Tropospheric expansion under global warming reduces tropical lower stratospheric ozone

Aaron Match[1], **Edwin P. Gerber**[1]

[1]Center for Atmosphere Ocean Science, Courant Institute of Mathematical Sciences, New York University

Key Points:

- In response to global warming, tropical stratospheric ozone appears to shift upwards, with decreases at lower altitudes and increases aloft.
- The apparent shift arises from two distinct processes: increases aloft from stratospheric cooling and decreases below from surface warming.
- Ozone loss from surface warming is due in roughly equal parts to strengthening upwelling and expansion of tropospheric ozone destruction.

Corresponding author: Aaron Match, aaron.match@nyu.edu

Abstract

In response to global warming, stratospheric ozone appears to shift upwards, increasing aloft and decreasing below. The decreases have been attributed to strengthening upwelling and reverse self-healing, whereby increased ozone aloft blocks photons from forming ozone below. Separately, the decreases have been attributed to tropospheric expansion shifting the ozone layer upwards. Using chemistry-climate model experiments (CMIP6) and an idealized model undergoing Chapman photochemistry, tropospheric destruction of ozone, and transport between a tropical and extratropical column via a leaky tropical pipe, this paper disentangles how ozone responds to global warming. Tropospheric expansion is argued not to shift the ozone upwards but to shift the tropospheric destruction of ozone upwards, "taking a bite" out of the ozone layer then advected upwards. This mechanism contributes roughly half of the ozone reduction. This work cautions that tropospheric expansion can suggest incompatible predictions when applied to different variables, e.g. ozone concentration versus tropospheric destruction rate.

Plain Language Summary

The ozone layer absorbs ultraviolet light otherwise harmful to life. Due to compliance with the Montreal Protocol, the ozone layer is generally recovering from depletion. But global warming is predicted to also affect the ozone layer, leading to an apparent upward shift of ozone in the tropics, with increases in the upper stratosphere and decreases in the lower stratosphere. These decreases in the lower stratosphere have previously been attributed to strengthening of stratospheric upwelling or to an upward shift caused by the expansion of the troposphere under global warming. We draw on elements of these prevailing explanations to quantify a new mechanism, which explains roughly half of the decrease: expansion of the troposphere converts stratospheric air into tropospheric air, the low ozone anomalies from which are then transported upwards into the tropical lower stratosphere by the background upwelling.

1 Introduction

Chemistry-climate models robustly predict that, in response to global warming, ozone will increase in the upper stratosphere and decrease in the tropical lower stratosphere (Shepherd, 2008; Chiodo et al., 2018). Figures 1A-C show three such simulated responses to a quadrupling of CO_2 in chemistry-climate models with interactive ozone chemistry

contributed to Coupled Model Intercomparison Project Phase 6 (CMIP6). The robustness of the simulated decrease in tropical lower stratospheric ozone suggests models may capture consistent mechanisms for the change. Global warming might already be reducing ozone in the tropical lower stratosphere, where the recovery since 2000 due to the Montreal Protocol has been notably absent (Petropavlovskikh et al., 2019).

There are two prevailing families of explanations for the decrease in tropical lower stratospheric ozone under global warming. A first family of explanations emphasizes stratospheric processes: the decrease in tropical lower stratospheric ozone is explained to result from (1) the strengthening Brewer-Dobson circulation, which reduces ozone in the tropics by advecting ozone-poor air from below, and (less significantly) (2) "reverse self-healing" due to stratospheric cooling, which increases ozone in the upper stratosphere thereby attenuating the ultraviolet radiation available to form ozone in the tropical lower stratosphere (Groves et al., 1978; Rosenfield & Schoeberl, 2005; Shepherd, 2008; Plummer et al., 2010; Chiodo et al., 2018).

A second family of explanations, in a largely separate body of literature, emphasizes tropospheric expansion under global warming. As the troposphere deepens with surface warming at roughly 500 m K^{-1} (Vallis et al., 2015; Match & Fueglistaler, 2021), variables thermodynamically connected to the troposphere or dynamically connected to the tropopause are expected to shift upwards (Singh & O'Gorman, 2012). Ozone has been proposed as one such variable, and ozone trends have been transformed into tropopause-following coordinates on the premise that this would remove the trends from tropospheric expansion (Forster & Tourpali, 2001; Thompson et al., 2021). Figure 1D shows the tropically-averaged ozone response to quadrupling of CO_2 in the CMIP6 experiments (solid curves), overlaid with the change predicted from a 2 km upward shift in the ozone profile (dotted). The prediction from the vertical shift closely matches the actual response.

That two families of explanations for the ozone response to global warming coexist begs for an attempt at unification. Are predictions based on stratospheric processes constrained to agree with those based on tropospheric expansion? For example, the strengthening of the Brewer-Dobson circulation under global warming can be viewed as an upward shift under tropospheric expansion (Oberländer-Hayn et al., 2016). Yet, it is not clear why photochemical processes would shift upwards throughout the stratosphere un-

der tropospheric expansion, raising the possibility that the agreement between the ozone response and that predicted by the vertical shift is coincidental.

Attempts to unify these explanations must be able to distinguish the effects of stratospheric versus tropospheric processes. Past studies have imposed stratospheric cooling without surface warming, isolating the reverse self-healing (Sigmond et al., 2004; Fomichev et al., 2007). They have also analyzed surface warming without stratospheric cooling, yielding combined changes in the Brewer-Dobson circulation and tropopause height. To separate these two effects requires a model that can distinguish the impact of the rising tropopause on ozone chemistry from its impact on the Brewer-Dobson circulation. Coupling Chapman photochemistry to the leaky tropical pipe of Neu and Plumb (1999) allows us to disentangle the proposed mechanisms to explain the gross features of the ozone response to global warming.

Figure 1E previews our main results, indicating that the idealized model reproduces the coarse shape (decreases below, increases aloft) and magnitudes (anomalies of order 10^{12} molec cm^{-3} for a quadrupling of CO_2) of the comprehensive model response. Decomposing the response by term, it is clear that the increases aloft are due to stratospheric cooling (blue curve), consistent with previous work (Isaksen et al., 1980; Jonsson et al., 2004; Sigmond et al., 2004; Fomichev et al., 2007). Roughly half of the reduction in tropical lower stratospheric ozone is due to the strengthening Brewer-Dobson circulation (green curve). Roughly the other half of the reduction in tropical lower stratospheric ozone is due to expansion of tropospheric ozone destruction (red curve), which "takes a bite" out of the stratospheric ozone profile, the low ozone anomalies from which are then advected upwards by the background upwelling. This hybrid troposphere/stratosphere process draws on elements from the two prevailing families of explanations, revealing how they can be reformulated to provide a more complete picture of the ozone response. A similar mechanism has been identified in the annual cycle of ozone as it relates to the annual cycle of convective detrainment rate at different altitudes (Folkins et al., 2006).

2 The idealized model

Our model couples together a tropical and extratropical column, each undergoing Chapman photochemistry, transport via a leaky tropical pipe, and tropospheric ozone destruction. Our goal is to compute the annual, spatial mean ozone concentration in the

tropics $[O_3]_T$ and the extratropics $[O_3]_E$. The prognostic equations for ozone number density [units: molec cm^{-3}] are as follows:

$$\frac{\partial [O_3]_i}{\partial t} = \frac{\partial [O_3]_i}{\partial t}|_{\text{photochemistry}} + \frac{\partial [O_3]_i}{\partial t}|_{\text{transport}} + \frac{\partial [O_3]_i}{\partial t}|_{\text{tropospheric destruction}} \quad (1)$$

where subscript i corresponds to tropics (T) or extratropics (E):

2.1 Photochemistry

Ozone photochemistry is represented by the paradigmatic Chapman cycle (Chapman, 1930), following the treatment of Jacob (1999). Chapman photochemistry considers the evolution of three oxygen species: O, O_2, and O_3. The key reactions are:

$$O_2 + h\nu \rightarrow O + O \qquad (\lambda < 240nm) \qquad \text{(R1)}$$

$$O + O_2 + M \rightarrow O_3 + M \qquad \text{(R2)}$$

$$O_3 + h\nu \rightarrow O_2 + O \qquad (\lambda < 320nm) \qquad \text{(R3)}$$

$$O_3 + O \rightarrow 2O_2 \qquad \text{(R4)}$$

where M represents third bodies with the number density of air molecules. The rate constants k_1 and k_3 are photolysis rate constants, so must be calculated accounting for radiative transfer. Reaction 2 has temperature-dependent rate constant $k_2 = k_0^{300}(T/300)^{-n}$, where $k_0^{300} = 6.0 * 10^{-34}$ cm^6 $molec^{-2}$ s^{-1} and n = 2.4. Reaction 4 has temperature-dependent rate constant $k_4 = A \exp(-E_a/RT)$ with A = 8.0 * 10^{-12} cm^3 $molec^{-1}$ s^{-1} and E_a/R = 2060 K (Brassuer & Solomon, 2005).

Acknowledging that Reactions 2 and 3 proceed quickly, a slowly-evolving family of odd oxygen can be defined (O_x = O + O_3) that is dominated by O_3 and evolves as follows:

$$\frac{\partial [O_3]}{\partial t}|_{\text{photochemistry}} = 2k_1 C_{O_2} n_a - \frac{2k_3 k_4}{k_2 C_{O_2} n_a^2}[O_3]^2 \quad (2)$$

where C_{O_2} is the molar fraction of O_2 in air (today, 0.21), and $n_a(z)$ is the number density of air molecules as a function of height [molec cm^{-3}] (assumed exponentially-decaying with scale height H = 7 km).

The photolysis rate constants k_1 and k_3 depend on the radiation as it interacts with O_2 and O_3. The photolysis rates are calculated as an integral across all wavelengths of the spectrally-resolved photolysis rate:

$$k = \int_\lambda q(\lambda)\sigma(\lambda)I_\lambda d\lambda \tag{3}$$

with wavelength λ, quantum yield q (molecules produced per photon absorbed), absorption coefficient (cm^2 molec^{-1}), and actinic flux density with respect to wavelength I_λ (photons cm^{-2} s^{-1} nm^{-1}). The actinic flux is a radiative quantity that depends on the absorption (and scattering) aloft:

$$I_\lambda(z) = I_{\lambda,\infty} \exp(-\frac{\tau_\lambda(z)}{\cos\theta}) \tag{4}$$

where τ_λ is the wavelength-dependent optical depth resulting from absorption by chemical species aloft and θ is the solar zenith angle. We consider overhead sun in the tropics ($\theta = 0°$) and low sun in the extratropics ($\theta = 60°$). Only absorption by O_2 and O_3 is considered:

$$\tau_\lambda(z) = \int_z^\infty (\sigma_{O_2}(\lambda)[O_2] + \sigma_{O_3}(\lambda)[O_3])dz' \tag{5}$$

Analytical solutions of the equilibrium ozone profile do not exist for Chapman photochemistry because the radiation that forms ozone is a function of the ozone profile itself. The ozone equilibrium is found numerically, accounting also for transport and tropospheric destruction.

2.2 Tropospheric Destruction

The troposphere is represented as a region of fast destruction of ozone through chemical sinks or dry deposition (e.g. Wild, 2007).

$$\frac{\partial[O_3]_i}{\partial t}|_{\text{tropospheric destruction}} = -\frac{[O_3]_i}{\tau_i(z)} \tag{6}$$

where the ozone number density relaxes exponentially towards zero with damping rate $\tau(z)$. Our results herein consider $\tau(z)$ approaching infinity in the troposphere and zero in the stratosphere, so that tropospheric destruction instantaneously destroys tropospheric

ozone. For the control climatology, the tropical tropopause is set at 16 km and the extratropical tropopause at 10 km.

2.3 Transport

Ozone is transported between the tropics and extratropics by advection and mixing, a leaky tropical pipe (Plumb, 1996; Neu & Plumb, 1999) following the treatment of Stolarski et al. (2014). The mass flux divergence of the upwelling in the tropics transports ozone to the extratropics. The tropics and extratropics are laterally mixed on a timescale μ.

$$\frac{\partial [O_3]_T}{\partial t}|_{\text{transport}} = -w n_a \frac{\partial}{\partial z}(\frac{[O_3]_T}{n_a}) - \mu([O_3]_T - [O_3]_E) \tag{7}$$

$$\frac{\partial [O_3]_E}{\partial t}|_{\text{transport}} = w n_a \frac{\partial}{\partial z}(\frac{[O_3]_E}{n_a}) + (D + \mu)([O_3]_T - [O_3]_E) \tag{8}$$

where w is the rate of upwelling/downwelling (units: m s^{-1}), and μ is the lateral mixing rate between the tropics and extratropics (units: s^{-1}).

$$D = -e^{z/H} \frac{\partial}{\partial z}(w e^{-z/H}) \tag{9}$$

where D is the mass flux divergence of the tropical upwelling (units: s^{-1}).

2.4 Model set-up and parameters

The idealized shortwave radiative transfer and photolysis rates are solved on a wavelength grid with 151 discretized wavelengths ranging from 180 nm to 360 nm. The default temperature is taken to be uniformly 240 K. Molecular oxygen absorption coefficients (σ_{O_2}) are taken from Ackerman (1971) and ozone absorption coefficients (σ_{O_3}) from Demore et al. (1997). Solar actinic flux is calculated from the Solar Spectral Irradiance Climate Data Record (Coddington et al., 2015), averaged from 01-01-2020 to 02-04-2021. Spectrally-resolved parameters are linearly interpolated to the wavelength grid.

The vertical dimension is discretized into 81 vertical levels separated by 500 meters from 0 to 40 km. The timestep is 10,000 seconds, and the model is run to approximate equilibrium for 50,000 time steps (roughly 15 years).

Chapman photochemistry is known to simulate roughly double the observed ozone concentration, due to neglected catalytic destruction from NOx and HOx (Jacob, 1999). Incorporating catalytic chemistry can increase the number of chemical reactions by an order of magnitude (Crutzen, 1971), yet these reactions are not required to explain leading-order aspects of the ozone response to global warming. By neglecting catalytic chemistry, our idealized model trades off the quantitative accuracy of more complex models in order to distill the mechanisms essential to the ozone response to global warming.

3 CMIP6 models

Our idealized model of Chapman photochemistry will be compared to various experiments with global chemistry-climate models that have interactive stratospheric ozone chemistry contributed to the Coupled Model Intercomparison Project Phase 6 (CMIP6) (Eyring et al., 2016). The full ozone response to quadrupled CO_2 concentrations, including internally-consistent representations of stratospheric cooling, strengthening Brewer-Dobson circulation, and expansion of tropospheric destruction, is examined by comparing the final 100 years of the abrupt-4xCO2 experiments to the piControl experiment. Similar results were presented in Chiodo et al. (2018). For that comparison, we examine CESM2-WACCM-FV2, CNRM-ESM2-1, and MRI-ESM2-0, as shown in Figure 1.

This paper provides the first intermodel comparison isolating the effects of stratospheric cooling and surface warming on stratospheric ozone. The effects of stratospheric cooling are isolated by evaluating the ozone response to quadrupled CO_2 at prescribed pre-industrial sea surface temperature (piClim-4xCO2 minus piControl). The piClim-4xCO2 experiments were conducted as part of the Radiative Forcing Model Intercomparison Project (RFMIP) (Pincus et al., 2016), and we analyze CNRM-ESM2-1, GFDL-ESM4, MRI-ESM2-0, and UKESM1-0-LL. The effects of surface warming are isolated by evaluating the ozone response to an increase in prescribed sea surface temperatures of 4 K within historical simulations (amip-p4K minus amip). The amip-p4K experiments were conducted as part of the Cloud Feedback Model Intercomparison Project (Webb et al., 2017), and we analyze CNRM-CM6-1, E3SM-1-0, and MRI-ESM2-0.

4 Results

Our goal is to understand what drives the reductions in tropical lower stratospheric ozone under global warming. We decompose the ozone response to CO_2 forcing mech-

anistically into separate responses to stratospheric cooling, strengthening of the Brewer-Dobson circulation, and expansion of tropospheric ozone destruction. Our decomposition is performed using the idealized model, but our confidence is boosted by comparing to relevant CMIP6 experiments. A key result, shown in Figure 1E is that the response to all components is quite linear, even at the level of quadrupling CO_2, justifying our reductive approach.

4.1 Stratospheric cooling

Stratospheric cooling can increase ozone by quickening Reaction 2, which partitions odd oxygen even more in favor of ozone, thereby slowing ozone loss from Reaction 4 (Groves et al., 1978; Jonsson et al., 2004). Reaction 4 is also directly slowed at decreasing temperature. The effect of stratospheric cooling on ozone in CMIP6 is isolated by comparing piControl experiments with the piClim-4xCO2 experiments, in which CO_2 is quadrupled at prescribed pre-industrial sea surface temperatures (Fig. 2A-E). Stratospheric cooling increases tropical ozone above 25 km, robustly leading to increases of roughly $5*10^{11}$ molec cm^{-3} at 30 km. Although stratospheric cooling extends down to 20 km in the tropics, ozone is actually reduced in the lower stratosphere, due to reverse self-healing, whereby the anomalously increased ozone aloft absorbs ultraviolet photons that would have otherwise formed odd oxygen in the lower stratosphere. The simulated reductions due to reverse self-healing are not as robust as the increases due to stratospheric cooling aloft.

In response to a uniform cooling of 10 K, the idealized model reproduces the ozone increases in the upper stratosphere and reverse self-healing in the lower stratosphere (Figure 2F). The idealized response to stratospheric cooling agrees with comprehensive models on the order of magnitude (10^{12} molec cm^{-3}), although the exact magnitudes are overestimated. The altitude at which increases transition to decreases occurs around 30 km in the idealized model but around 25 km in the comprehensive models. Interestingly, reverse self-healing seems to vanish in the lowermost stratosphere, although this feature should not be taken literally given that realistic stratospheric cooling does not extend much below 20 km.

The ozone reductions in the tropical lower stratosphere are about an order of magnitude smaller in the stratospheric cooling experiments than in the total response to quadru-

pled CO_2 (Figure 2E vs. 1D). Thus, stratospheric cooling is only a minor contributor to tropical lower stratospheric ozone reductions.

4.2 Disentangling the strengthening of the Brewer-Dobson circulation from expansion of tropospheric destruction

Stratospheric cooling cannot explain the large reductions in tropical lower stratospheric ozone, thereby implicating surface warming. The two competing families of explanations for how surface warming reduces ozone in the tropical lower stratosphere are that it strengthens the Brewer-Dobson circulation or that it shifts the ozone upwards due to tropospheric expansion. We draw on both families by showing that the strengthening Brewer-Dobson circulation explains about half the response, with the other half explained by an upward shift in the tropospheric destruction of ozone, which predicts a different response than an upward shift of ozone itself.

In CMIP6, the effects of surface warming on ozone are analyzed by comparing amip-p4K experiments with amip experiments. The amip experiments are driven by realistic boundary conditions over the historical period, to which the amip-p4K experiments prescribe 4 K increases in sea surface temperatures. Figure 3A-D shows that in response to the surface warming of 4 K, ozone is reduced in the tropical lower stratosphere, with peak reductions around 20 km extending upwards with diminishing magnitude above 25 km.

The gross features of this response can be explained using the idealized model of the ozone response to global warming. Figure 3E shows the response of the idealized model to a uniform strengthening of the Brewer-Dobson circulation of 0.1 mm s^{-1} (green curve), which corresponds to a 2 km upward shift of the vertical mass flux profile (Oberländer-Hayn et al., 2016). Upwelling induces large ozone anomalies in the lower stratosphere, where transport is fast compared to photochemical recovery timescales (years around 23 km and months around 28 km) (Dutsch, 1968; Jacob, 1999). Upwelling barely perturbs ozone in the upper stratosphere, where transport is slow compared to photochemical recovery timescales (weeks around 33 km and days around 38 km).

Expansion of tropospheric ozone destruction plays a key role, too, as shown by the red curve in Figure 3E. Mechanistically, expansion of tropospheric ozone destruction destroys ozone by transforming stratospheric air into tropospheric air with a shorter ozone

lifetime, i.e. "taking a bite" out of the stratosphere. The anomalies from the bite are then advected upwards by the background upwelling into regions where photochemical equilibrium is eventually re-established. Figure 4 illustrates this two-step (bite then advect) mechanism. The idealized model suggests that the effects of strengthening of the Brewer-Dobson circulation and expansion of tropospheric ozone destruction are remarkably linear, even with the perturbation corresponding to a 2 km upward shift of the tropopause.

There are some notable quantitative differences between the idealized model and the chemistry-climate model response. In the idealized model, the reductions in ozone are overestimated by roughly a factor of two, and they extend upwards to 35 km, versus roughly to 30 km in the CMIP6 models. Despite these caveats, the idealized model demonstrates that the newly identified mechanism of expansion of tropospheric ozone destruction contributes at leading order to tropical lower stratospheric ozone reductions under global warming. In the simulations analyzed herein, strengthening Brewer-Dobson circulation and the expansion of tropospheric ozone destruction contribute equally to total column ozone loss (i.e. in Figure 3E, the integrals of the red and green curves are almost identical).

4.3 Revisiting the vertical shift of the ozone profile

Previous studies have suggested that tropospheric expansion under global warming shifts the ozone profile upwards (Forster & Tourpali, 2001; Thompson et al., 2021). Indeed, the ozone response to quadrupling of CO_2 strongly resembles a vertical shift (Figure 1D). But, when the role of tropospheric expansion is isolated in Figure 3D, it becomes clear that any vertical shift from tropospheric expansion does not extend above 25 km. It appears to be coincidental that stratospheric cooling increases ozone by an amount matching the prediction from a vertical shift (in Figure 1D).

The ozone response well below 25 km seems theoretically relatable to a vertical shift. By construction in the idealized model, expansion of tropospheric destruction of ozone induces a vertical shift in the ozone *below* the tropopause. If upwelling is sufficiently strong above the tropopause, then there forms a region of ozone depressed below its photochemical equilibrium above the tropopause, the structure of which could move upwards along with the tropopause. This region is not very deep, as shown in Figure 3E by the disagree-

ment within a few kilometers above the tropopause of the ozone anomalies predicted from tropospheric expansion (red) versus those predicted by a vertical shift (dotted).

5 Rejecting a naive hypothesis of tropospheric expansion

Tropospheric expansion might be hoped to support the naive hypothesis that all variables shift upwards under global warming. Indeed, variables in disparate regions and dynamical regimes appear to shift upwards under global warming, including cloud and microphysical quantities (Tompkins & Craig, 1999), static stability (Kushner et al., 2001), transient kinetic energy and momentum flux (Lorenz & DeWeaver, 2007), relative humidity (Sherwood et al., 2010), the Brewer-Dobson circulation (Oberländer-Hayn et al., 2016), and QBO amplitude (Match & Fueglistaler, 2021).

Yet, past work has noted some caveats that would limit a naive tropospheric expansion hypothesis. For example, Sherwood et al. (2010) reported that the tropospheric expansion hypothesis underestimates relative humidity changes in certain regions that are controlled by distant last points of saturation. Singh and O'Gorman (2012) argued that, in theory, temperature and pressure velocity should not only be shifted, but also rescaled. Our work emphasizes that the naive hypothesis of tropospheric expansion can lead to incompatible predictions when applied to two variables that cannot both shift upwards in a dynamically consistent way. In the idealized ozone model, an upward shift of the tropospheric destruction rate of ozone is incompatible with an upward shift of the ozone profile itself.

Therefore, the naive hypothesis must be rejected in favor of a more cautious approach: tropospheric expansion should be justified by the internal dynamics of the variable of interest as it responds to global warming. For ozone dynamics, tropospheric expansion should be applied to the tropospheric destruction rate of ozone, not to the ozone profile itself.

6 Conclusions

In response to CO_2 forcing, tropical stratospheric ozone appears to shift upwards, with decreases in the lower stratosphere and increases aloft. There are two co-existing families of explanations for the ozone reduction in the tropical lower stratosphere. The first attributes the decrease to a strengthening Brewer-Dobson circulation and (to a lesser

degree) reverse self-healing. The second attributes the vertical shift itself to tropospheric expansion.

Drawing on some elements from these two families but revising others, we argue that the tropical lower stratospheric decrease in ozone arises in roughly equal parts from the strengthening Brewer-Dobson circulation and expansion of the tropospheric destruction of ozone. Tropospheric expansion does not directly shift the ozone profile upwards, but rather shifts the tropospheric destruction of ozone upwards. This expanded tropospheric destruction "takes a bite" out of the stratospheric ozone profile, the low ozone anomalies from which are advected upwards by the background upwelling until the anomalies are damped by photochemistry. The increases in ozone aloft result from stratospheric cooling, so their resemblance to a vertical shift is coincidental. Although stratospheric cooling and tropospheric expansion both depend on CO_2 concentration, tropospheric expansion is mediated by climate sensitivity through the surface warming, whereas stratospheric cooling comes from the local radiative effects of CO_2.

7 Data Availability Statement

O2 absorption coefficients taken from Ackerman (1971), as accessed from `http://satellite.mpic.de/spectral_atlas/cross_sections/Oxygen/O2_Ackerman(1971)_298K_116.3-243.9nm(int-c).txt`. O3 absorption coefficients taken from Demore et al. (1997), as accessed from `http://satellite.mpic.de/spectral_atlas/cross_sections/Ozone/O3_JPL-2002(2002)_273K_175-363nm(rec).txt`. Solar actinic flux developed by Coddington et al. (2015), as accessed from `https://www.ncei.noaa.gov/products/climate-data-records/solar-spectral-irradiance`. CMIP6 data is accessible from `https://esgf-node.llnl.gov/search/cmip6/`.

Acknowledgments

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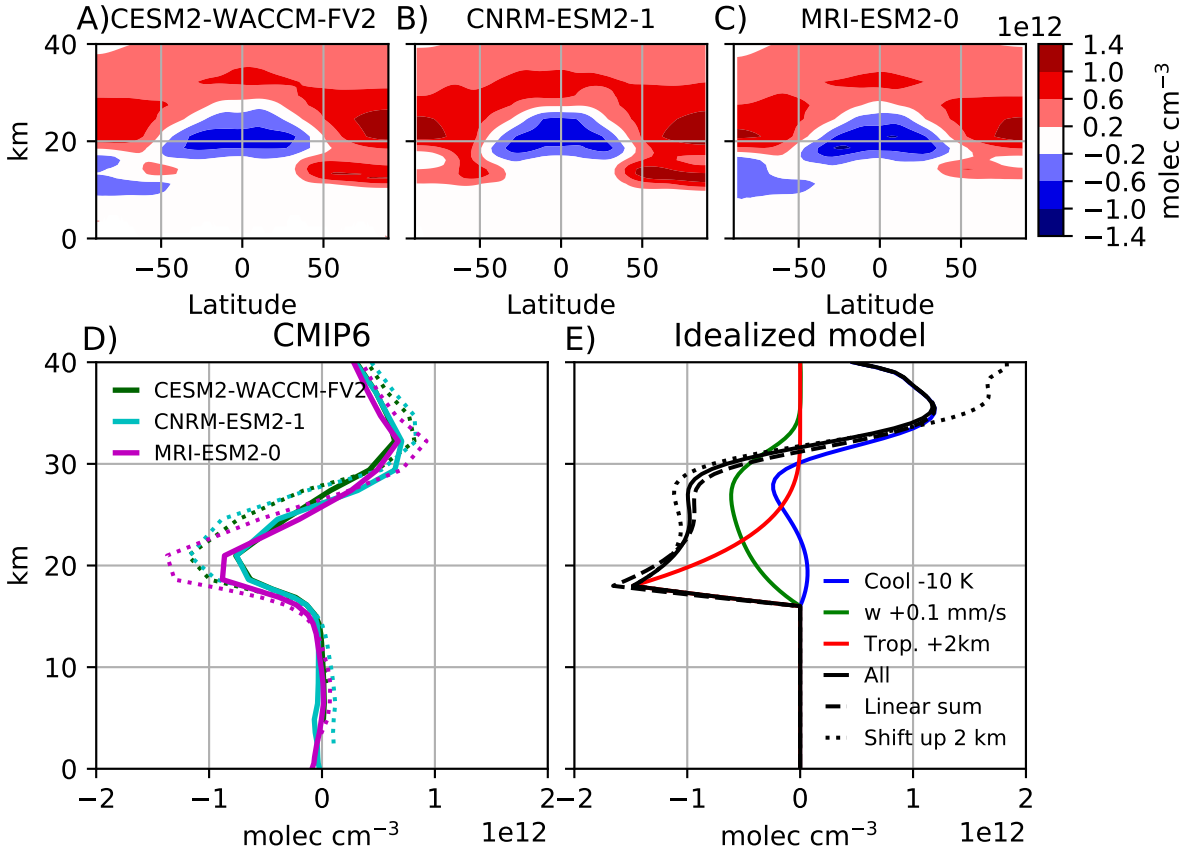

Figure 1. Effect on ozone number densities of CO_2 quadrupling. (A-C) Ozone number densities in CMIP6 models: abrupt-4xCO2 (final 100 years) minus piControl. (D) Tropical average (30S-30N) of panels A-C, and change in ozone number densities from shifting the piControl ozone profile up by 2 km (dotted). (E) Tropical ozone anomalies in an idealized two-column model of ozone photochemistry, destruction and transport (described in the Methods), where anomalies result from a uniform cooling of 10 K (blue), a uniform increase in tropical upwelling by 0.1 mm s^{-1} (green), an upward shift of the troposphere by 2 km (red), all of the above (black solid), the linear sum of each individual perturbation (black dashed), and the upward shift of the control ozone profile by 2 km (black dotted).

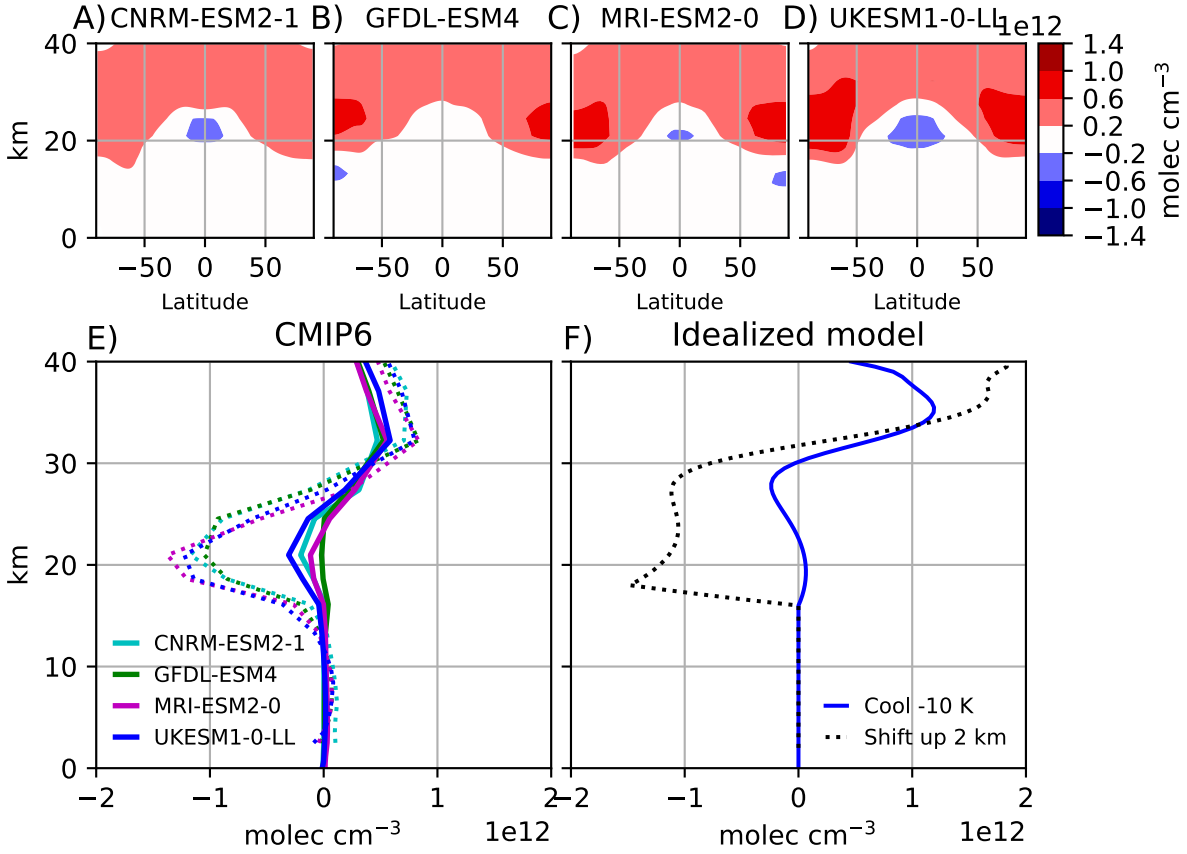

Figure 2. Effect on ozone number densities of stratospheric cooling. (A-D) Ozone number densities in piClim-4xCO2 minus piControl.(E) Tropical average (30S-30N) of panels A-D, and change in ozone number densities from shifting the piControl ozone profile up by 2 km (dotted). (F) Ozone anomalies in the idealized model in response to a uniform cooling of 10 K (blue) compared to upward shift of the control ozone profile by 2 km (black dotted).

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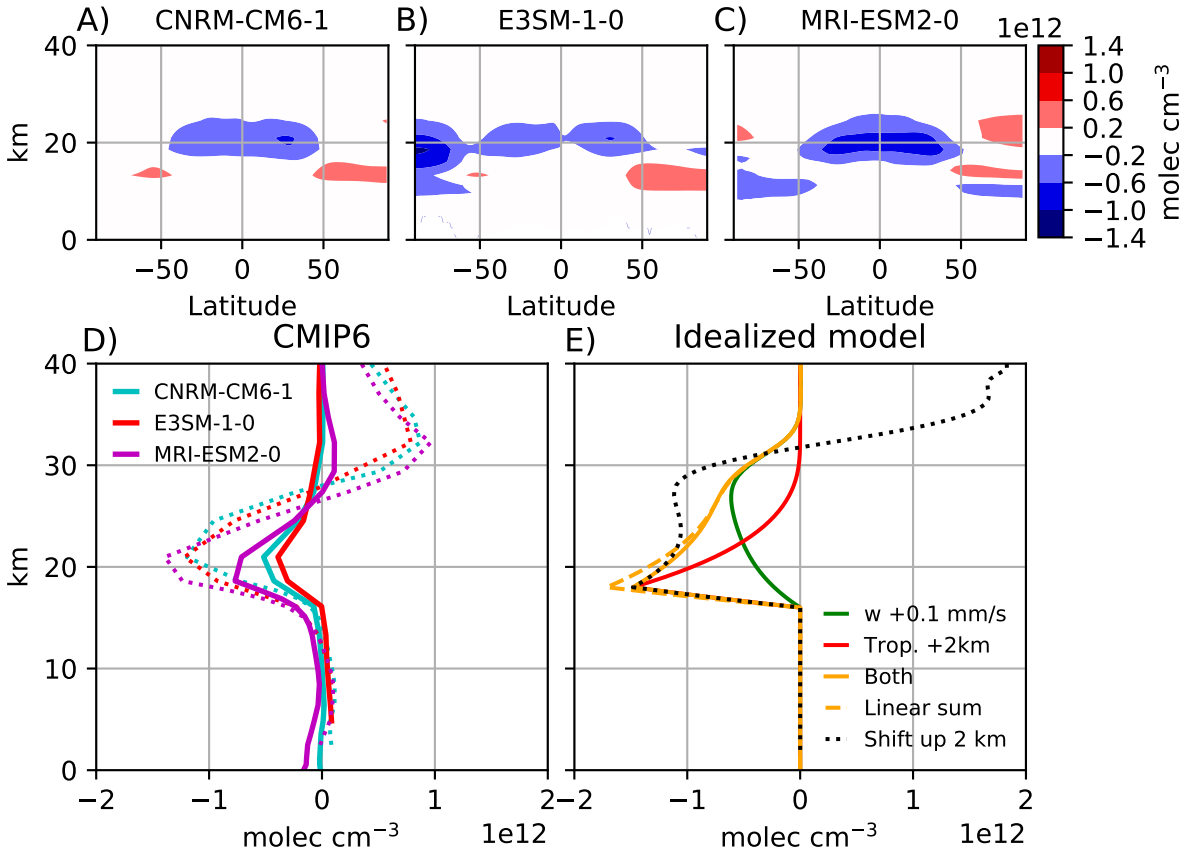

Figure 3. Effect on ozone number densities of surface warming. (A-C) Ozone number densities in amip-p4K minus amip. (D) Tropical average (30S-30N) of panels A-C (solid) and change in ozone number density from shifting the amip ozone profile up by 2 km (dotted). (E) Ozone anomalies in the idealized model in response to strengthening of the Brewer-Dobson circulation by 0.1 mm s^{-1} (green), tropospheric expansion by 2 km (red), both (orange), their linear sum (dashed orange), and an upward shift of the control ozone profile by 2 km (black dotted).

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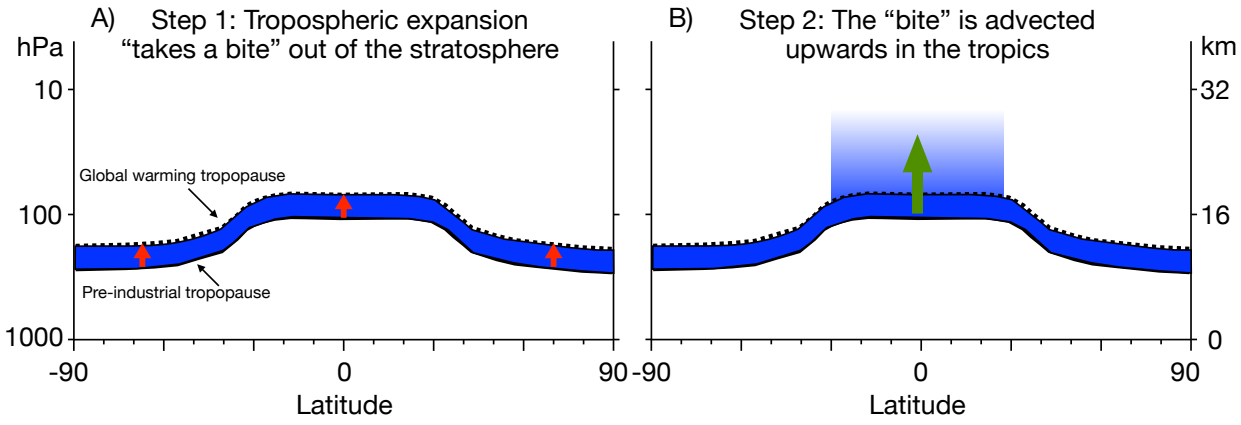

Figure 4. Cartoon illustrating the impact of expansion of tropospheric destruction of ozone. (A) Tropospheric expansion "takes a bite" out of the stratosphere by converting stratospheric air to tropospheric air. (B) The "bite" is advected upwards in the tropics by the background Brewer-Dobson circulation until photochemical equilibrium is gradually re-established.

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