AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER

10.1002/2016GL067997

Key Points:

- Tropical total column ozone is projected to decrease due to rising greenhouse gas concentrations
- Consequently, a significant increase of harmful UV radiation is predicted for parts of the tropics
- Largest uncertainties exist about the future amount of tropical tropospheric ozone

Supporting Information:

- Supporting Information S1

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Citation:
Meul, S., M. Dameris, U. Langematz, J. Abalichin, A. Kerschbaumer, A. Kubin, and S. Oberländer-Hayn (2016), Impact of rising greenhouse gas concentrations on future tropical ozone and UV exposure, *Geophys. Res. Lett.*, *43*, doi:10.1002/2016GL067997.

Received 28 JAN 2016
Accepted 2 FEB 2016
Accepted article online 5 FEB 2016

Impact of rising greenhouse gas concentrations on future tropical ozone and UV exposure

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Abstract Future projections of tropical total column ozone (TCO) are challenging, as its evolution is affected not only by the expected decline of ozone depleting substances but also by the uncertain increase of greenhouse gas (GHG) emissions. To assess the range of tropical TCO projections, we analyze simulations with a chemistry-climate model forced by three different GHG scenarios (Representative Concentration Pathway (RCP) 4.5, RCP6.0, and RCP8.5). We find that tropical TCO will be lower by the end of the 21st century compared to the 1960s in all scenarios with the largest decrease in the medium RCP6.0 scenario. Uncertainties of the projected TCO changes arise from the magnitude of stratospheric column decrease and tropospheric ozone increase which both strongly vary between the scenarios. In the three scenario simulations the stratospheric column decrease is not compensated by the increase in tropospheric ozone. The concomitant increase in harmful ultraviolet irradiance reaches up to 15% in specific regions in the RCP6.0 scenario.

1. Introduction

Since the 1990s, when the regulation of man-made ozone depleting substances (ODSs), defined in the Montreal Protocol and its amendments, came into effect, much effort was put into monitoring the evolution of these compounds as well as their effect on the ozone layer. Recent ground- and space-based observations suggest that near-global total column ozone (TCO) has increased by around 1% between 2000 and 2013, albeit with distinct discrepancies between individual data sets [*World Meteorological Organization (WMO)*, 2014]. Based on estimates of a continuing ODS decline, comprehensive chemistry-climate models (CCMs) predict a recovery of TCO toward 1980 benchmark levels during the 21st century (21C) in the extratropics [*WMO*, 2014]. However, in addition to the chemical effects on ozone by ODSs, increasing concentrations of the well-mixed greenhouse gases (GHGs) carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) affect stratospheric ozone through chemical and dynamical processes: GHG-induced radiative cooling of the upper stratosphere will accelerate ozone recovery, as temperature-dependent ozone loss reactions will slow down [*Jonsson et al.*, 2004]. Furthermore, chemical reaction rates are altered by the increase of the CH_4 and N_2O loading. While the CH_4 increase will enhance TCO, the rising level of nitrogen oxides (NO_x), which is related to increased N_2O emissions, will reduce stratospheric ozone [*Fleming et al.*, 2011].

In addition to the effect on chemistry, increasing sea surface temperatures (SSTs) will enhance the upwelling of tropical air masses in the lower stratosphere [e.g., *Garny et al.*, 2011] by an acceleration of the stratospheric Brewer-Dobson circulation (BDC) [e.g., *Butchart et al.*, 2010], affecting ozone transport from the production region in the tropics to middle and high latitudes [e.g., *Li et al.*, 2009]. Consequently, in the extratropics, growing GHG concentrations will speed up the ODS-related recovery of stratospheric partial column ozone (PCO).

In the tropics, however, the future evolution of TCO is more difficult to assess. The enhanced export of lower stratospheric ozone counteracts the chemically induced ozone increase in the middle and upper stratosphere [e.g., *Plummer et al.*, 2010]. Furthermore, increasing tropospheric PCO levels can partly offset stratospheric changes [*Shepherd et al.*, 2014]. While non-climate-change experiments show that the expected ODS decline would lead to a return of tropical TCO to 1980 levels around the middle of 21C [*Eyring et al.*, 2010], no such return is predicted until 2100 including the GHG increase under the moderate A1B scenario

[*Intergovernmental Panel on Climate Change (IPCC)*, 2001; *Austin et al.*, 2010]. The dynamically driven ozone decrease in the tropical lower stratosphere exceeds the ODS- and GHG-induced ozone increase in the upper stratosphere as well as the ozone increase in the troposphere. Thus, following the A1B scenario, CCM projections indicate that in the tropics increasing GHG concentrations will offset the benefits of the ODS regulation, which will be accompanied by an increase in harmful ultraviolet (UV) irradiance at the surface [e.g., *Hegglin and Shepherd*, 2009; *Bais et al.*, 2011].

However, a range of future GHG emission scenarios is possible. Therefore, updated boundary conditions of the new Representative Concentration Pathways (RCPs) [*Meinshausen et al.*, 2011] are provided for model integrations with different future GHG scenarios, as the Climate Model Intercomparison Project (CMIP5). Analyses of selected CMIP5 models including interactive or semioffline chemistry revealed that by the end of 21C a range of tropical stratospheric PCO levels is possible, depending on the RCP scenario [*Eyring et al.*, 2013]. Nevertheless, all scenarios lead to a reduction compared to 1980. In contrast, a substantial spread is found for TCO (10 Dobson unit (DU)) with positive changes in the RCP8.5 and negative in other scenarios [*WMO*, 2014]. This spread is explained by the growing role of tropospheric ozone change [*Shepherd et al.*, 2014] which is strongly scenario dependent. However, the dependence of the results on the different ensemble sizes and models used for the scenario model means is controversially discussed [*WMO*, 2014]. Furthermore, a quantitative deviation of the model mean anomaly from the CCMVal-2 projections is reported with respect to the timing of minimum TCO which may also affect the projected TCO level at the end of 21C.

A more detailed analysis of model data regarding the future evolution of tropical ozone is required, not only to underpin the statements made in *WMO* [2014] but also to enable a critical evaluation and designation of uncertainties. Moreover, the potential changes in harmful solar radiation at the surface, which are of interest for the population in the tropics, are not estimated for the different RCP scenarios in the previous studies.

In our study we investigate the future evolution of tropical ozone under the RCP scenarios 4.5, 6.0, and 8.5 projected with one individual CCM. In addition to the total and stratospheric ozone column, we analyze the scenario-dependent evolution in the upper and lower stratosphere to separate the response of the different processes. Ozone changes in the model are calculated interactively as a reaction to the ODS and GHG evolution and are directly linked to changes in UV irradiance at the surface. This allows us to assess the range of projected future TCO, PCO, and UV changes in the tropics in a consistent way.

2. Model and Experiments

Simulations have been performed with the state-of-the-art CCM EMAC (ECHAM/MESSy Atmospheric Chemistry) [*Jöckel et al.*, 2006]. EMAC is a numerical chemistry and climate simulation system that includes submodels describing tropospheric and middle atmosphere processes. We applied EMAC (with the core atmospheric model ECHAM5 version 5.3.01 [*Roeckner et al.*, 2006] and the coupling system MESSy version 1.10) in the T42L39MA-resolution, i.e., with a spherical truncation of T42 (approximately 2.8°× 2.8°) with 39 hybrid pressure levels up to 0.01 hPa.

We performed three transient simulations, following the RCP scenarios 4.5, 6.0, and 8.5 [*Meinshausen et al.*, 2011], respectively. After the spin-up time of at least 5 years each experiment has been integrated from 1960 to 2100. The corresponding SST and SIC (sea ice concentration) input fields are obtained from the Max Planck Institute Earth System Model (MPI-ESM) [*Schmidt et al.*, 2013] for the RCP4.5 and RCP8.5 scenarios and (due to availability) from CMIP5 simulations with the Hadley Global Environment Model 2 - Earth System (HadGEM2-ES) model [*Jones et al.*, 2011] for the RCP6.0 scenario.

ODS boundary conditions are prescribed following the adjusted A1 scenario from *WMO* [2007] in all simulations. The quasi-biennal oscillation in the tropics is included by assimilating observed wind data into the model. For the future, observations are repeated. The observed 11 year solar cycle is prescribed between 1960 and 2008. As in the future the prescribed solar variability is not identical for the scenarios, we remove the solar variability from the analyzed time series by using the multiple linear regression analysis [*Bodeker et al.*, 1998].

The statistical significance of differences between the chosen time periods is tested with the Student's *t* test using the time series without solar variations as input.

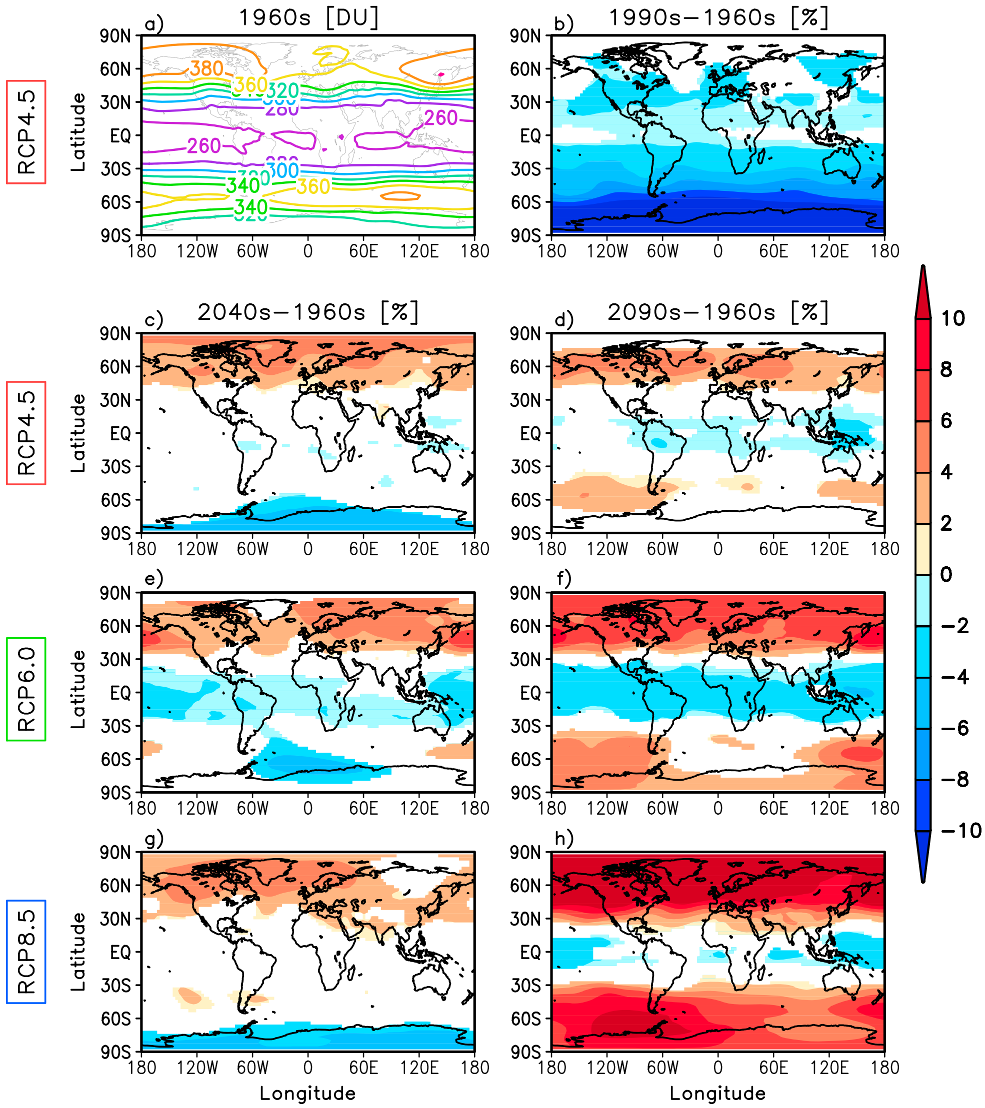

Figure 1. Geographical distribution of the annual mean TCO in DU in (a) the RCP4.5 simulation for the 1960s mean and the relative TCO difference between the (b) 1990s and 1960s, (c) 2040s and 1960s, and (d) 2090s and 1960s in % in the RCP4.5 simulation. (e and f) Same as Figures 1c and 1d but for the RCP6.0 simulation. (g and h) Same as Figures 1c and 1d but for the RCP8.5 simulation. Statistically significant changes on the 95% confidence niveau are colored.

3. Results

3.1. Effects of Different GHG Emission Scenarios on Ozone

In our reference period (1960s), the annual mean TCO distribution, shown for the RCP4.5 simulation (Figure 1a), is characterized by low values in the tropics and high values in the extratropics with the maximum in the Northern Hemisphere (NH). In the 1990s, TCO is globally reduced reaching 20% over Antarctica compared to the 1960s (Figure 1b). This is in qualitative agreement with values derived from observations [*WMO*, 2003] (not shown), although the ozone hole area in October is underestimated by the model as in other CCMs [*Austin et al.*, 2010] (see also Figure S4 in the supporting information). For the past period, we obtain nearly identical results for the three scenarios (not shown), reflecting the use of quasi identical boundary conditions.

The projected changes for the middle (2040s) and the end of 21C (2090s) to the 1960s are shown for the most moderate (Figures 1c and 1d), the medium (Figures 1e and 1f), and the strongest GHG scenario (Figures 1g and 1h). In the 2040s, TCO will have returned to and will partly exceed 1960s values in the NH extratropics in all scenario simulations but stay significantly lower at Southern Hemisphere high latitudes where ODSs decline slower (Figures 1c, 1e, and 1g). In the tropics, however, TCO reacts differently to the RCP scenarios.

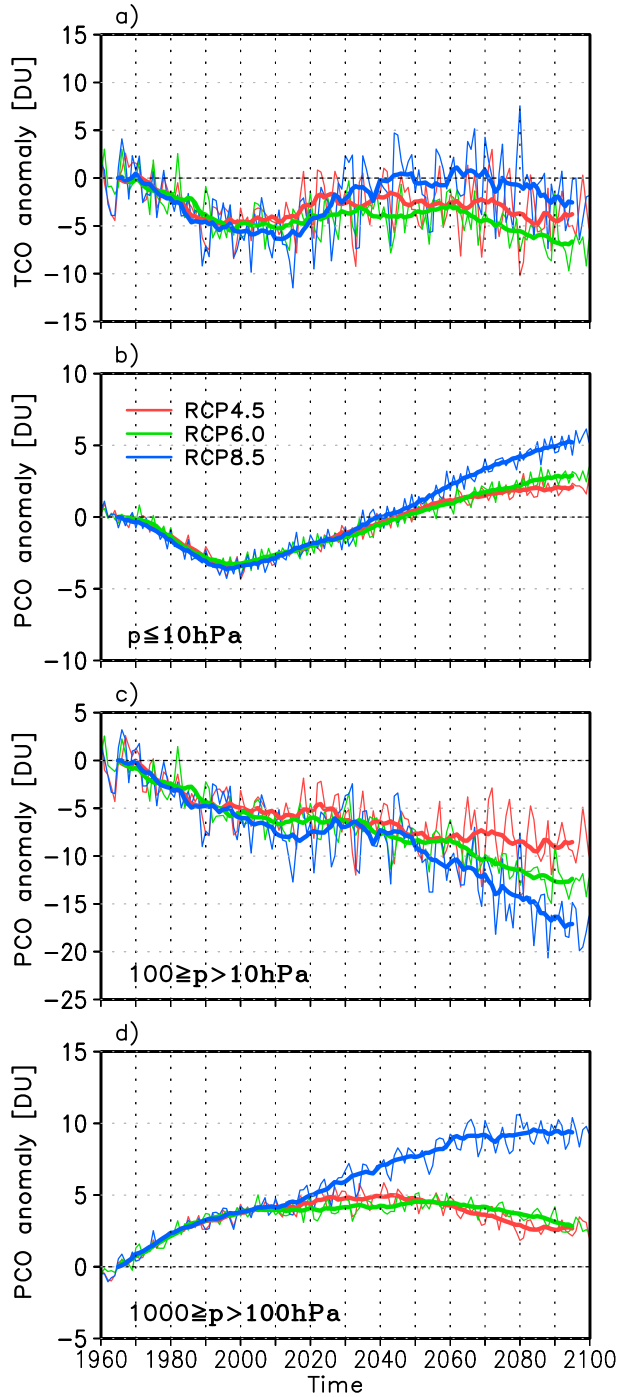

Figure 2. (a) Time series of annual mean tropical mean (20°S–20°N) TCO anomaly to the 1960–1970 mean for the RCP4.5 (red), RCP6.0 (green), and RCP8.5 (blue) simulations, (b–d) same as Figure 2a but for the PCO for the (Figure 2b) upper stratosphere (pressure ≤ 10 hPa), (Figure 2c) middle stratosphere (100 hPa ≥ pressure > 10 hPa), and (Figure 2d) troposphere (1000 hPa ≥ pressure > 100 hPa). Thick solid lines indicate the smoothed time series.

Following the RCP4.5 and RCP8.5 scenarios, TCO will have returned to 1960s values by the middle of 21C (i.e., no significant difference to the 1960s), while significantly lower TCO is projected for the RCP6.0 scenario. In the far future, TCO will increase in the extratropics in all scenarios (Figures 1d, 1f, and 1h). In contrast, tropical TCO will be significantly lower than 1960s values. Thus, by the end of the integration period all scenarios lead to a tropical TCO decrease, but the largest values as well as the largest area of significant tropical TCO decrease relative to the 1960s are found for the medium RCP6.0 scenario.

3.2. Scenario-Dependent Processes Affecting Ozone

To investigate the sensitivity of the processes, which are driving the tropical TCO changes, to the different scenarios, tropical anomaly time series are shown for TCO (Figure 2a) and the PCO in three altitude regions, i.e., the upper and lower stratosphere and the troposphere (Figures 2b–2d).

In the past the evolution of tropical TCO in the simulations is comparable to observations (relative to 1979–1988 mean; Figure S2). The future TCO anomaly clearly shows a different reaction to the RCP scenarios as indicated by Figure 1. In the upper stratosphere PCO will increase with largest changes in the strongest GHG scenario and smallest in the weakest scenario with a spread of about 3 DU (Figure 2b). This is related to the GHG-induced stratospheric cooling and the concomitant reduced ozone loss.

In the lower stratosphere, where the chemical lifetime of ozone is larger and ozone is affected by both chemistry and transport changes [e.g., *Austin et al.*, 2010; *Meul et al.*, 2014], PCO is projected to slightly increase between 2010 and 2030 but to decrease afterwards until 2100 (Figure 2c). Again, the PCO change in 2100 is largest in the RCP8.5 and smallest in the RCP4.5 simulation varying by about 9 DU. In this region, the rising branch of the BDC causes a net export of ozone which is enhanced in future climate, forced by higher tropical SSTs and a strengthening of the BDC [e.g., *Oberländer et al.*, 2013]. The magnitude of the simulated PCO decrease is linked to the increase of the vertical velocity of the mean residual circulation (not shown). Thus, the projected ozone decrease in the tropical lower stratosphere in the different scenarios is consistent with the trends of the prescribed SST time series (Figure S1c), which diverge between the scenario runs after the 2040s, with the largest increase in the RCP8.5 scenario.

Tropospheric PCO has increased in the past and is projected to continue increasing until 2100 for the RCP8.5 scenario (Figure 2d), while the RCP4.5 and RCP6.0 scenarios will lead to a decline of tropospheric PCO in the second half of 21C. The chemical production of tropospheric ozone is driven by the emissions of ozone precursor gases, including NO_x, carbon monoxide (CO), CH_4, and non methane volatile organic compounds [*IPCC*, 2001]. We find that the tropospheric PCO difference between the scenario runs is strongly connected to the concentrations of CH_4 (Figures 2d and S1b): While the emissions of the other precursors will decrease in all scenarios after 2050 (Figure S1a), a decline of surface CH_4 is predicted only for the weak and medium scenarios. In the RCP8.5, however, it more than doubles between 2000 and 2100. The resulting variation of tropospheric PCO between the scenarios is about 6 DU at the end of 21C.

Hence, in all GHG scenarios, the decline of TCO in the tropics by the end of 21C is a result of the predominant PCO decrease in the lower stratosphere which is reduced but not completely outweighed by the concurrent ozone recovery in the upper stratosphere and tropospheric ozone increases. Due to the particularly enhanced tropospheric ozone increase in the RCP8.5 scenario, the negative tropical TCO anomaly relative to the 1960s (but also to the 1980s; Figure S3) will be smaller in the extreme GHG scenario than in the weaker RCP6.0 scenario.

These results are partly in contradiction to *WMO* [2014] that shows positive TCO changes in the RCP8.5 scenario simulations from CMIP5 models also when comparing to the 1960s. However, the largest decrease of tropical TCO being projected for the medium RCP6.0 scenario is in agreement with *WMO* [2014]. The spread between the different TCO projections by about 4 DU is smaller than reported in *WMO* [2014].

How the counteracting effects on the ozone development in the different altitude regions vary between the scenarios is analyzed in more detail based on the vertical structure of PCO changes (Figure 3). The 1960s (Figure 3a) are characterized by a maximum increase of the PCO lying above each level between 10 and 50 hPa, reflecting the location of the stratospheric ozone layer. In all scenarios the change between the 2040s and 1960s (Figure 3b) is small and nearly identical in the upper stratosphere down to 10 hPa. Between 20 and 70 hPa, the dynamically induced reduction of the above lying PCO is comparable in the RCP6.0 and RCP8.5 runs but larger than in the moderate RCP4.5 scenario. This results in a decreased PCO at the tropopause in all scenarios, but with the largest changes in the RCP6.0 simulation (−8 DU). Thus, middle of 21C we already detect an effect of different GHG scenarios in the lower stratosphere but not in the upper stratosphere. In the troposphere, the same increase of the PCO is projected in the RCP4.5 and RCP6.0 simulation (≈5 DU), leading to a smaller decline of the ozone column at the surface than at the tropopause (Figure 3b). In the RCP8.5 simulation the positive trend of ozone in the troposphere offsets the negative trend in the stratosphere, and no significant change is projected at the surface (see also Figure 1g).

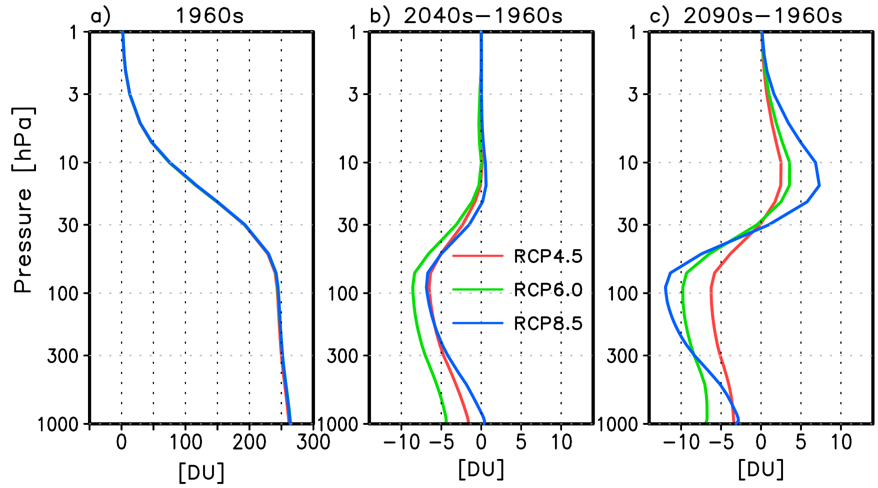

Figure 3. Vertical profile of PCO (20°S–20°N) lying above each level for (a) the 1960s mean and the difference between (b) the 2040s and 1960s, and (c) the 2090s and 1960s in DU. Color coding as in Figure 2. Note the nearly identical lines in Figure 3a.

At the end of 21C (Figure 3c), all scenarios show an increase of PCO in the upper stratosphere with the smallest increase in the RCP4.5 and the largest in the RCP8.5 simulation (2.5/6.8 DU, respectively, seen at 10 hPa). Between 15 and 70 hPa the dynamically driven ozone decrease exceeds the chemically induced ozone increase above, leading to a reduced stratospheric PCO at the tropopause in all scenarios compared to the 1960s. This is consistent with the results by *Eyring et al.* [2013]. However, the tropospheric ozone increase will not cancel out the stratospheric ozone decrease in the RCP8.5 simulation (as shown in *WMO* [2014]) but will lead to a comparable TCO reduction in the extreme GHG scenario as in the moderate RCP4.5 scenario. Despite the less extreme GHG increase in the RCP6.0 compared to the RCP8.5 scenario, the largest tropical TCO decrease is projected for this medium scenario. Regarding the change of column ozone at the surface, these results show that the evolution of tropospheric ozone is crucial for predicting future TCO levels.

3.3. Changes in Harmful UV-B Irradiance

Which consequences will these ozone changes have for life in the tropics? This is answered by analyzing the changes in surface UV-B irradiance ($\lambda = 280 - 315$ nm) that is able to damage human DNA (deoxyribonucleic acid) for the three RCP scenarios. The integral of UV-B radiation weighted with the DNA-damage action spectrum (UVB-DNA; based on *Setlow* [1974 and *National Research Council (NRC)* [1982]) is calculated online during the model integration and therefore consistently accounts for changes in ozone concentration and cloudiness.

Figures 4a and 4b show the 1960s mean and the past change of the surface UVB-DNA for the RCP4.5 simulation. The low climatological TCO prevailing in the tropics (Figure 1a) comes along with high values of UVB-DNA. In the past, a significant increase of UVB-DNA by 10–30% in the southern subtropics and by more than 100% at high latitudes occurred due to the development of the Antarctic spring ozone loss. Tropical latitudes were not significantly affected by ODS-induced ozone depletion (Figure 4b).

Consistent with the projected TCO changes in Figure 1, a significant increase of UVB-DNA is found in some tropical regions in the mid-21C depending on the scenario (Figures 4c, 4e, and 4g). At the end of 21C at both the northern and southern high latitudes less harmful UV-B irradiance will reach the surface compared to the 1960s, whereas in the tropics all scenarios show the tendency to an UVB-DNA increase (Figures 4d, 4f, and 4h). The largest significant increase of 15% is expected for the RCP6.0 scenario, with South America, South Africa, and Australia being affected most. In the tropical mean the UVB-DNA changes from the 1960s to the 2090s vary between +1 to +5% between the RCP scenario simulations (not shown). The geographical pattern found in the EMAC simulations is qualitatively similar to changes in erythemally weighted UV-B radiation derived from A1B-projections of CCMVal-2 models [*Bais et al.*, 2011].

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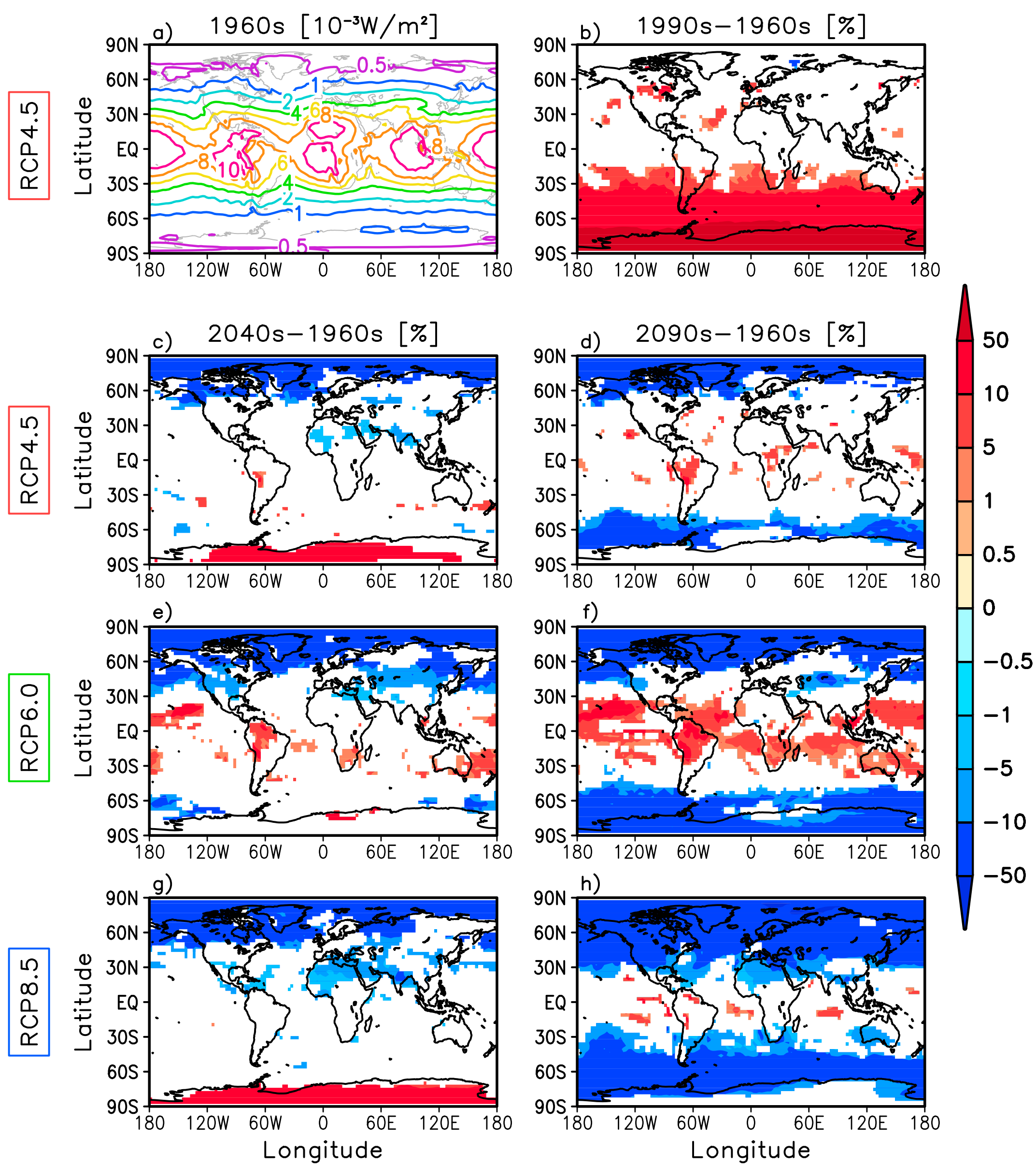

Figure 4. Same as Figure 1 but for UVB-DNA (integral of UV-B irradiance weighted with the DNA-damage action spectrum; see text for details) at the surface in (a) 10^{-3} W/m^2 and (b–h) %.

4. Discussion and Conclusions

Projections of tropical ozone in 21C are difficult due to dynamical and chemical processes in different altitude regions competing with each other. In this study, we have analyzed the sensitivity of future ozone changes to the three GHG scenarios RCP4.5, RCP6.0, and RCP8.5 in simulations with the CCM EMAC. This is done separately for the lower and upper stratosphere and the troposphere to better understand the changes of the total column.

The range of upper stratospheric TCO increase relative to the 1960s that is projected for the three scenarios is found to be relatively small (3 DU) compared to the spread that occurs in lower stratospheric PCO decrease (9 DU) at the end of 21C. This results in decreased stratospheric PCO in all scenarios ranging from −6 DU in the RCP4.5 to −12 DU in the RCP8.5 run. The negative change in stratospheric PCO in all scenarios is in qualitative agreement with *Eyring et al.* [2013]. However, we find the largest decrease in the strongest scenario (RCP8.5), whereas *Eyring et al.* [2013] show a slightly larger decrease in the RCP6.0 scenario. This might be due to different models contributing to the scenario means in *Eyring et al.* [2013] and/or due to an effect of different SST/SIC sources in the EMAC simulations. In the RCP6.0 simulation a slightly smaller sensitivity of stratospheric circulation changes to SST changes is found compared to the RCP4.5 and RCP8.5 runs (Figure S5). To roughly estimate this effect on the lower stratospheric PCO changes, we use the linear relationships of the RCP8.5 run between surface temperature, tropical upwelling, and lower stratospheric PCO. This results in a larger decrease of lower stratospheric PCO by ≈1.5 DU for the RCP6.0 run. Vice versa the lower stratospheric PCO decrease in the RCP4.5 and RCP8.5 scenarios is smaller by ≈1 DU and ≈2.5 DU, respectively, using the linear relationship derived from the RCP6.0 run. In both cases, the lower stratospheric PCO decrease remains largest in the RCP8.5 scenario, but for the stratospheric column the changes become similar for the RCP6.0 and RCP8.5 runs.

The evolution of tropospheric PCO in the EMAC simulations is shown to be highly scenario dependent since the different emission scenarios for CH_4 strongly affect the chemical production of ozone. Thus, at the end of 21C the tropospheric PCO increase in the RCP8.5 run is projected to be 6 DU larger than in the RCP4.5 and RCP6.0 simulations. This different behavior in the troposphere leads to the result that the largest TCO decrease at the end of 21C is projected for the medium GHG scenario, consistent with the results in *WMO* [2014]. However, regarding the RCP8.5 scenario, no positive sign is found for the TCO change in the EMAC simulations in contrast to the CMIP5 model selection [*WMO*, 2014], even when accounting for potential effects of the SST/SIC data set.

These results indicate that there is a possibility for a future decrease in tropical TCO leading to intensified harmful UV-B irradiance at the surface. In the EMAC scenario simulations the tropical mean change of UVB-DNA from the 1960s to the 2090s is between 1 and 5% with the largest increase projected for the RCP6.0 run. We show that in specific regions UVB-DNA significantly increases by 15% in the medium scenario. Enhanced GHG concentrations are identified as the major drivers for these changes. Nevertheless, it is a challenge to estimate the integral effect in the tropics. On the one hand, the expected increase of ozone in the middle and upper stratosphere due to lower temperatures is well understood and can precisely be quantified; on the other hand, the strength of the ozone decrease in the lower stratosphere caused by stronger future upwelling is less certain. Nonetheless, our model results show a clear reduction of stratospheric PCO in all three RCP scenarios. This finding agrees with the CMIP5 results [*Eyring et al.*, 2013] and suggests that a future reduction of the stratospheric ozone column is a robust feature. So far, determining the future change of the tropospheric ozone column is uncertain because its sign strongly depends on the assumed emission scenario and the differently complex tropospheric chemistry schemes used in the models. In the EMAC scenario simulations the sign of tropical tropospheric ozone change differs after year 2030, with a positive trend in RCP8.5 and negative trends in RCP6.0 and RCP4.5. Combining all effects, the EMAC simulations show a general decrease of the tropical TCO in the future. In contrast, the CMIP5 results show a slight future increase of TCO in RCP8.5 only. By comparing the RCP8.5 results from *Eyring et al.* [2013] and *WMO* [2014], we derive an increase of tropospheric PCO of 9 DU which is comparable to the EMAC simulations (Figure 2d). Thus, the differences in the RCP8.5 TCO projections arise from the stratospheric PCO changes. This points out that the sign of the TCO change depends on both the projected magnitude of lower stratospheric PCO decrease and the tropospheric ozone changes. To summarize, a future reduction of the tropical stratospheric ozone is most likely, but the TCO effect may be masked by a strong increase of the tropospheric ozone amount.

Acknowledgments
Data for this paper are available at the Freie Universität Berlin on the SHARP ftp data archive under GRL_trop_ozone_Meul_et_al_2015.tar. Please contact ulrike.langematz@met.fu-berlin.de. This work has been funded by the Deutsche Forschungsgemeinschaft (DFG) within the Research Unit SHARP (LA 1025/14-2, 1025/13-2, and 1025/15-1). J.A. was supported by the DFG within ISOLAA (LA 1025/19-1), and A. Ku. was supported by the BMBF within MiKlip (01LP1168A). We thank M. Kunze for providing the solar irradiance input for multiple linear regression. Furthermore, we would like to thank the North-German Supercomputing Alliance (HLRN) and the ECMWF computing center in Reading for computing time and support.

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