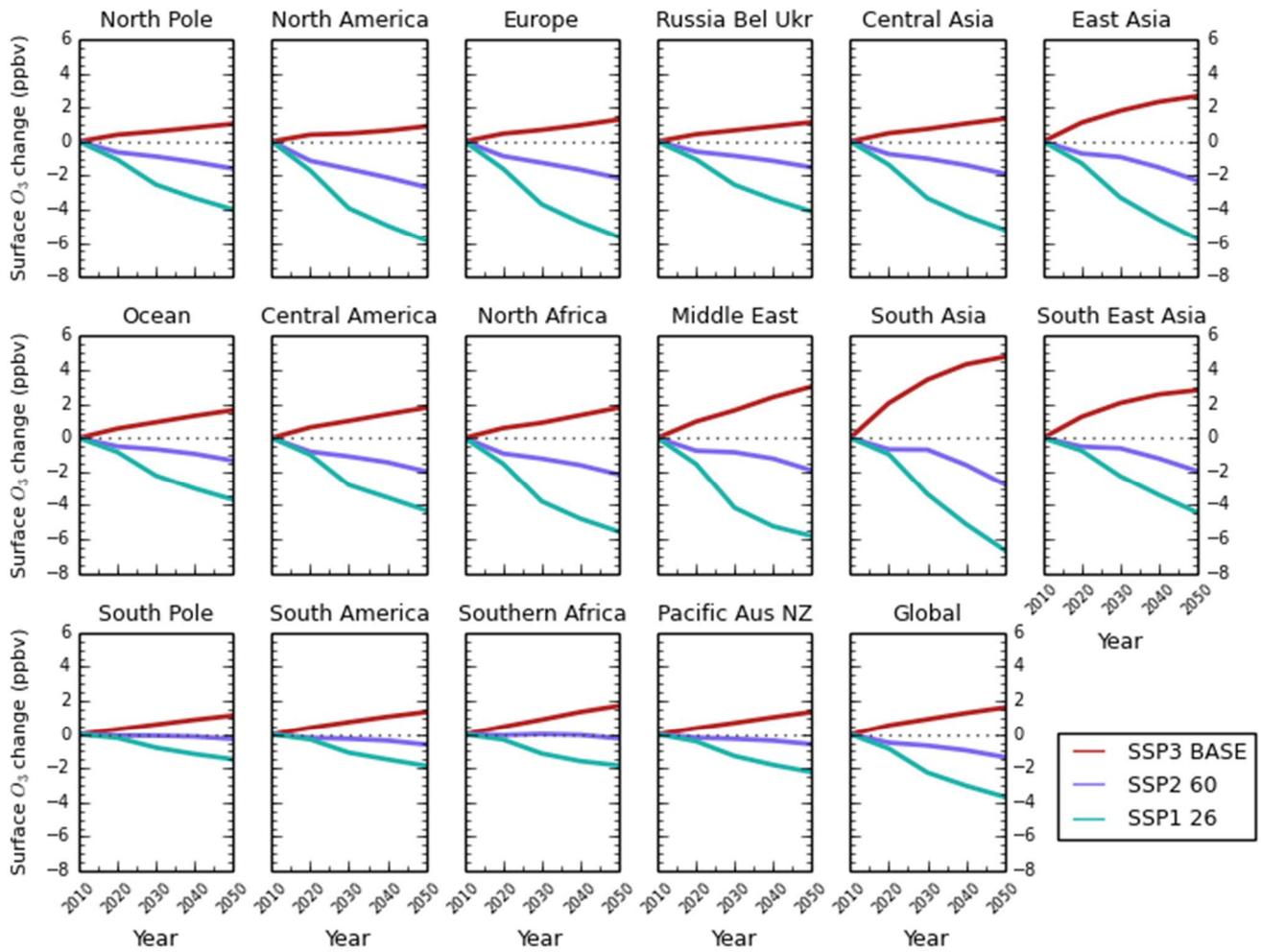
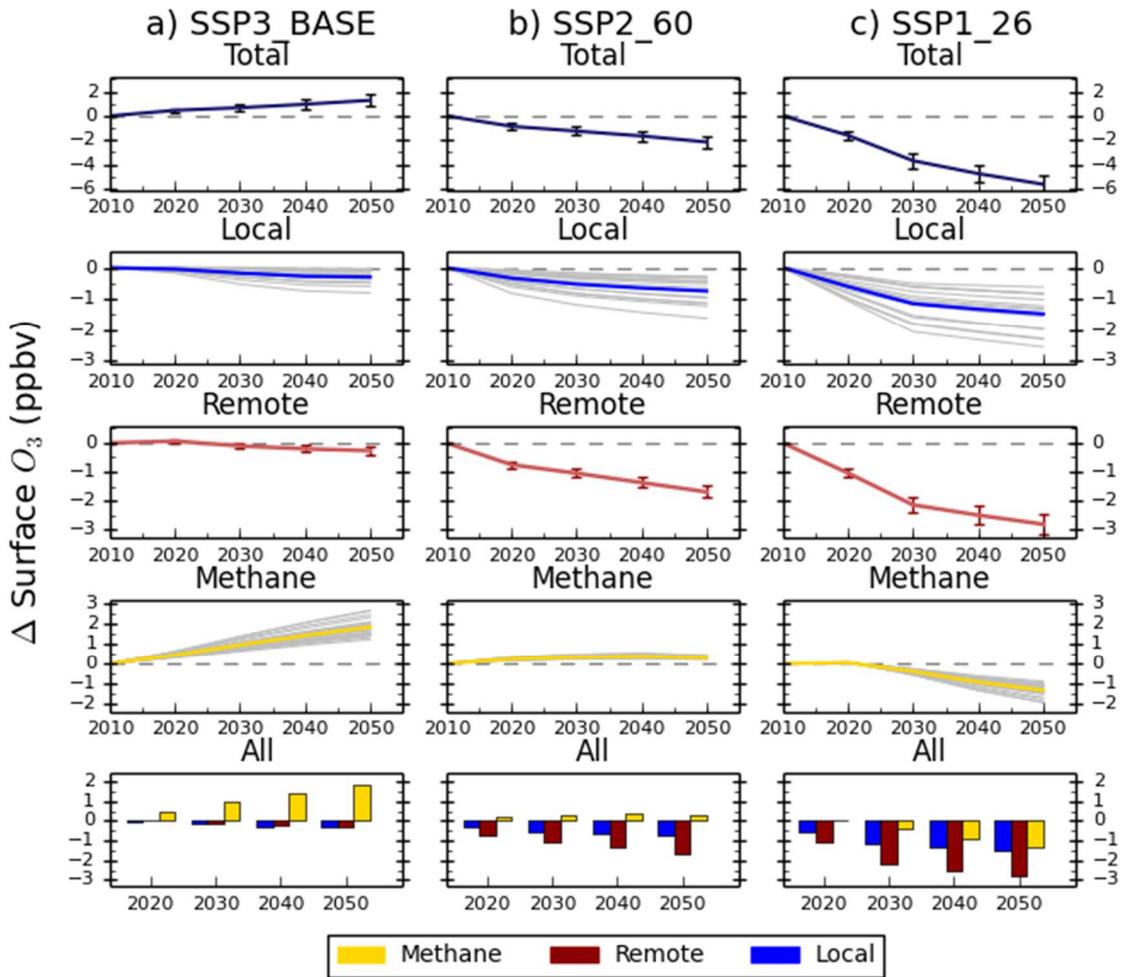


Figure 8: Same as Fig 7. but for the South Asian region.



5 **Figure 9: Annual mean change in regional surface O_3 concentrations between 2010 and 2050 from the parameterisation for the CMIP6 emissions scenarios of SSP3 baseline (red), SSP2 with a radiative forcing target of 6.0 W m^{-2} (purple) and SSP1 with a radiative forcing target of 2.6 W m^{-2} (green).**



5 **Figure 10:** Total annual mean change in regional surface O_3 concentrations over Europe and the contribution of local (blue), remote (red) or methane (gold) sources between 2010 and 2050 from the parameterisation for the CMIP6 emissions scenarios of SSP3 BASE (a), SSP2 6.0 (b) and SSP1 2.6 (c). Grey lines on the local and methane panels represent individual model estimates of O_3 changes, showing the spread in model responses; solid lines show the multi-model mean. Error bars represent one standard deviation over the model range. The last row of panels shows the O_3 response from individual sources plotted together for each year.

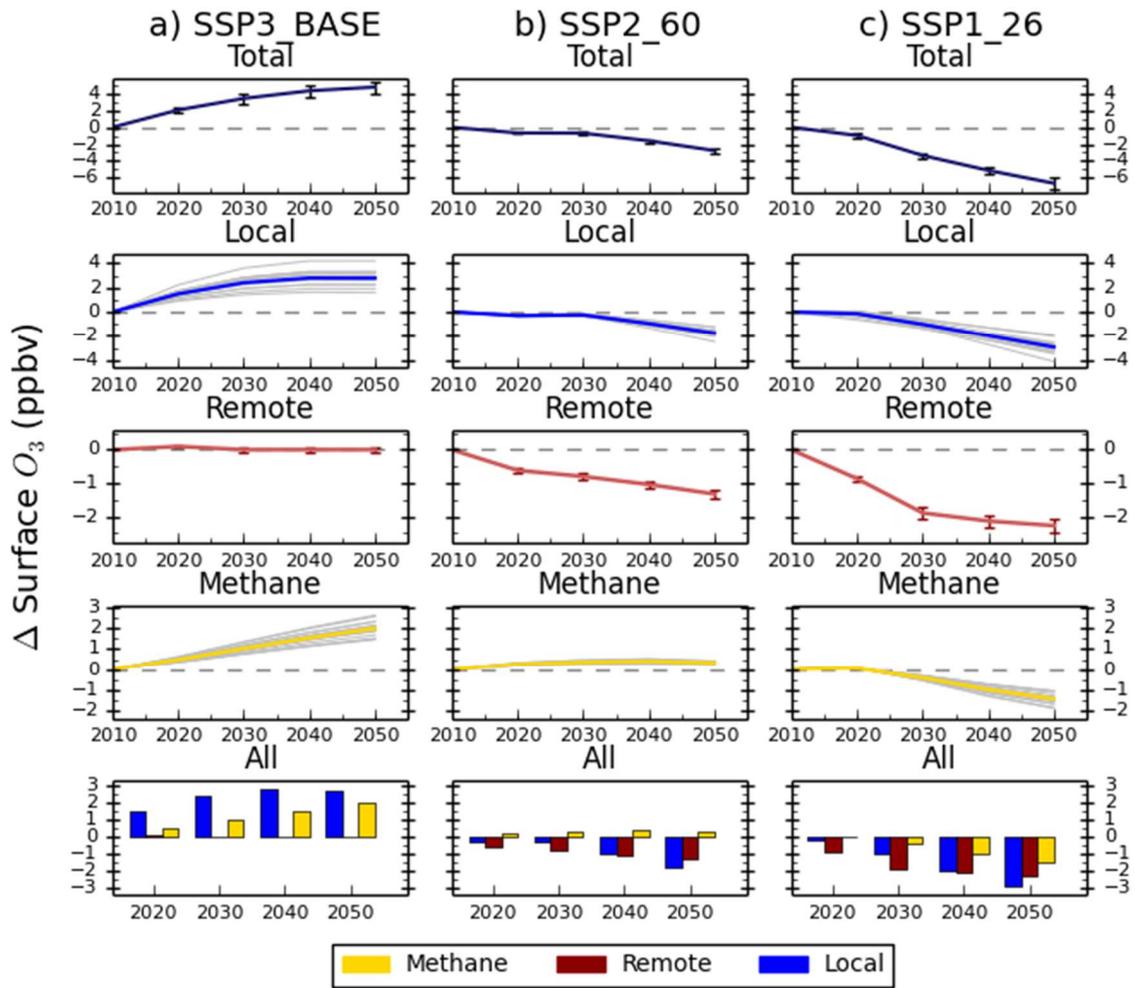


Figure 11: Same as Fig. 10 but for the South Asian region.

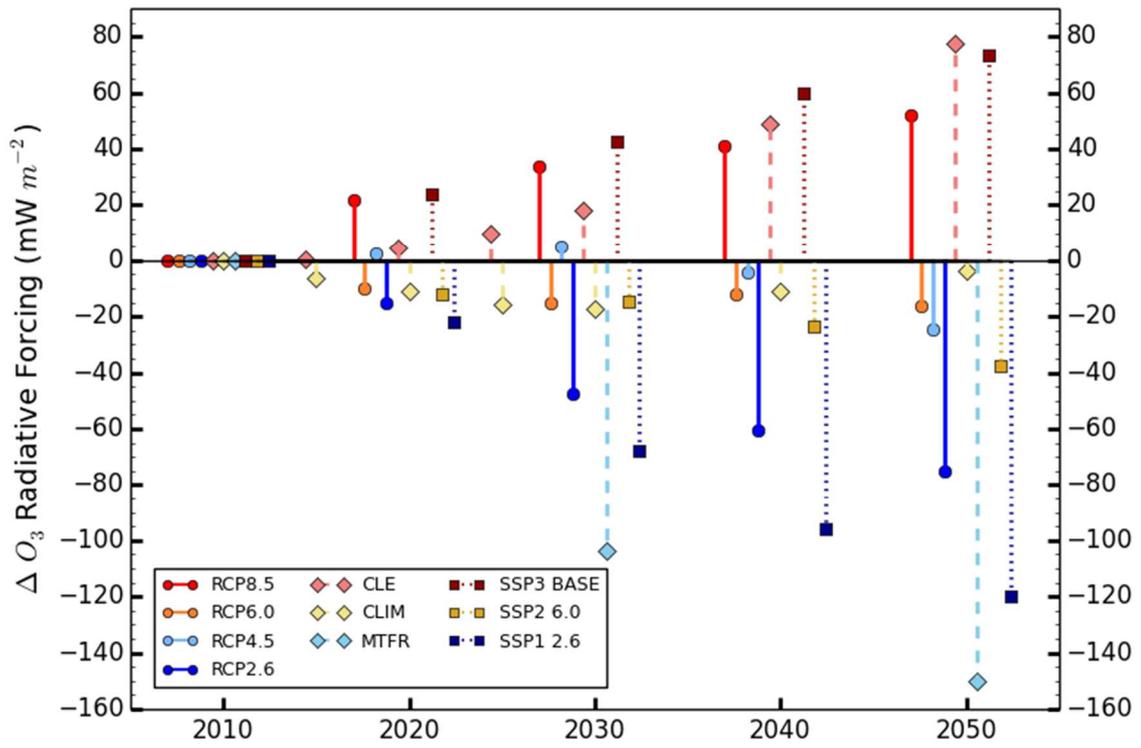


Figure 12. Parameterised response in the global annual mean ozone radiative forcing relative to 2010 for the different CMIP5 RCPs (circles), ECLIPSE (diamonds) and CMIP6 SSPs (squares).

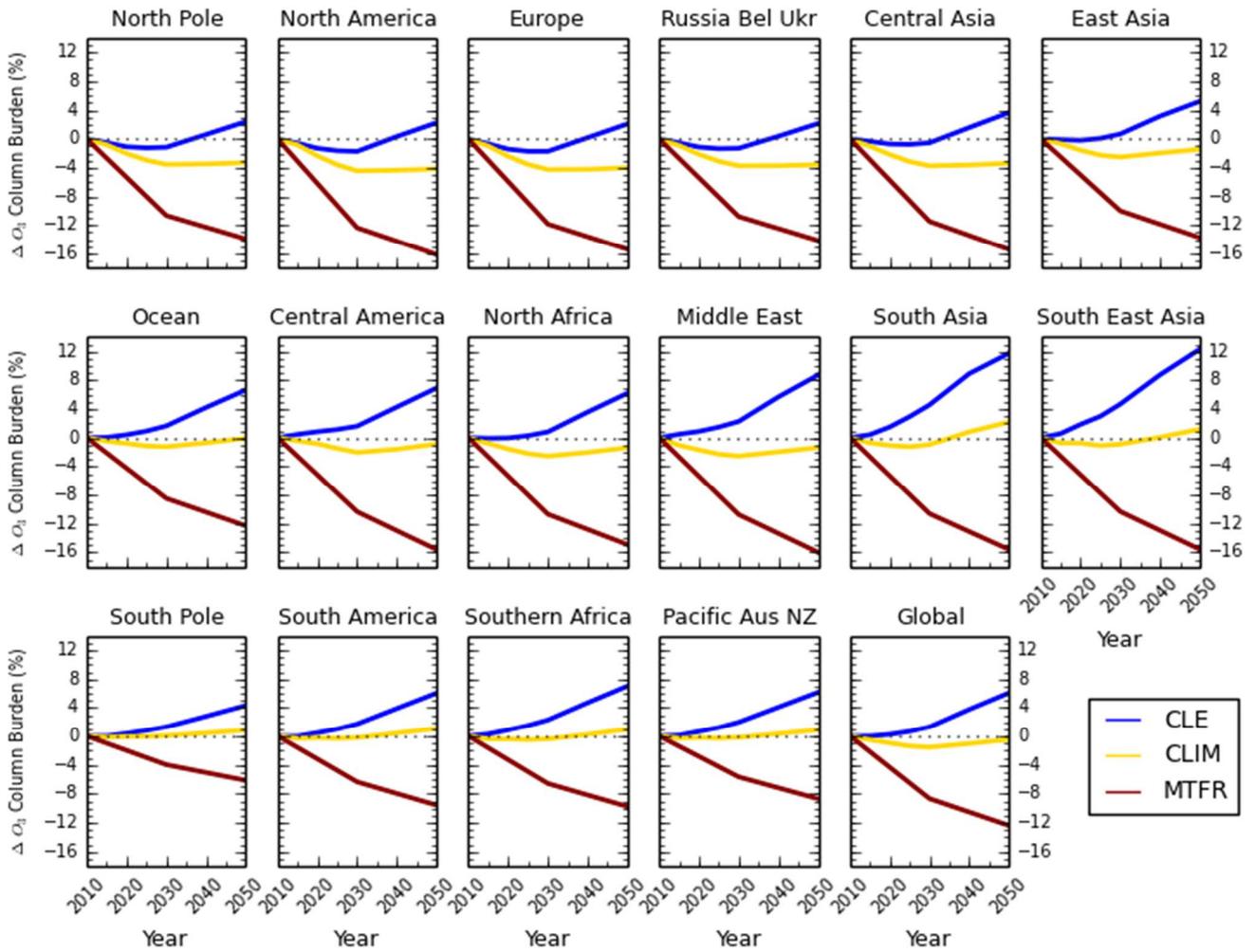


Figure 13: Annual mean percentage change in the regional and global tropospheric O₃ burden over the period 2010 to 2050 from the parameterisation for the ECLIPSEv5a emissions under CLE (blue), CLIM (gold) and MTRF (red) scenarios.

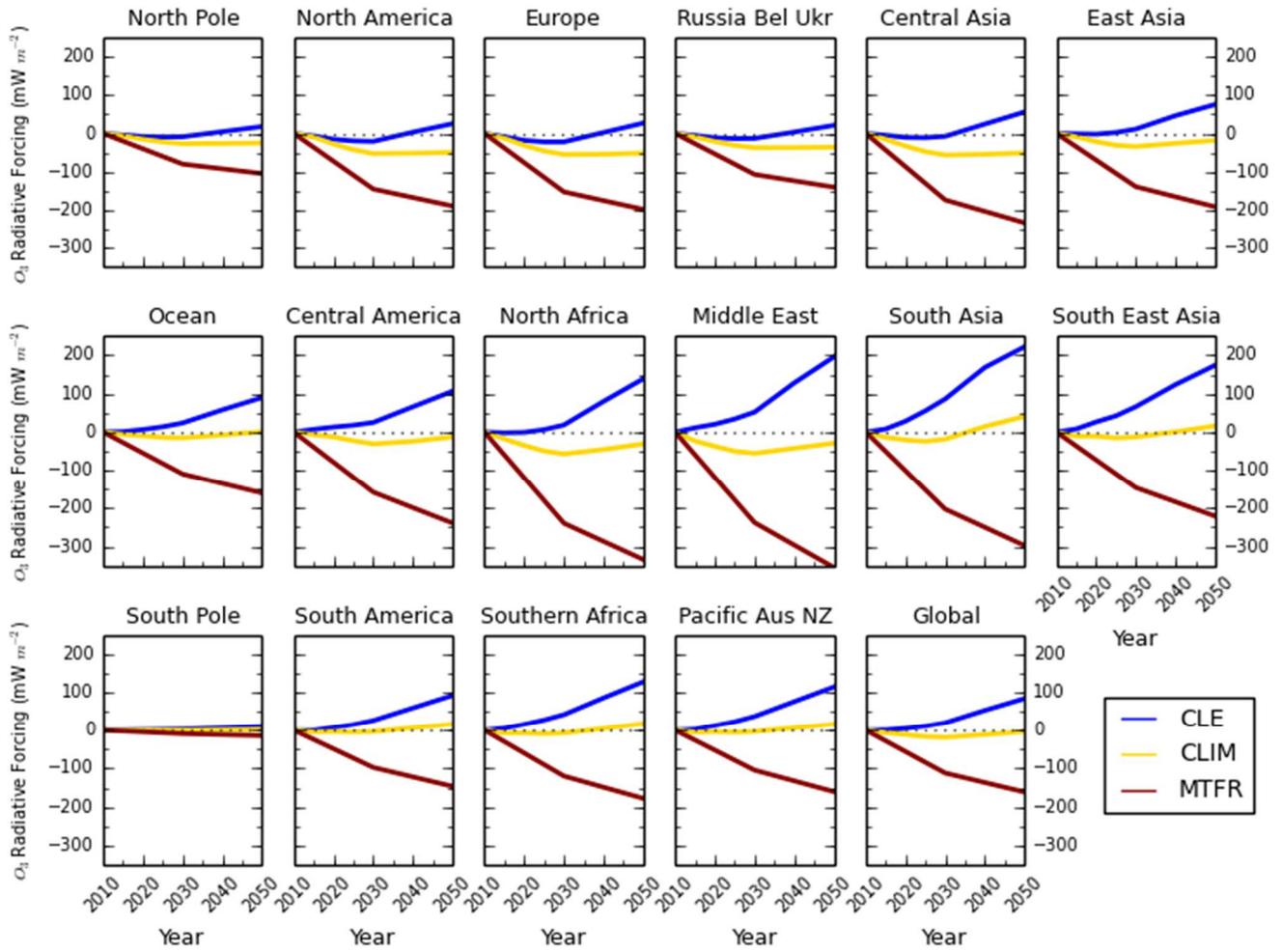


Figure 14: Annual mean regional O₃ radiative forcing relative to 2010 from the parameterisation for the ECLIPSEv5a emissions under the CLE (blue), CLIM (gold) and MTFR (red) scenarios.