The double dip: how tropospheric expansion counteracts increases in extratropical stratospheric ozone under global warming

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Key Points:

- Under increasing CO_2, models project extratropical stratospheric ozone to increase except around 10 km (lower dip) and 17 km (upper dip).
- The lower dip is due to expansion of the extratropical troposphere, whereas the upper dip is due to expansion of the tropical troposphere.
- The lower dip is strongest in winter when ozone is greatest. The upper dip is strongest in summer when Brewer-Dobson downwelling is weakest.

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Abstract

In response to rising CO_2, chemistry-climate models project that extratropical stratospheric ozone will increase, except around 10 km and 17 km. We call the muted increases or reductions at these altitudes the "double dip". The double dip results not from stratospheric cooling but from surface warming. Using an idealized photochemical-transport model, surface warming is found to produce the double dip via tropospheric expansion, which converts ozone-rich stratospheric air into ozone-poor tropospheric air. The lower dip results from expansion of the extratropical troposphere, as previously understood. The upper dip results from expansion of the *tropical* troposphere, low-ozone anomalies from which are then transported into the extratropics. Large seasonality in the double dip in chemistry-climate models can be explained by seasonality in the Brewer-Dobson circulation. The remote effects of the tropical tropopause on extratropical ozone complicate the use of (local) tropopause-following coordinates to remove the effects of global warming.

Plain Language Summary

In response to rising atmospheric CO_2 primarily from the burning of fossil fuels, beneficial ozone in the mid-latitude stratosphere tends to increase due to changes in the winds and temperature-dependent reaction rates. However, these broad increases in ozone are punctuated by reductions (or muted increases) around 10 km and 17 km, which we term the "double dip". We find that the double dip exists because the warming of the troposphere allows the tops of rainclouds to reach higher altitudes, reducing stratospheric ozone through the injection of ozone-poor tropospheric air. The lower dip around 10 km results intuitively from the deepening of the mid-latitude troposphere. Counterintuitively, the upper dip around 17 km results from deepening of the faraway tropical troposphere, whose remote reductions in tropical ozone are then transported laterally over the mid-latitudes. Thus, the double dip depends on both the local and remote deepening of the troposphere, which could complicate a common practice of filtering out the effects of tropospheric expansion that only considers the local component.

1 Introduction

Whereas the largest anthropogenic effects on the ozone layer have resulted from chemical perturbations due to ozone-depleting substances, the ozone layer is also being

perturbed thermodynamically and dynamically due to rising CO_2. Rising CO_2 leads to both increases and decreases in ozone at different locations, and chemistry-climate models (CCMs) robustly agree on the spatial pattern of this response (e.g., Haigh & Pyle, 1982; Shepherd, 2008; Chiodo et al., 2018; Match & Gerber, 2022). Examples of this robust response are shown from three CCMs in Fig. 1. Ozone is simulated to increase in the mid- to upper-stratosphere (i.e., above the ozone maximum), because stratospheric cooling speeds up the three-body reaction that forms O_3 and slows down certain collisional loss reactions (Haigh & Pyle, 1982; Jonsson et al., 2004). Ozone is simulated to decrease in the tropical lower stratosphere for two main reasons: (1) a strengthening of the Brewer-Dobson circulation (BDC), which upwells ozone-poor air from below (e.g., Shepherd, 2008; Li et al., 2009), and (2) tropospheric expansion, which erodes the ozone layer from below to lead to reductions of ozone that are then upwelled by the climatological BDC (Match & Gerber, 2022).

The ozone response in the extratropical lower stratosphere is less straightforward, as it is not uniform in sign, and, unlike the changes elsewhere, exhibits some sign asymmetries between hemispheres and sign disagreements between models (Fig. 1). Nonetheless, a robust vertical structure of the response is evident: although ozone generally increases in the extratropical lower stratosphere under global warming, these increases are punctuated by two "dips" (i.e., reductions in the magnitude of the increase possibly rising to the level of an absolute decrease): a *lower dip* around 10 km and an *upper dip* around 17 km. We call this response the "double dip". The double dip exists when averaging either poleward of 30°, where the upper dip might be contaminated by a direct contribution from ozone reductions in the tropical upwelling regime, or when averaging poleward of 60°, far from any tropical contamination (Fig. 1, bottom row, solid vs. dashed curves).

The lower dip has been previously described and attributed to tropospheric expansion, which erodes the ozone layer from below by replacing ozone-rich stratospheric air with ozone-poor tropospheric air (Plummer et al., 2010; Dietmüller et al., 2014). The upper dip, on the other hand, has not been the focus of prior work and has not been explicitly discussed despite appearing (sometimes subtly) in figures from numerous previous studies, including Fomichev et al. (2007, their Fig. 12), Shepherd (2008, their Fig. 11), Plummer et al. (2010, their Fig. 2), Dietmüller et al. (2014, their Fig. 1a), Banerjee et al. (2016, their Fig. 1), Chiodo et al. (2018, their Fig. 2), and Keeble et al. (2021, their

Fig. 10). Our goals are to explain the mechanisms that lead to the double dip and frame their implications for interpreting ozone trends in the extratropical lower stratosphere.

We begin by analyzing pairs of CMIP6 experiments that isolate the two pathways by which rising CO_2 affects ozone: stratospheric cooling and surface warming. The double dip will be found to result from surface warming (Section 2). Surface warming is *a priori* understood to affect stratospheric ozone through tropospheric expansion and by strengthening the BDC (the latter arguably connected to the former, e.g., as in Oberländer-Hayn et al. (2016), although our analysis treats their effects separately). To disentangle the effects of tropospheric expansion and the strengthening BDC, we analyze a highly simplified photochemical-transport model within which the tropopause height and BDC strength can be independently varied, an extension of that developed by Match and Gerber (2022) (Section 3). Our results support previous arguments that the lower dip arises from expansion of the extratropical troposphere (Plummer et al., 2010; Dietmüller et al., 2014) (Section 4). The upper dip also arises from tropospheric expansion, but not of the (local) extratropical troposphere but rather of the *tropical* troposphere, reductions in ozone from which are then laterally transported into the extratropics at the altitude of the tropical tropopause around 17 km. We proceed to explain seasonality in the double dip as a consequence of the seasonal cycle of the BDC (Section 5), then discuss implications of these results for the interpretation of tropopause-following coordinates (Section 6).

2 The double dip: surface warming, not stratospheric cooling

The extratropical ozone response to a quadrupling of CO_2 is shown in Fig. 1, and for a quantitative comparison among CCMs in Fig. 2a. The increase of CO_2 is thought to affect the ozone layer by perturbing the thermodynamical and dynamical conditions that determine ozone reaction rates and transport. These effects can be distinguished by considering the separate effects of stratospheric cooling and surface warming, whose contributions to the double dip can be assessed by comparing pairs of chemistry-climate model experiments that isolate each in turn (similar decompositions appear in, e.g., Fomichev et al., 2007; Match & Gerber, 2022). These pairs of experiments are drawn by opportunity from the CMIP6 archive. The pairs of experiments have slightly different background states, pre-industrial for isolating stratospheric cooling versus historical for isolating surface warming.

Surface warming is isolated by comparing two experiments with prescribed sea surface temperatures (SSTs). A standard experiment with prescribed historically-evolving SSTs (amip) is compared to one in which those historical SSTs are uniformly warmed by 4 K (amip-p4K). Warming the SSTs warms the troposphere, but does not lead to stratospheric cooling, which occurs under rising CO_2 due to the direct radiative effects of the enhanced CO_2 in the stratosphere (Manabe & Wetherald, 1967). Surface warming expands the troposphere and strengthens the BDC. Fig. 2b shows the extratropical ozone response to surface warming in three CCMs. Surface warming also leads to a pronounced double dip, with localized reductions around 10 km and 17 km.

To determine whether stratospheric cooling also contributes to the double dip, we isolate its effects. Stratospheric cooling is isolated by comparing a control experiment (piControl) to an experiment in which CO_2 is quadrupled while holding SSTs fixed (piClim-4xCO2). The increase of CO_2 increases the radiative cooling of the stratosphere to lead to stratospheric cooling, whereas the warming effects of the CO_2 in the troposphere are suppressed by keeping surface temperatures fixed. Fig. 2c shows that stratospheric cooling leads to an increase in extratropical ozone above about 17 km, primarily by perturbing the photochemical reaction rates that produce and destroy ozone. The ozone response in the extratropical lower stratosphere is small, and there is no evidence of reductions that would project significantly onto either the upper or lower dip. Therefore, we conclude that the double dip results primarily from surface warming.

3 Methods: A simple model to disentangle contributions from the Brewer-Dobson circulation and tropospheric expansion

In order to understand how surface warming leads to the double dip, it is necessary to mechanistically disentangle its two primary pathways for affecting ozone: tropospheric expansion and strengthening of the Brewer-Dobson circulation. Tropospheric expansion is a direct thermodynamic consequence of global warming (Singh & O'Gorman, 2012; Vallis et al., 2015). Based on the idea that convection must deepen in order for a moist adiabatic parcel launched at the surface to reach an approximately fixed tropopause temperature (Hartmann & Larson, 2002; Seeley et al., 2019; Jeevanjee & Fueglistaler, 2020; McKim et al., 2024), Match and Fueglistaler (2021) derived a simple scaling for tropospheric expansion of 6 hPa per Kelvin of surface warming. The strengthening Brewer-Dobson circulation is a circulation response that also scales with surface warming, and

Oberländer-Hayn et al. (2016) have argued that it is a direct consequence of the upward shift of the circulation under tropospheric expansion, although we can assess their effects independently. To do so, we formulate an idealized photochemical-transport model that distills the processes that control extratropical stratospheric ozone, but allows us to independently prescribe values for the tropopause height and BDC strength.

The model is a Chapman+2 photochemical-transport model, and its formulation draws from two streams of previous work. From the first stream, we adopt a Chapman Cycle-based model of spectrally-resolved UV photochemistry in an isothermal atmosphere with transport between the tropics and extratropics via a leaky tropical pipe, and zero ozone below the prescribed tropopause (Match & Gerber, 2022). From the second stream, we augment our Chapman Cycle reactions with two additional reactions representing generalized catalytic sinks of O and O_3 (the Chapman+2 model: Match et al., 2024a, 2024b). The rates of these generalized catalytic sinks depend on the climatological distribution of catalysts drawn from a chemistry-climate model as tabulated in Brasseur and Solomon (2005). The Chapman+2 model has previously been described in a steady-state formulation with transport parameterized as a damping in the above reference, but here we represent transport explicitly using the leaky tropical pipe. To produce a realistic annual cycle of the double dip, we split the extratropics into a Northern Hemisphere column and a Southern Hemisphere column, and impose an annual cycle in the strength of the BDC, which peaks in the winter in each hemisphere.

The leaky tropical pipe is generally formulated to represent the mid- to upper-stratosphere, where the tropics are isolated from the extratropics (Neu & Plumb, 1999). In order to extend the leaky tropical pipe down to lower altitudes, we impose a jump towards larger lateral mixing below the tropical tropopause, which has the effect of damping ozone in the extratropical lower stratosphere and could be interpreted to represent known lateral mixing pathways above the subtropical jet (e.g. Hoor et al., 2004; Gettelman et al., 2011).

A schematic showing the basic formulation of the resulting Chapman+2 photochemical-transport model is shown in Fig. S1. A detailed description of the model and our numerical approach is provided in Texts S1 and S2. The climatological seasonal cycle of ozone in our model is shown in Fig. S2, indicating a favorable comparison to that from the chemistry-climate model MRI-ESM2-0.

Perturbations are applied to the Chapman+2 photochemical transport model to represent the three key effects of rising CO_2. Stratospheric cooling, represented as a uniform cooling of 10 K, perturbs the temperature-dependent reaction rates for the Chapman+2 reactions. Strengthening of the Brewer-Dobson circulation, represented by a uniform increase of $\bar{w}^*$ by 0.05 mm s^{-1}, perturbs transport by the leaky tropical pipe. Tropospheric expansion, represented by a 1 km upward shift of the tropopause and lateral mixing, takes a bite out of the ozone layer from below that is then transported by advection and mixing. Together, these perturbations will be shown to approximate the response to a quadrupling of CO_2 (see also Match & Gerber, 2022). Critically for our understanding, they can be imposed separately or in various combinations to emulate experiments from the CMIP6 models, allowing us to assess the linearity of the response, and ultimately disentangle the relative contributions to the double dip.

Omitted in our model is another proposed decadal driver of extratropical ozone trends: changes in the latitudinal structure of two-way mixing. An idealized model suitable to represent such changes would require higher latitudinal resolution. This caveat prevents our model from reproducing this particular proposed mechanism for recent unexpected declines in extratropical lower stratospheric ozone (Ball et al., 2018; Wargan et al., 2018; Ball et al., 2020; Orbe et al., 2020).

4 Results: The double dip is due to tropospheric expansion

The Chapman+2 photochemical-transport model was designed to emulate the extratropical ozone changes in chemistry-climate models in response to surface warming and/or stratospheric cooling. To establish its fitness, we show three validation benchmarks, one in each row of Fig. 2. The model successfully reproduces the qualitative structure of the extratropical stratospheric ozone response to each perturbation. In response to simultaneous surface warming and stratospheric cooling, ozone increases aloft and there is a double dip in the lower stratosphere (Fig. 2a vs. 2d). Isolating the response to surface warming retains the double dip, now with only small ozone changes above 20 km (Fig. 2b vs. 2e). Isolating the response to stratospheric cooling retains the increased ozone above 20 km, but without the double dip (Fig. 2c vs. 2f).

The success of the Chapman+2 model in these mechanism denial experiments builds confidence that it can also further decompose the response to surface warming into the

distinct effects of a strengthening BDC and tropospheric expansion. This decomposition is shown in Figs. 2d and 2e. Consistent with prior literature, the strengthening BDC (magenta curves) increases ozone in the extratropical lower stratosphere (e.g., Shepherd, 2008). These increases arise from stronger downwelling of ozone-rich air that can equilibrate at a higher concentration against sinks from photochemistry and mixing. The leaky tropical pipe framework also captures the increase in lateral transport from tropics to extratropics. Potential reductions of extratropical ozone from this enhanced lateral transport of ozone-poor air from the tropics are overwhelmed by the increases of ozone from enhanced downwelling.

With the strengthening BDC leading to broad increases of ozone, the double dip must instead result from tropospheric expansion (Fig. 2d and 2e, red and cyan curves). As others have argued, expansion of the extratropical troposphere leads directly to the lower dip by eroding the ozone layer from below (Plummer et al., 2010; Dietmüller et al., 2014) (cyan curves). A new result of this figure is that the upper dip comes from expansion of the *tropical* troposphere (red curves). The expanding tropical tropopause erodes the ozone layer in the tropical lower stratosphere, low ozone anomalies from which are then advected and mixed into the extratropical lower stratosphere. As represented in our model, tropical tropospheric expansion induces an upper dip due to both the upward shift in tropical tropospheric destruction of ozone and the upward shift in the lateral mixing profile that jumps to larger values below the tropical tropopause. These two effects both contribute at leading order, as seen in the decomposition of Fig. S3. In total, the column-integrated response to (remote) tropical tropospheric expansion is not only significant but is actually several factors larger than the response to (local) extratropical tropospheric expansion.

The double dip is thus a function of both local (extratropical) and remote (tropical) tropospheric expansion. The lower dip occurs around 10 km at the altitude of the extratropical tropopause, and the upper dip is centered around 17 km at the altitude of the tropical tropopause. The gap of approximately 7 km separating the two dips therefore reflects the tropopause break in the vicinity of the subtropical jet, whereby the tropopause drops discontinuously between the tropics and the extratropics. A pathway of lateral mixing from the tropical lower stratosphere into the extratropical lower stratosphere has previously arisen in other contexts, such as when considering transport of short-lived sub-

stances and idealized tracers (Hoor et al., 2004; Bönisch et al., 2009; Gettelman et al., 2011; Abalos et al., 2017).

These results suggest that tropospheric expansion is uniquely responsible for the double dip. This result is quantitative but also qualitative; the strengthening BDC and stratospheric cooling strictly increase ozone without imparting much structure from the tropopause onto their responses, whereas tropospheric expansion strictly reduces ozone while strongly imprinting tropopause structure. The singular role of tropospheric expansion in leading to the double dip can be contrasted to the case of tropical ozone changes under global warming, in which tropospheric expansion and the strengthening BDC reinforce each other in leading to reductions of ozone (Match & Gerber, 2022), making the attribution of their relative contributions a quantitative question.

5 Results: Seasonality of the double dip due to seasonality of the Brewer-Dobson circulation

So far, we have presented results and mechanistic arguments that appeal to the annually-averaged double dip. However, the double dip is not predicted in CCMs to exist year-round. Figs. 3a (Northern Hemisphere) and S4a (Southern Hemisphere) show that the lower dip is strongest in winter, whereas the upper dip is strongest in summer and vanishes in winter. This seasonal cycle is evident in both hemispheres and in other chemistry-climate models (Fig. S5). Compared to the time-averaged double dip, the seasonal cycle provides a more stringent test of our explanatory framework.

There are many seasonally-varying aspects of ozone photochemistry and transport that could be important for the modulating the double dip. On the photochemistry side, there are annual cycles in solar zenith angle, catalyst concentrations, and temperature-dependent reaction rates, among others. On the transport side, there are annual cycles in tropopause height and the strength of the Brewer-Dobson circulation, which is strongest in winter due to the enhanced planetary wave activity propagating up from the troposphere (e.g., Holton et al., 1995; Butchart, 2014). There is also an annual cycle in the lateral mixing from the tropical tropopause layer above the subtropical jet into the extratropical lower stratosphere, which maximizes during summer associated with the Asian summer monsoon anticyclone (e.g. Hoor et al., 2004; Gettelman et al., 2011; Stolarski et al., 2014). Although all of these photochemical and transport factors could in principle contribute significantly to the seasonal cycle in the double dip, here we show that

just one factor—the seasonal cycle in BDC strength—is sufficient to reproduce the seasonality of the double dip. This does not, however, rule out contributions from other factors.

Figure 3 shows the seasonally-resolved response of ozone in the Northern Hemisphere extratropics in the Chapman+2 photochemical-transport model to stratospheric cooling, strengthening BDC, tropospheric expansion, and all three together, compared to the Northern Hemisphere ozone response to an abrupt quadrupling of CO_2 in MRI-ESM2-0. We emphasize that the only seasonally-varying boundary condition of the Chapman+2 photochemical-transport model is the strength of the BDC, which varies sinusoidally from zero at the summer solstice to twice the annual mean value at the winter solstice (consistent with reanalyses, e.g., Seviour et al., 2012).

The lower dip is stronger in winter and weaker in summer (Fig. 3a,b), and this seasonality is easier to explain than that of the upper dip. The Chapman+2 photochemical-transport model suggests that this seasonality comes from seasonality in the effects of tropospheric expansion (Fig. 3i,j), and less so from the effects of stratospheric cooling or strengthening BDC. The seasonality of the ozone reduction in the lower dip depends simply on how much ozone is present to be eroded away: the lower dip is larger in winter because there is more ozone in the lowermost stratosphere during that time. The idealized model has more ozone in winter because of the seasonally stronger downwelling of the BDC.

The seasonality of the upper dip is more subtle to explain. This is because there is not much seasonality in the response to tropical tropospheric expansion itself (Fig. 3k,l). In the simple model, the seasonality of the upper dip comes from seasonality in the response to stratospheric cooling and the strengthening BDC, both of which exhibit larger increases of ozone in winter that mask the upper dip. Stratospheric cooling leads to a larger increase of mid-stratospheric ozone in winter than in summer (Fig. 3e) because the elevated ozone in the upper stratosphere from stratospheric cooling is downwelled more strongly in winter. Strengthening the Brewer-Dobson circulation leads to a stronger increase of ozone in winter because our prescribed fractional increase in the BDC is largest then. (Our seasonally-varying downwelling varies from zero on July 1 to $2\overline{w_j^*}$ on January 1, so strengthening the BDC by increasing $\overline{w_j^*}$ has a peak absolute effect on January 1 and no effect on July 1.) An open question is whether seasonality in lateral mixing, such

as from the Asian summer monsoon anticyclone could be a leading-order contributor to seasonality in the upper dip, a contribution that could be explored in future work. In summary, the upper dip vanishes in winter because it is masked by annually maximal increases of ozone from stratospheric cooling and the strengthening BDC, which both follow as a direct consequence of the climatological peak of the BDC. A schematic of this mechanistic understanding is shown in Fig. 4.

The seasonal cycle of the ozone response to a quadrupling of CO_2 includes large cancellation among opposing terms, so it is not surprising that the sign of the ozone response to a quadrupling of CO_2 is not robustly simulated in this region (Fig. 1). Although the sign is not robust, this paper demonstrates that key aspects of the pattern of the response, namely the double dip, can be understood. The sign of the response at each location and throughout the seasonal cycle could be a sensitive indicator for the effects of model disagreements in stratospheric cooling, the strengthening BDC, and tropospheric expansion.

Rising CO_2 is not the only perturbation that will affect the ozone layer in the coming decades. Ongoing recovery of the ozone hole due to the Montreal Protocol could potentially obscure part of the double dip. Fig. S6 compares ozone in two chemistry-climate models (MRI-ESM2-0 and CNRM-ESM2-1) between 2015-2044 and 2071-2100 in the high-development and high-emissions pathway of ssp585. The annually-averaged change in ozone is plotted as well as the change in only DJF or JJA. Recovery of polar ozone from declining CFCs generally dominates the response, although the upper dip from surface warming is evident in the Northern Hemisphere during JJA for both models.

6 Discussion: Implications for filtering global warming using tropopause-following coordinates

Filtering out trends in ozone from global warming is of great interest because the residual time series may help reveal the chemical recovery of the ozone layer due to declining ozone-depleting substances (Petropavlovskikh et al., 2019). A growing practice intended to remove the impact of warming is to transform ozone trends into tropopause-following coordinates, based on the understanding that the tropopause rises under global warming (Thompson et al., 2021). The use of tropopause-following coordinates has proceeded with different methods reflecting different assumptions. Some studies assess ozone trends in tropopause-following coordinates through most of the stratosphere (Wargan

et al., 2018; Bognar et al., 2022), while others restrict tropopause-following coordinates to an empirically-determined region within roughly 5 km of the tropopause (Pan et al., 2004; Hegglin et al., 2008; Millán et al., 2024). The results in this paper suggest precautions towards each approach.

Using tropopause-following coordinates throughout the stratosphere assumes that ozone is conserved with respect to the local tropopause under dynamical perturbations in tropopause height. Above 25-30 km, however, the ozone layer is typically in photochemical equilibrium (e.g., Perliski et al., 1989; Brasseur & Solomon, 2005) where it is unaffected by dynamical anomalies in ozone due to local tropopause variability (Match & Gerber, 2022).

Restricting attention to the dynamically-controlled regime below 25-30 km, we have shown that if both tropopauses rise equally, the resulting change in ozone can be approximated by a shift with respect to the local tropopause (Figs. 2, 3, S7). Yet, this only works for a uniform rise in both tropopauses, which is not necessarily expected in response to warming. Non-uniform tropopause changes introduce problems: if only the tropical tropopause rises, tropopause-following coordinates in the extratropics cannot capture the resulting upper dip (Fig S7b); if only the extratropical tropopause rises, tropopause-following coordinates predict a spurious upper dip (Fig. S7c). There does not exist a single tropopause-following coordinate that can filter out arbitrary changes in tropopause structure.

Restricting tropopause-following coordinates to an empirically-determined window near the tropopause can avoid contamination from the photochemically-controlled region, but introduces other challenges. The empirical window is often chosen by using past data of ozone and tropopause heights to identify where tropopause-following coordinates reduce the variance of ozone compared to absolute height coordinates (Hegglin et al., 2008; Millán et al., 2024). This empirical window thus demarcates where the variability in ozone is dominated by variability in extratropical tropopause height, and has generally been found to extend 2-5 km above the extratropical tropopause. Yet, because the empirical window excludes most of the the upper dip, a major part of the warming response occurs outside its frame.

7 Conclusions

The extratropical stratospheric ozone response to rising CO_2 has a robust shape: increases in ozone throughout the stratosphere are punctuated by two dips, i.e., reductions in the size of increase, potentially large enough to yield absolute reductions. The upper dip is at 17 km and is strongest in summer, and the lower dip is at 10 km and is strongest in winter. With the use of CMIP6 chemistry-climate model results and the Chapman+2 photochemical-transport model, the double dip has been explained as follows:

- The lower dip results from expansion of the extratropical troposphere. The lower dip is strongest in winter when extratropical lower stratospheric ozone is largest.
- The upper dip results from expansion of the remote tropical troposphere. The upper dip is strongest in summer, whereas it is masked in winter by the annually maximal increases in ozone from stratospheric cooling and the strengthening BDC.

The sensitivity of extratropical lower stratospheric ozone to both local and remote properties of the tropopause complicates the growing practice of using local tropopause-following coordinates to filter out the effects of changes in tropopause height on ozone.

8 Data Availability Statement

Python software version of the Chapman+2 photochemical-transport model is publicly available at `10.5281/zenodo.13412270`, along with the run script used to produce the main experiments analyzed herein. CMIP6 data is freely accessible from `https://esgf-node.llnl.gov/search/cmip6/`.

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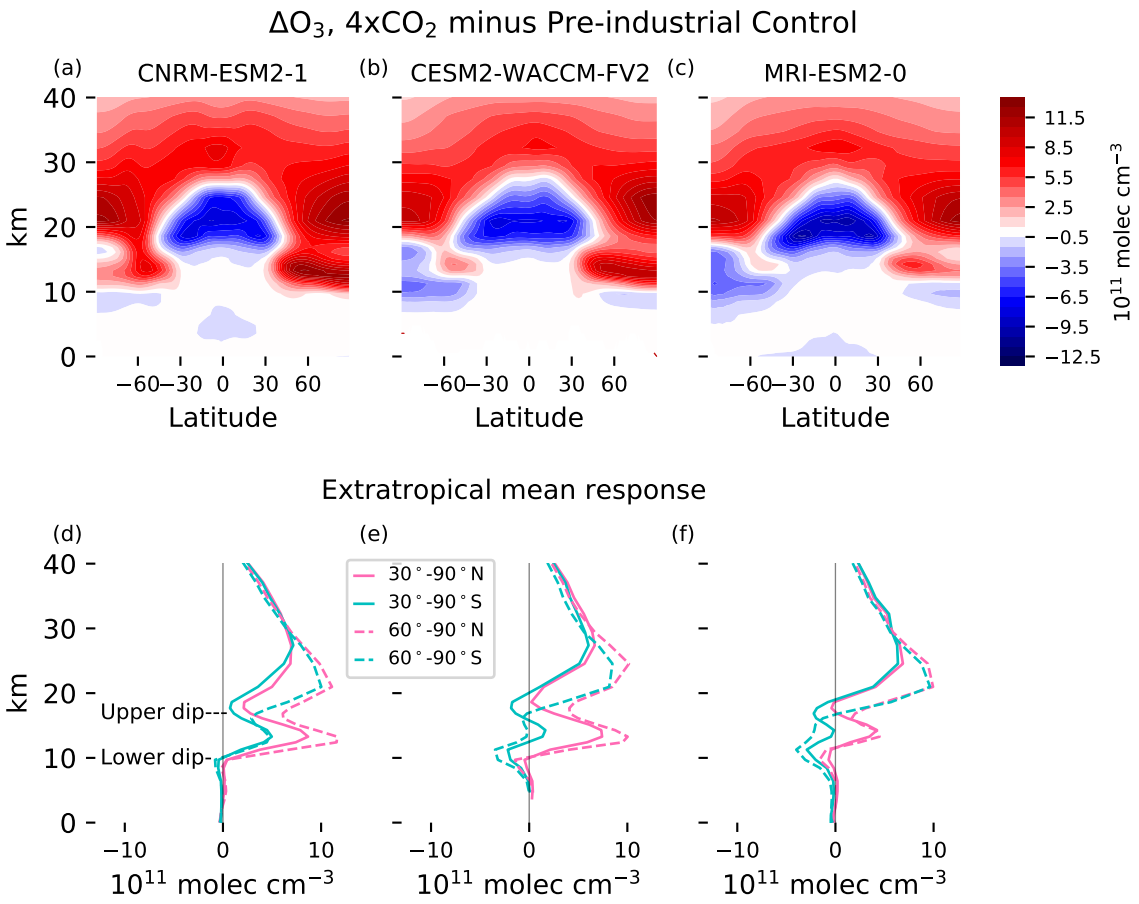

Figure 1. The double dip is evident in the response of extratropical O_3 to a quadrupling of CO_2 in three CMIP6 models with interactive chemistry. Top row: Change in [O_3] in abrupt-4xCO2 (years 50-150) minus piControl. Bottom row: Extratropical mean changes in [O_3] in the Northern Hemisphere (pink) and Southern Hemisphere (cyan) when averaged poleward of 30° (solid) or poleward of 60° (dashed). The lower dip occurs around 10 km, and the upper dip occurs around 17 km.

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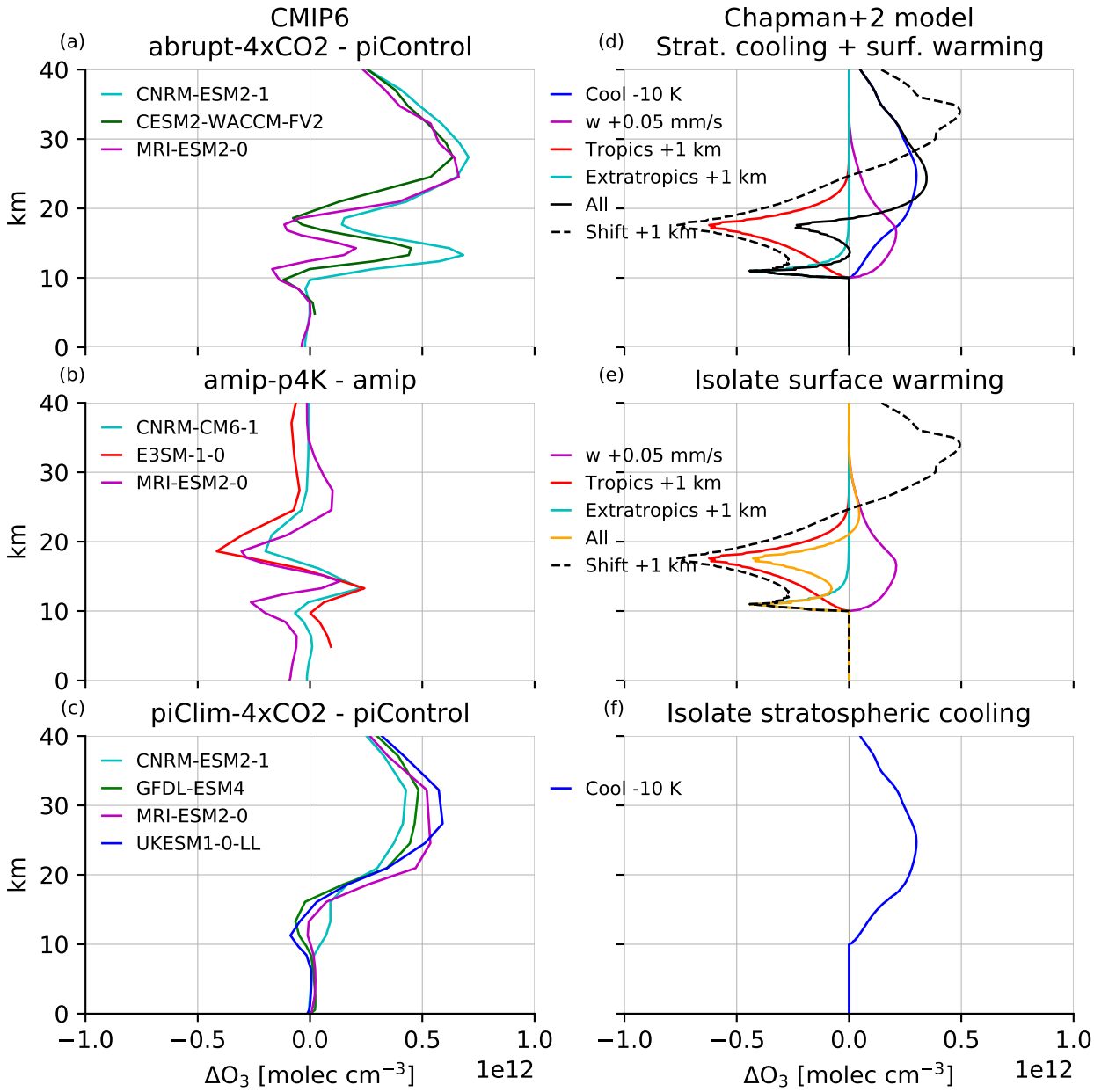

Figure 2. Decomposing the mechanisms by which increasing CO_2 affects extratropical O_3 in CMIP6 models (left column) and the Chapman+2 model (an idealized photochemical-transport model, right column). (a) Response of extratropical O_3, averaged poleward of 30°, to abrupt-4xCO2 minus piControl in the three CMIP6 models shown in Fig. 1. (b) As above, but isolating surface warming through amip-p4K minus amip. (c) As above, but isolating stratospheric cooling through piClim-4xCO2 minus piControl. The double dip is due to surface warming and not stratospheric cooling. (Right column) Extratropical O_3 response to the key mechanistic drivers of the response: stratospheric cooling of 10 K (blue), strengthening Brewer-Dobson circulation by 0.05 mm s^{-1} (magenta), expansion of the tropical troposphere by 1 km (red), expansion of the extratropical troposphere by 1 km (cyan), and all together (black). (Dashed black) Change in O_3 from a 1 km upward shift of the control profile. The lower dip is due to expansion of the extratropical troposphere, and the upper dip is due to expansion of the tropical troposphere.

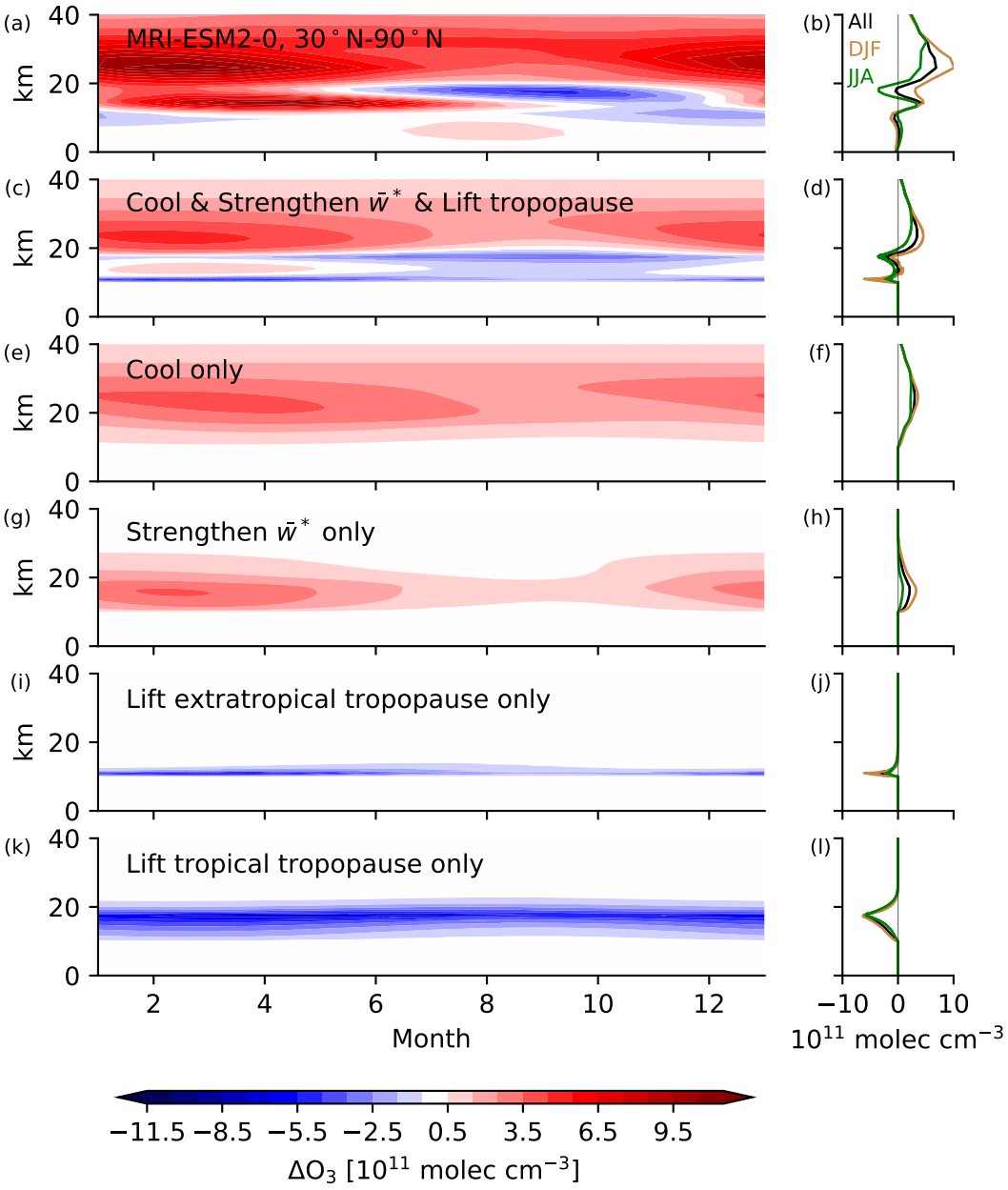

Figure 3. Mechanistic decomposition of the annual cycle in the Northern Hemisphere extratropical O_3 response to global warming. (Left column) (a) MRI-ESM2-0 for abrupt-4xCO2 minus piControl and (c,e,g,i,k) Chapman+2 mechanism denial experiments, in which all seasonality arises solely from a prescribed annual cycle in the Brewer-Dobson Circulation (strongest in winter of each hemisphere). In MRI-ESM2-0, the upper dip around 17 km is strongest in summer whereas the lower dip around 10 km is strongest in winter, with both aspects reproducible from the Chapman+2 model whose sole seasonally-varying driver is BDC strength. (Right column) Temporal average of the left column across All months (black), DJF (brown), and JJA (green).

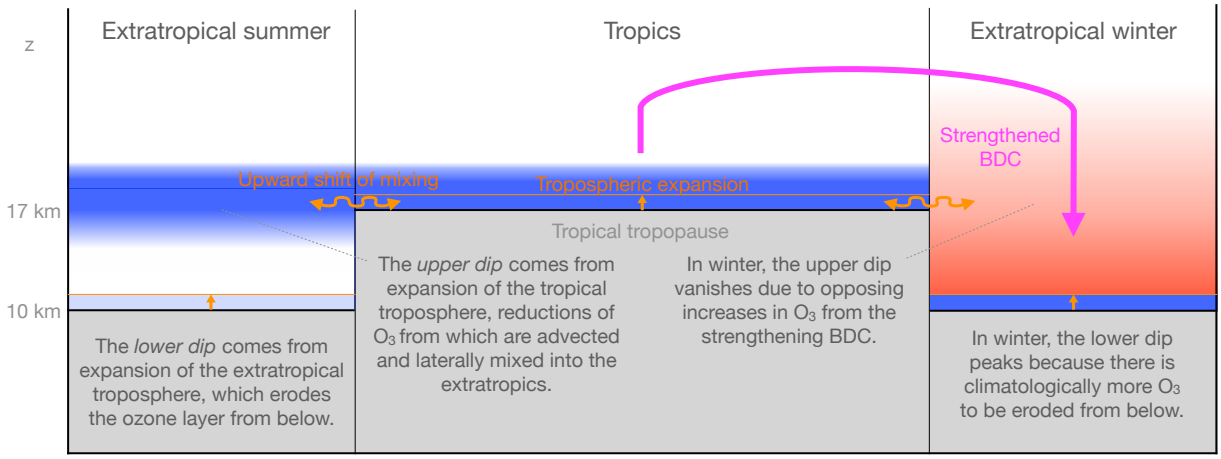

Figure 4. Schematic illustrating how surface warming leads to each dip and its seasonality. Reductions of ozone are in blue and increases are in red.