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## LETTER

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A H Butler<sup>1,2</sup>, J S Daniel<sup>1</sup>, R W Portmann<sup>1</sup>, A R Ravishankara<sup>3</sup>, P J Young<sup>4</sup>, D W Fahey<sup>1</sup> and K H Rosenlof<sup>1</sup>

<sup>1</sup> National Oceanic and Atmospheric Administration/Earth Systems Research Laboratory/Chemical Sciences Division, Boulder, CO, USA

<sup>2</sup> Cooperative Institute for Research in Environmental Sciences, University of Colorado-Boulder, Boulder, CO, USA

<sup>3</sup> Departments of Chemistry and Atmospheric Science, Colorado State University, Fort Collins, CO, USA

<sup>4</sup> Lancaster University, Lancaster, UK

E-mail: [amy.butler@noaa.gov](mailto:amy.butler@noaa.gov)

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### Abstract

Due to the success of the Montreal Protocol in limiting emissions of ozone-depleting substances, concentrations of atmospheric carbon dioxide, nitrous oxide, and methane will control the evolution of total column and stratospheric ozone by the latter half of the 21st century. As the world proceeds down the path of reducing climate forcing set forth by the 2015 Conference of the Parties to the United Nations Framework Convention on Climate Change (COP 21), a broad range of ozone changes are possible depending on future policies enacted. While decreases in tropical stratospheric ozone will likely persist regardless of the future emissions scenario, extratropical ozone could either remain weakly depleted or even increase well above historical levels, with diverse implication for ultraviolet (UV) radiation. The ozone layer's dependence on future emissions of these gases creates a complex policy decision space for protecting humans and ecosystems, which includes unexpected options such as accepting nitrous oxide emissions in order to maintain historical column ozone and surface UV levels.

## 1. Introduction

Now that the world has agreed to curtail global warming following the 2015 Conference of the Parties to the United Nations Framework Convention on Climate Change (COP 21) in Paris, implementation requires emissions reductions of various greenhouse gases, including carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O). These gases not only warm the Earth, but also affect ozone abundances (WMO (World Meteorological Organization) 2014). Consequently, changes in these gas emissions and their simultaneous impacts on climate and the ozone layer become important considerations for future policy decisions.

The stratospheric ozone layer protects Earth's surface from the most damaging ultraviolet (UV) radiation. It has been substantially depleted over the last 35 years due to halocarbon emissions, which are both potent greenhouse gases and ozone-depleting substances (ODSs). An undisputed environmental policy-

making success story, the Montreal Protocol and its amendments will return ODSs to near historical levels (1955–1975, prior to significant depletion) by the latter half of this century and prevent significant health consequences of excessive UV exposure (Newman and McKenzie 2011, Chipperfield *et al* 2015). However, the evolution of stratospheric ozone beyond the middle of the 21st century, when ODS emissions are minimal and their atmospheric concentrations continue to decrease, will largely be determined by CO<sub>2</sub>, N<sub>2</sub>O, and CH<sub>4</sub> (Oman *et al* 2010, Fleming *et al* 2011, Portmann *et al* 2012, Revell *et al* 2012, Eyring *et al* 2013, Stolarksi *et al* 2015, Iglesias-Suarez *et al* 2016). Under some future emission scenarios, extratropical ozone far exceeds historical levels, reducing surface UV below their historical levels (Hegglin and Shepherd 2009, Watanabe *et al* 2011, Bais *et al* 2015).

N<sub>2</sub>O destroys stratospheric ozone by increasing reactive odd-nitrogen species. N<sub>2</sub>O is currently the largest ozone-depletion-potential (ODP)-weighted emission (Ravishankara *et al* 2009) and a potent

**Table 1.** Concentrations of CO<sub>2</sub>, N<sub>2</sub>O, CH<sub>4</sub>, and Cl<sub>y</sub> used in each time-slice simulation, and the climatology period for sea surface temperatures (SSTs)/sea ice. Concentrations are from either the year 1957 (historical) or 2095 (future runs) RCPs.

Experiment	CO <sub>2</sub> (ppm)	N <sub>2</sub> O (ppb)	CH <sub>4</sub> (ppb)	Cl <sub>y</sub> (ppb)	SSTs/Ice climatology
1957 historical	314	291	1211	0.73	1955–1975
RCP 2.6		344	1259		
RCP26_N2O_85	423	<b>428</b>	1259	1.19	RCP 2.6 2075–2095
RCP26_CH4_85		344	<b>3698</b>		
RCP 4.5	535	371	1591	1.16	RCP 4.5 2075–2095
RCP 8.5		428	3698		
RCP85_N2O_26	889	<b>344</b>	3698	1.11	RCP 8.5 2075–2095
RCP85_CH4_26		428	<b>1259</b>		

greenhouse gas; its potential mitigation via the Montreal Protocol has been examined (Kanter *et al* 2013). The ODP of N<sub>2</sub>O changes with CO<sub>2</sub> and CH<sub>4</sub> abundances (Ravishankara *et al* 2009, Wang *et al* 2014, Revell *et al* 2015). The dominant effect of increased CH<sub>4</sub> is increased ozone production in the lower stratosphere and troposphere down to the surface (Fleming *et al* 2011, Eyring *et al* 2013). Increased CO<sub>2</sub> cools the stratosphere and consequently increases ozone particularly in the upper stratosphere (Fleming *et al* 2011). Increased CO<sub>2</sub> is expected to accelerate the Brewer-Dobson Circulation (BDC), leading to stratospheric ozone decreases in the tropics and increases in the extratropics (Butchart 2014). While CH<sub>4</sub> and N<sub>2</sub>O affect ozone predominantly via chemical reactions, CO<sub>2</sub> affects ozone indirectly via temperature and dynamical changes. Although N<sub>2</sub>O, CH<sub>4</sub>, and CO<sub>2</sub> all exacerbate climate change, they have different, and possibly non-linear (Meul *et al* 2015), influences on both total column and stratospheric column ozone.

The Montreal Protocol's regulations to mitigate ODSs are expected to reduce their radiative forcing while increasing global stratospheric ozone back towards historical levels. Conversely, protecting the future ozone layer under evolving CO<sub>2</sub>, N<sub>2</sub>O, and CH<sub>4</sub> emissions may involve the conundrum of weighing benefits for climate change against maintaining the ozone layer and surface UV radiation near historical levels.

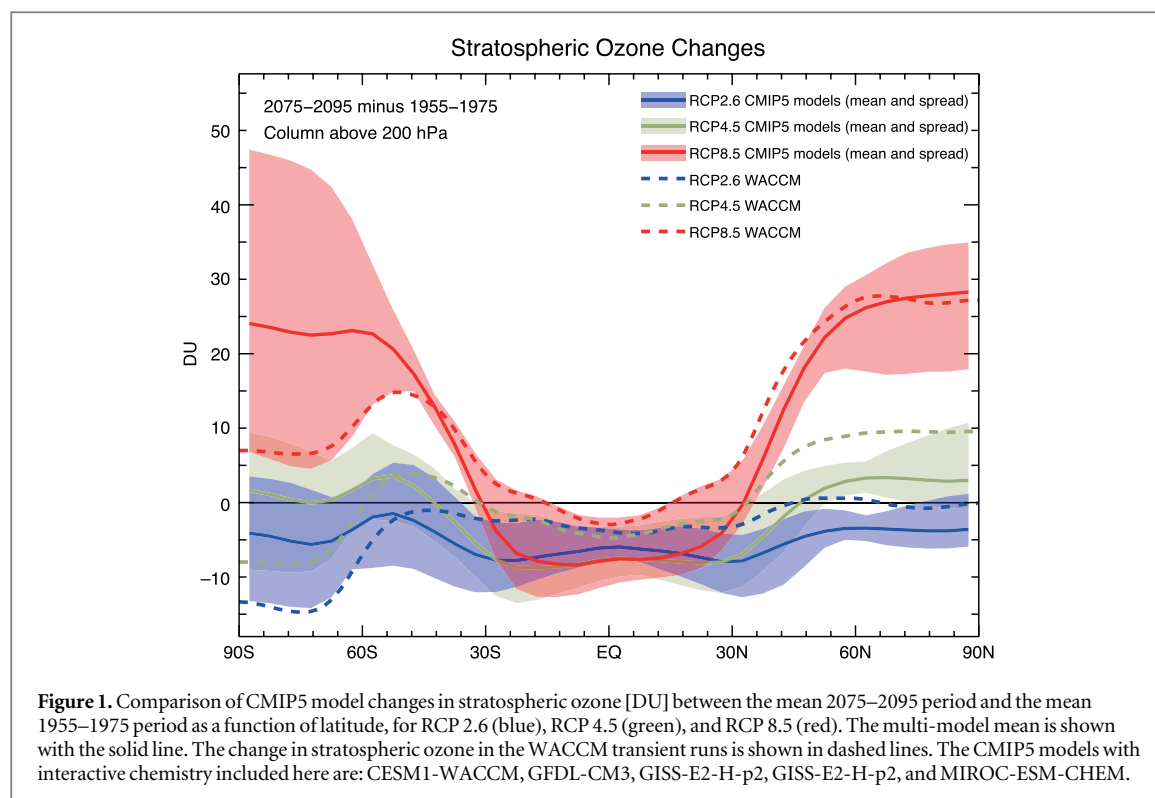
## 2. Methods

To demonstrate the complexities in future ozone changes when N<sub>2</sub>O or CH<sub>4</sub> concentrations are increased or reduced in climates with varying CO<sub>2</sub>, we ran simulations using the National Center for Atmospheric Research (NCAR) Whole Atmosphere Community Climate Model (WACCM) for the Representative Concentration Pathways (RCPs), RCP 2.6, 4.5, and 8.5. WACCM version 4 (Marsh *et al* 2013) is an atmospheric extension to NCAR's Community Earth System Model (CESM) with a high model top (~140 km) and fully interactive chemistry in the

middle atmosphere, though tropospheric chemistry has a limited representation CH<sub>4</sub> and carbon monoxide oxidation). The horizontal resolution is 1.9 degrees latitude by 2.5 degrees longitude with 66 vertical levels. The quasi-biennial oscillation is represented by nudging the tropical winds to match observed interannual variability.

A historical transient simulation from 1955–2005 was performed using time-evolving observed forcings (surface concentrations of radiatively active species, daily solar spectral irradiance, and volcanic sulfates). The ocean was initialized from a reference case and allowed to freely evolve. Future RCP transient simulations were run from 2006–2095 using projected concentrations of radiatively active species (Meinshausen *et al* 2011). From these transient simulations, we used the sea surface temperature (SST) and sea ice climatologies for both the historical (1955–1975) and the future (2075–2095) climates to force 20-year ‘time-slice’ experiments (following 10 years of spin-up) with varying concentrations of N<sub>2</sub>O, CH<sub>4</sub>, and CO<sub>2</sub> (table 1) based on the RCP scenarios. Time-slice runs simulate ‘slices’ of time rather than the full transient response; the atmosphere responds to fixed climatological SSTs and forcings held at a constant value for a particular year. In runs where we modulate N<sub>2</sub>O or CH<sub>4</sub> concentrations, we assume that the associated changes in SSTs/sea ice are not large (i.e., they are dominated by the changes in CO<sub>2</sub>).

For comparison with values in table 1, 2016 global concentrations of CO<sub>2</sub>, N<sub>2</sub>O, and CH<sub>4</sub> are ~402 ppm, ~328 ppb, and ~1840 ppb, respectively. Note that in the end-of-the-century RCP 8.5 climate, CO<sub>2</sub> and CH<sub>4</sub> have increased substantially (roughly a factor of three), while N<sub>2</sub>O has increased only by ~50%, compared to the historical climate. Total chlorine (Cl<sub>y</sub>) is higher for all future climates compared to Cl<sub>y</sub> in the historical run because the long-lived chlorofluorocarbons persist. Cl<sub>y</sub> concentrations, aerosols, and tropospheric emission precursor concentrations in the time-slice experiments are based on corresponding historical or RCP scenario values, but we performed an additional sensitivity run to test the role of Cl<sub>y</sub> in the RCP 8.5



scenario (not shown). Solar spectral irradiances (SSI), specified from the Lean *et al* (2005) model and used for solar heating and photolysis reactions, vary on daily timescales in the transient runs, but are set constant in the historical time-slice run to 1960 values and in the future time-slice runs to 2095 values. The SSI integrated over all wavelengths is the total spectral irradiance (TSI); for comparison, the TSI is  $\sim 1362 \text{ W m}^{-2}$  in 1960 and  $\sim 1361 \text{ W m}^{-2}$  in 2095. For solar and geomagnetic parameters (e.g., the F 10.7 cm flux,  $K_p$ ,  $A_p$ ), a constant value (the solar cycle average) was used in all time-slice runs.

We also consider changes in stratospheric ozone in coupled climate models with interactive chemistry from the Fifth Coupled Model Intercomparison Project (CMIP5) (Taylor *et al* 2012). Stratospheric column ozone is determined by summing ozone above 200 hPa (more complex methods using a latitudinally-varying tropopause yielded similar results).

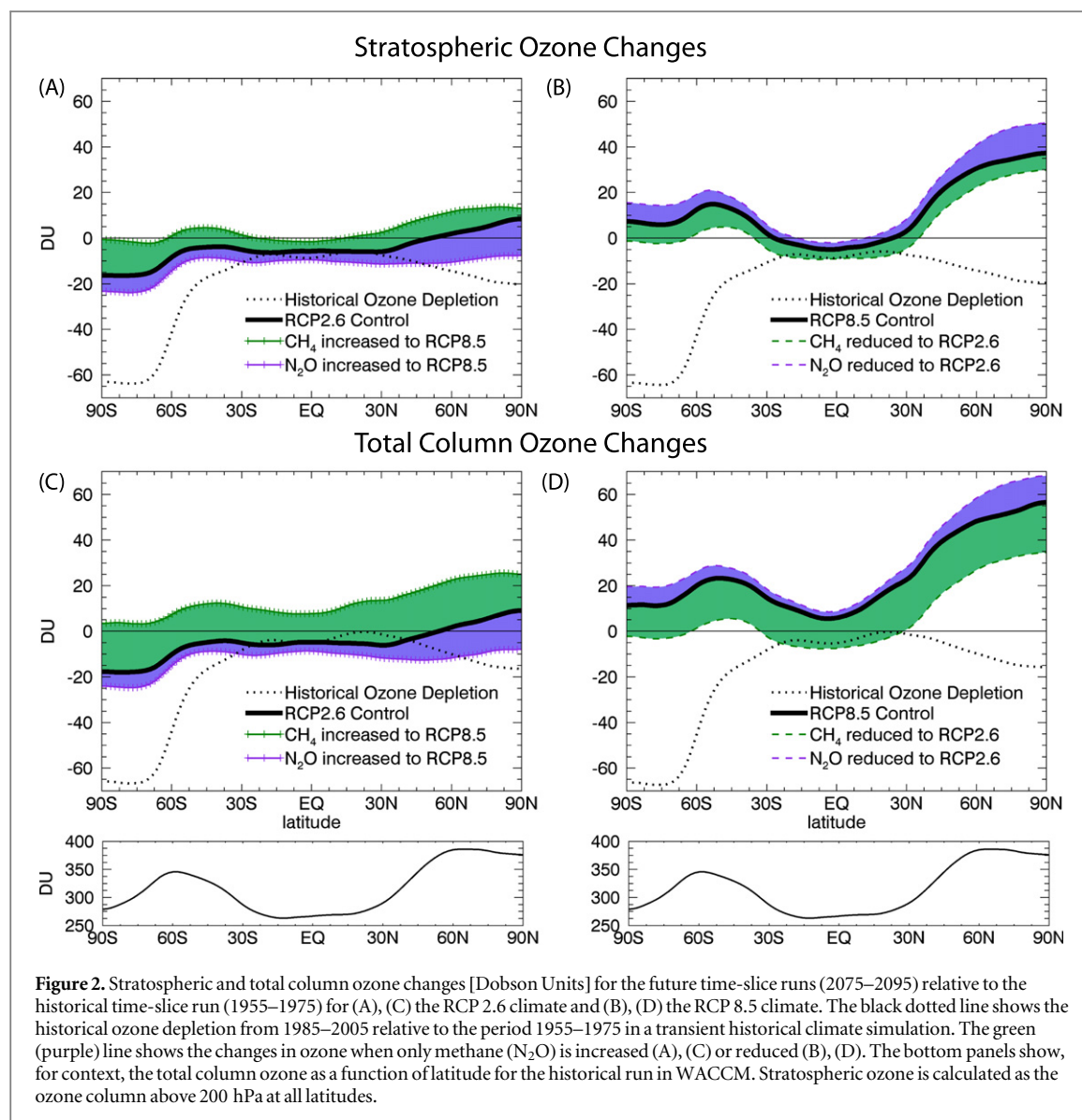
### 3. Results

In agreement with previous studies (Oman *et al* 2010, Fleming *et al* 2011, Portmann *et al* 2012, Revell *et al* 2012, Eyring *et al* 2013, Stolarski *et al* 2015, Iglesias-Suarez *et al* 2016), there is a broad range of possible changes to the stratospheric ozone layer by the end of the century (2075–2095) compared to the historical period (1955–1975) (shown in figure 1 for CMIP5 models with interactive chemistry). In an aggressive greenhouse gas mitigation scenario (RCP 2.6; blue shading), stratospheric ozone at most

latitudes remains slightly below historical levels by the end of the 21st century. The  $\sim 5$ – $10$  Dobson Unit ozone depletion is due to slightly elevated  $\text{N}_2\text{O}$  and anthropogenic halocarbons remaining in the atmosphere.

However, under continued greenhouse gas emissions (RCP 8.5; red shading), stratospheric ozone remains depleted in the tropics but substantially exceeds historical levels in the extratropics. Lower tropical stratospheric ozone relative to the historical period is mostly due to the accelerated BDC and partly due to  $\text{N}_2\text{O}$ -related loss processes (Fleming *et al* 2011, Eyring *et al* 2013). The extratropical ozone increases are primarily associated with the cooling effects of  $\text{CO}_2$  in the mid- to upper stratosphere, the increased advection of ozone-rich air via the strengthened BDC, and enhanced production due to increased  $\text{CH}_4$  (Fleming *et al* 2011). In a moderate emission scenario (RCP 4.5; green shading), stratospheric ozone remains depleted in the tropics, but extratropical ozone returns to near historical levels, due to a balance between increases caused by  $\text{CO}_2$  and  $\text{CH}_4$ , and depletion caused by ODSs and  $\text{N}_2\text{O}$ . The changes in stratospheric ozone from the historical period (1955–1975) to the end of the century (2075–2095) in our WACCM RCP 2.6, 4.5, and 8.5 transient runs (dashed lines) are largely consistent with the CMIP5 simulations, with the exception of weaker ozone recovery in the Southern Hemisphere extratropics and greater ozone recovery in the Northern Hemisphere extratropics (figure 1; dashed lines).

Time-slice experiments were forced using SST and sea ice climatologies from the transient simulations



and the concentrations of  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{CH}_4$ , and total Cly shown in table 1. Here we focus on the RCP 2.6 and 8.5 scenarios, since the stratospheric ozone response to RCP 4.5 falls in between these extreme scenarios (figure 1). For the RCP 2.6 future time-slice run (figures 2(A), (C); bold black line), ozone is slightly below historical levels at most latitudes by the end of the 21st century in agreement with the transient simulation (figure 1). We can then examine what would happen if either  $\text{CH}_4$  or  $\text{N}_2\text{O}$  were not aggressively mitigated. If  $\text{CH}_4$  were increased to RCP 8.5 concentrations for 2095, column ozone increases at all latitudes due to large tropospheric ozone increases (figures 2(A), (C); green shading). If  $\text{N}_2\text{O}$  were increased to RCP 8.5 concentrations for 2095, ozone decreases at all latitudes due to stratospheric ozone depletion (figures 2(A), (C); purple shading). Note that the tropical ozone depletion in this scenario would potentially be larger than during the period of maximum historic ozone layer depletion (1985–2005, dotted line). A key point is that if the world were to

achieve reductions of  $\text{CO}_2$  and  $\text{CH}_4$  concentrations to RCP 2.6 levels,  $\text{N}_2\text{O}$  mitigation would become important to avoid exacerbation of both climate change and ozone layer depletion. On the other hand, if  $\text{CO}_2$  and  $\text{N}_2\text{O}$  were reduced to RCP 2.6 levels but  $\text{CH}_4$  concentrations increased, we expect stratospheric ozone increases towards historical levels and large increases in global tropospheric ozone.

In contrast, modulating  $\text{N}_2\text{O}$  or  $\text{CH}_4$  in a climate with high greenhouse gas concentrations has different consequences for the total and stratospheric column ozone. In the RCP 8.5 future time-slice run, stratospheric ozone remains depleted in the tropics but substantially exceeds historical levels in the extratropics (figure 2(B); bold black line). Large  $\text{CH}_4$  increases in the RCP 8.5 scenario (and thus large increases in tropospheric ozone) leads to increases in the total column ozone above historical levels at all latitudes (figure 2(D); bold black line). Note that increases in column ozone due to enhanced  $\text{CH}_4$  occurs more in the troposphere than in the stratosphere, as to be



expected. In this climate, a reduction in  $\text{N}_2\text{O}$  concentrations would reduce anthropogenic climate forcing and slightly reduce stratospheric ozone depletion at low latitudes, but would increase column ozone beyond historical levels in the extratropical stratosphere and at all latitudes (figures 2(B), (D); purple shading). A reduction in  $\text{CH}_4$  concentrations would also reduce anthropogenic climate forcing and tropospheric ozone, and thus decrease extratropical ozone levels towards historical levels, but could exacerbate tropical stratospheric ozone decreases (figures 2(B), (D); green shading).

There are many potential impacts of changes in tropospheric and stratospheric ozone on human health and the ecosystem. There are large uncertainties in tropospheric ozone due to uncertainties in tropospheric precursor emissions, but the changes simulated in WACCM fall within the CMIP5 multi-model spread (Eyring *et al* 2013, Young *et al* 2013). We assess the latitudinal changes in the UV index (UVI) at local noon using an approximation based on calculated total column ozone for cloud-free, aerosol-free, low-surface albedo conditions (Madronich 2007):

$$\text{UVI} \sim 12.5 \mu_o^{2.42} \left( \frac{\Omega}{300} \right)^{-1.23}, \quad (1)$$

where  $\mu_o$  is the cosine of the solar zenith angle at solar noon on the 15th of each month, and  $\Omega$  is the total column ozone. Note that the noon UVI ranges from zero at the poles to  $\sim 12$  at the equator, and varies substantially with latitude and season, with the highest values in the extratropical summer hemisphere. Thus a 20% change in UVI at  $45^\circ$  latitude represents absolute increases in the UVI of  $\sim 2$  in the summer but only  $\sim 0.5$  in the winter.

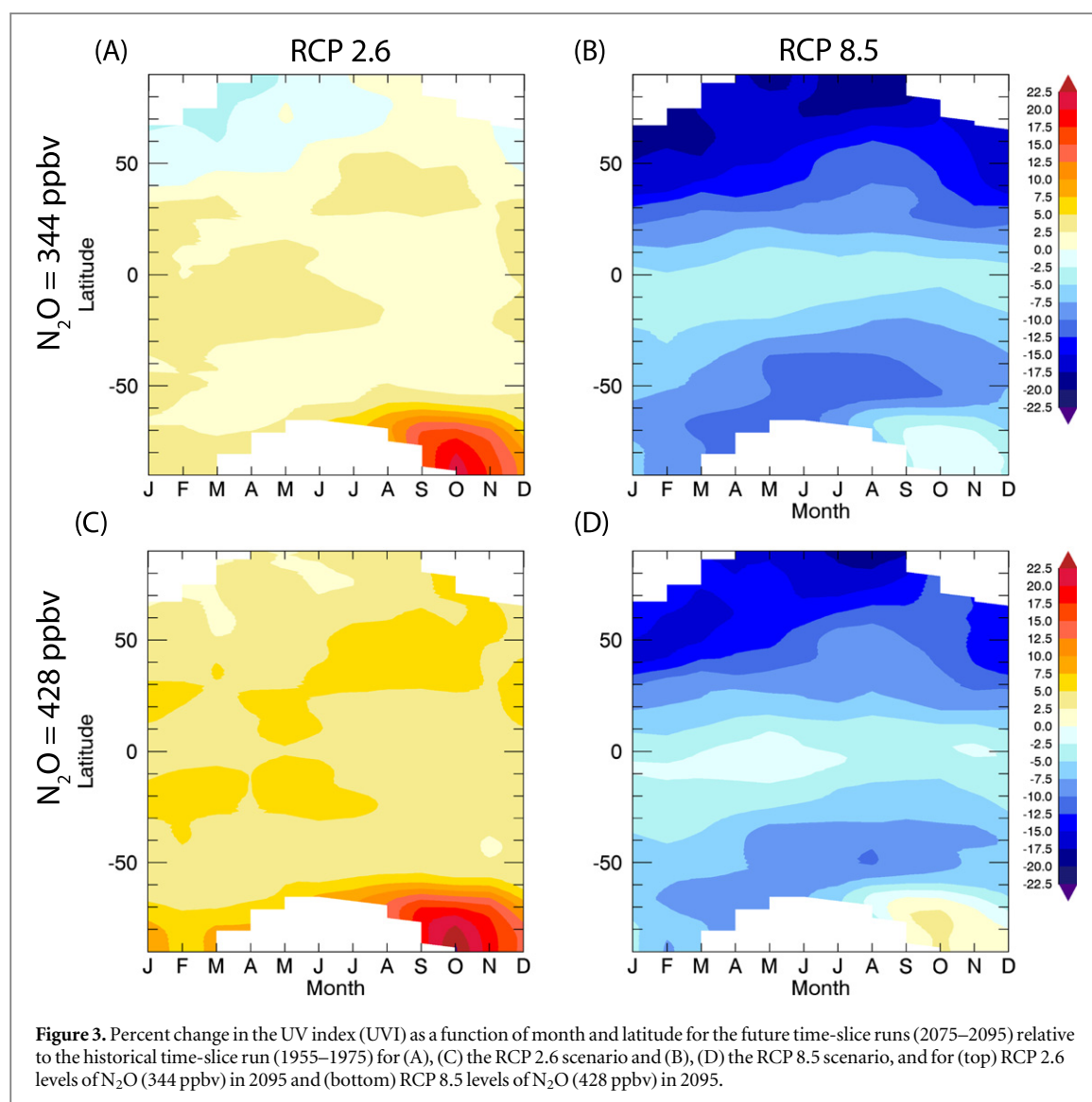
Figure 3 shows the percent change in the UVI (based on total, not stratospheric, column ozone) compared to the historical period as a function of month for (A, C) the RCP 2.6 future climate and (B, D) the RCP 8.5 future climate for  $\text{N}_2\text{O}$  concentrations fixed at 344 ppbv (the 2095 RCP 2.6 value) or 428 ppbv (the 2095 RCP 8.5 value). Figures 3(A) and (D) represent the ‘control’ RCP 2.6 and 8.5 climates, respectively, while figures 3(B) and (C) correspond to the lower and upper boundary for potential future changes in ozone in figure 2. Clearly, changes in UVI by the latter half of the century depend more on future  $\text{CO}_2$  concentrations than on  $\text{N}_2\text{O}$  concentrations. In the RCP 2.6 future climate, UVI remains only slightly larger than historical levels, except in the Southern Hemisphere (SH) spring where remaining halocarbons preferentially deplete polar ozone and increase surface UV exposure (highlighting the need to phase out these compounds). Conversely, in the RCP 8.5 future climate, UVI decreases everywhere relative to historical levels, and especially in the extratropics. These results are in general agreement with previous studies, though some studies find weak increases in UV exposure in the tropics (Bais *et al* 2011, Watanabe

*et al* 2011). It is clear that lower concentrations of  $\text{N}_2\text{O}$  would reduce UVI towards historical levels (i.e., zero percent change) in the RCP 2.6 climate (figures 3(A) versus (C)) but further away from historical levels in the RCP 8.5 climate (figures 3(B) versus (D)). In the RCP 8.5 future climate with  $\text{N}_2\text{O}$  mitigated to RCP 2.6 levels (figure 3(B)), the UVI is reduced up to  $\sim 20\%$  in the Northern Hemisphere (NH) extratropics. The UVI is actually closer to historical levels when  $\text{N}_2\text{O}$  is increased (figure 3(D)).

#### 4. Discussion and conclusions

The negative health effects of enhanced UV exposure, namely increased incidence of skin cancers and cataracts (Williamson *et al* 2014, Lucas *et al* 2015), could continue to impact the Southern Hemisphere as well as much of the globe in an RCP 2.6 future climate, particularly if  $\text{N}_2\text{O}$  concentrations increase. This enhanced UV exposure could be especially detrimental in the tropics, where UV exposure is already high. In contrast, consequences of reduced UV exposure are not as clear and need to be better understood, particularly if greenhouse gas emissions continue to increase (Heggin and Shepherd 2009, Bais *et al* 2015). For humans, one primary concern of lower UV is a reduction in vitamin D synthesis, which is associated with increased risks of osteoporosis, rickets, certain types of cancer, cardiovascular disease, multiple sclerosis, and rheumatoid arthritis (Autier *et al* 2014). Some recent studies are unable to find evidence of vitamin D supplementation reducing risk of these diseases (Lucas *et al* 2015). Extratropical UV reduction may be particularly important because it occurs in regions where vitamin D deficiencies are already prevalent (Correa *et al* 2013, Lucas *et al* 2015). Reductions in UV could also affect terrestrial and aquatic ecosystems, as well as biogeochemical and carbon cycles (Ballare *et al* 2011, Williamson *et al* 2014, Bornman *et al* 2015, Erickson III *et al* 2015, Hader *et al* 2015).

The UV changes presented here are based on a number of assumptions, such as cloud-free and unpolluted conditions. Potential future increases in cloud cover and reductions in surface reflectivity over the Arctic would cause additional decreases in UV at northern high-latitudes, while projected decreases in aerosols may counter these surface UV reductions; however, these effects are uncertain (Watanabe *et al* 2011, Correa *et al* 2013, Bais *et al* 2015). In addition, there are uncertainties in the tropospheric emission scenarios, and limitations of the model simulations themselves (such as limited tropospheric chemistry). Volcanic eruptions are not considered in our future simulations but can have substantial transient effects on stratospheric ozone and associated UV. Warmer temperatures and changing precipitation patterns may also cause behavioral changes, such as



altering the time people spend outdoors and thus, possibly, their overall UV exposure (Lucas *et al* 2015).

Uncertainties aside, it is reasonable to assert that a comprehensive scientific foundation for future policy decisions to protect the ozone layer will include (a) the collective impacts of expected  $CO_2$ ,  $CH_4$ , and  $N_2O$  emissions on ozone, (b) the intricacies of balancing continued tropical stratospheric ozone decreases with potential extratropical ozone increases, and (c) the amount of total ozone increases or decreases that are deemed a concern for human and ecosystem health. Because of the large global warming influence of  $CO_2$  and its enhancing effects on ozone, reducing  $CO_2$  emissions may be the ideal policy for both reducing climate change and returning ozone to near historical levels at all latitudes by the latter half of the century. Reducing  $CH_4$  emissions would have similar co-benefits. In contrast, reducing  $N_2O$  emissions increases stratospheric ozone, which is beneficial in the present-day climate and for certain future scenarios where ozone depletion persists; but decreasing  $N_2O$  may actually raise ozone above historical levels in the latter

half of the century if  $CO_2$  and  $CH_4$  emissions continue unabated as in the RCP 8.5 scenario.

Reducing greenhouse gas emissions to at least the RCP 4.5 trajectory is required to obtain the goals set forth by COP 21, which aim to limit global temperature changes to less than  $2^\circ C$  above pre-industrial levels (Collins *et al* 2013). If achieved, this would also return stratospheric ozone and UV to near historical levels globally by 2100. If we aim to limit global temperature changes to less than  $1.5^\circ C$   $CO_2$ ,  $CH_4$ , and  $N_2O$  all need to be reduced to near RCP 2.6 levels, which would also benefit the ozone layer. If the world cannot achieve a reduction in  $CO_2$  emissions to at least RCP 4.5 levels, the range of policy options to protect the ozone layer would necessarily broaden. One possibility includes allowing increased  $N_2O$  emissions in order to maintain historical ozone and UV levels in the extratropics. For this option, the benefits of reducing ozone levels in the extratropics would need to be carefully weighed against slightly larger depletion in the tropics and the greenhouse gas effects of  $N_2O$ . This again highlights the importance of meeting COP21

goals to minimize future climate change and maintain surface UV at historical levels for human health and ecosystems.

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