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Key Points:
- Chemistry feedbacks not critical in determining equilibrium climate sensitivity in CESM
- Increased CO_2 levels lead to substantial changes in ozone from the upper troposphere to the stratopause
- Chemistry feedbacks do not drive large tropospheric water vapor changes

Supporting Information:
- Supporting Information S1

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Stratospheric ozone chemistry feedbacks are not critical for the determination of climate sensitivity in CESM1(WACCM)

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Abstract The Community Earth System Model-Whole Atmosphere Community Climate Model (CESM1-WACCM) is used to assess the importance of including chemistry feedbacks in determining the equilibrium climate sensitivity (ECS). Two $4\times CO_2$ model experiments were conducted: one with interactive chemistry and one with chemical constituents other than CO_2 held fixed at their preindustrial values. The ECS determined from these two experiments agrees to within 0.01 K. Similarly, the net feedback parameter agrees to within 0.01 W m^{-2} K^{-1}. This agreement occurs in spite of large changes in stratospheric ozone found in the simulation with interactive chemistry: a 30% decrease in the tropical lower stratosphere and a 40% increase in the upper stratosphere, broadly consistent with other published estimates. Off-line radiative transfer calculations show that ozone changes alone account for the difference in radiative forcing. We conclude that at least for determining global climate sensitivity metrics, the exclusion of chemistry feedbacks is not a critical source of error in CESM.

1. Introduction

As the number of Earth system models that include interactive chemistry increases, it becomes important to understand if this class of models has a markedly different response to increasing greenhouse gases than those that have typically been used to study climate change. The presence of a strong feedback due to the variations in composition induced by a change in radiative forcing (RF) could lead to a significant revision of estimated warming predicted under various future emission scenarios. A model's "climate sensitivity," typically defined as the change in global mean surface air temperature in response to a change in RF, is often assessed through idealized model simulations. One measure of the climate sensitivity due to RF from increasing CO_2 concentrations is the transient climate response (TCR), which is determined from simulations where CO_2 is increased at a rate of 1% yr^{-1} until doubling. Another measure is the equilibrium climate sensitivity (ECS) derived from simulations where CO_2 is abruptly increased to double or quadruple its preindustrial value [see, e.g., *Andrews et al.*, 2012; *Gregory*, 2004; *Forster et al.*, 2013].

In the study by *Muthers et al.* [2014], TCR was computed, with and without chemistry, in the SOCOL (SOlar Climate Ozone Links) model. TCR was determined from simulations where CO_2 was increased while all other forcings were fixed at 1990 levels. They found a small negative effect from including interactive chemistry on the order of −0.1 K (∼4%). The SOCOL ECS determined from abrupt $4\times CO_2$ experiments was 7.5 K with interactive chemistry and 8 K without, i.e., a negative chemistry feedback that reduces the model's ECS by 7%. This chemistry feedback appears to be consistent with the study by *Dietmüller et al.* [2014] using the ECHAM (European Centre/Hamburg)/MESSy (Modular Earth Submodel System) Atmospheric Chemistry (EMAC) model. They showed a decrease in ECS of 3.4% and 8.4% for $2\times CO_2$ and $4\times CO_2$ experiments, respectively. However, *Dietmüller et al.* [2014] suggest that the reduction in sensitivity in their model comes from large changes in stratospheric ozone, while changes in SOCOL ozone were <5%.

More recently, *Nowack et al.* [2014] reported a 1 K reduction in the global mean surface temperature 75 years after quadrupling CO_2 when interactive chemistry was included in the United Kingdom Met Office Unified Model version 7.3 coupled to the United Kingdom Chemistry and Aerosols (UKCA) scheme. As in the study by *Dietmüller et al.* [2014], the difference in sensitivity was attributed to changes in ozone in the upper troposphere and lower stratosphere. In their model, this leads to large changes in stratospheric water vapor and changes in cirrus clouds. A 1 K reduction is equivalent to 20% of the ECS in that model: if that result were

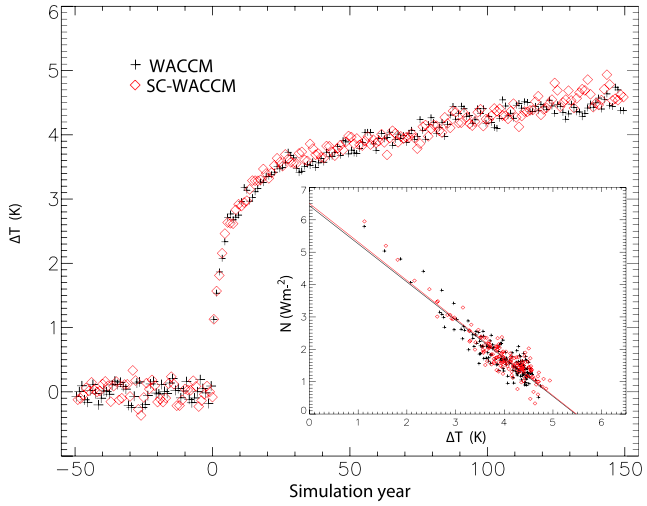

Figure 1. Time series of annual mean global surface temperature anomalies (K) for WACCM (black) and SC-WACCM (red). CO_2 concentrations were quadrupled at year 0. Inset panel shows annual and global mean top of the atmosphere net radiative flux (*N*) plotted against the surface temperature anomalies. Solid lines are linear regression fits.

representative of all Earth system models, it would imply a large bias in the simulated global temperature response to increasing greenhouse gases in models without interactive chemistry.

The aim of this paper is to examine the robustness of such a large ozone negative feedback on surface warming from increased CO_2. For this, we use a state-of-the-art stratosphere-resolving Earth system model, namely, the National Center for Atmospheric Research (NCAR) Community Earth System Model [*Hurrell et al.*, 2013] with the Whole Atmosphere Community Climate Model (CESM1-WACCM) as its atmosphere component. We confirm that there are significant changes in ozone from the upper troposphere to the stratopause. However, we find that these changes lead to statistically insignificant changes in the ECS, suggesting that models without interactive chemistry may still be used to reasonably determine ECS.

2. Experiment Description

We perform canonical CO_2-quadrupling climate sensitivity experiments using two versions of CESM1-WACCM: one with active chemistry [*Marsh et al.*, 2013] and the other with chemistry specified from a previous model simulation. The active-chemistry model version used here is identical to that used to conduct simulations as part of the Coupled Model Intercomparison Project Phase 5 [*Taylor et al.*, 2012], the climate of which is described by *Marsh et al.* [2013] and is referred to here as simply WACCM. The chemistry module in WACCM solves for the concentration of 59 species and includes both gas phase and heterogeneous chemistry. The model resolution is 1.9° latitude by 2.5° longitude, and the model top level is 5×10^{-6} hPa (approximately 140 km). The specified chemistry (SC) version (designated as SC-WACCM) [*Smith et al.*, 2014] uses the same model resolution, vertical grid, and physical parameterizations as WACCM but specifies the concentrations of ozone. SC-WACCM has been shown to have a tropospheric and stratospheric mean state and variability that is nearly identical to WACCM. It should be noted that the stratospheric water vapor distribution in both models is quite similar, since both models include sources from methane oxidation, unlike in the model used by *Muthers et al.* [2014] where the water vapor concentrations were up to 37% larger in the SOCOL model with interactive chemistry. The other active components in our model (land, ocean, and sea ice) are identical between WACCM and SC-WACCM. Our climate sensitivity simulations use as their initial conditions the model state from extended preindustrial (PI) control simulations. The global mean CO_2 concentration is fixed at 285 ppm, and SC-WACCM uses ozone fields from the WACCM PI control simulation. Both control simulations show no discernible trend in the global mean temperature and have long-term mean net radiative fluxes at the top of the atmosphere that are near zero (as shown in Figure 1 and Table 1).

3. Results

Figure 1 shows the annual mean global surface temperature anomaly (ΔT) time series for WACCM (black) and SC-WACCM (red) experiments. Anomalies for each experiment are calculated relative to the mean over the

Table 1. Climate Sensitivity and Feedback Components Derived From Abrupt $4\times CO_2$ Increase Experiments for WACCM and SC-WACCM

	Radiative Forcing (W m^{-2})				Feedbacks $-\alpha$ (W m^{-2} K^{-1})			
	PI TOA (W m^{-2})	$4\times CO_2$	ΔF_{O_3} [a]	ECS (K) $2\times CO_2$	LWC	SWC	CRE Derived	Net
WACCM	−0.05	6.44	−0.08	2.73	−1.90	0.88	-0.16	−1.18
SC-WACCM	0.01	6.50	NA	2.74	−1.85	0.87	−0.21	−1.19

[a]Ozone RF calculated offline using PORT.

last 50 years of their PI controls (i.e., WACCM and SC-WACCM anomalies were calculated separately), although the PI control 50 year mean temperatures matched to within 0.01°C. The mean temperature anomaly time series from the two model configurations are very similar. Over the last 50 years of the 150 year abrupt CO_2 increase simulations, the mean anomaly is 4.41 K for the run with interactive chemistry and 4.49 K with specified chemistry. Also in Figure 1, we show the top of the atmosphere (TOA) net radiative fluxes (N) plotted against the surface temperature anomalies. Following the methodology of *Gregory* [2004], the strength of the model's net feedback parameter α is the slope of the line determined from linear regression of ΔT and N fit to the following equation:

$$N = \mathrm{RF} - \alpha \Delta T \tag{1}$$

The ECS and imposed radiative forcing (RF) are determined from the ΔT and N intercepts, respectively, and are listed in Table 1. It is immediately clear from the fact that the fit lines are nearly overlapping that these three parameters (ECS, RF, and α) are almost identical in the two model configurations. The equivalent $2\times CO_2$ ECS agrees to within 0.01 K, the net feedback parameter (α) to within 0.01 W m^{-2} K^{-1}, and F to within 0.06 W m^{-2}. The differences between the two models are all less than 1% of the actual WACCM or SC-WACCM values.

The net feedback parameter, α, can be broken down into its clear-sky longwave (LWC) and shortwave (SWC) components and the cloud radiative effect (CRE). LWC and SWC feedbacks are calculated in the same manner as α using the net TOA shortwave and longwave fluxes (plotted versus ΔT in Figure 2). CRE is determined as the difference between the net feedback and net clear-sky feedbacks. The calculated feedbacks with and without interactive chemistry are also listed in Table 1. Here we see a slight differentiation between the two models in the LWC feedback, with interactive chemistry amplifying the negative feedback by about 2.6%. In addition, the regression lines are slightly offset, implying that the clear-sky RF is slightly different between the two simulations. The LW clear-sky RF estimated from the regression line intercepts is around 0.24 W m^{-2} less in the model with interactive chemistry. This is partly balanced by a 0.11 W m^{-2} increase in the SW clear-sky RF, leading to an estimated net clear-sky RF difference of −0.13 W m^{-2} (we show below that this difference is comparable to what we might expect from the RF of the $4\times CO_2$-induced ozone changes).

Since the studies by *Dietmüller et al.* [2014] and *Nowack et al.* [2014] attribute a relatively stronger chemistry feedback to changes in ozone, we next examine how ozone changes in WACCM in response to a quadrupling

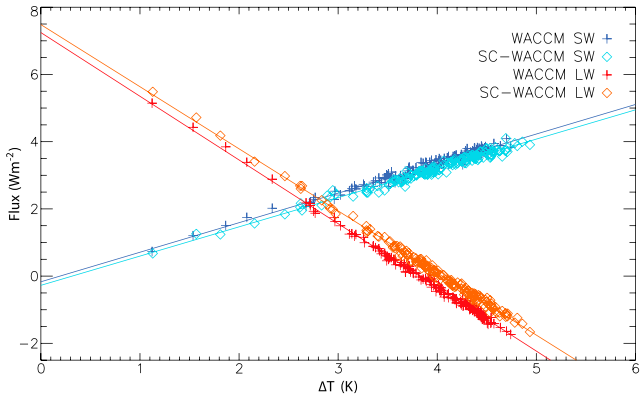

Figure 2. Changes in global mean clear-sky shortwave and longwave radiative fluxes versus changes in global mean surface temperature (ΔT) for WACCM and SC-WACCM for the 150 years following an abrupt quadrupling of CO_2. Solid lines are linear regression fits.

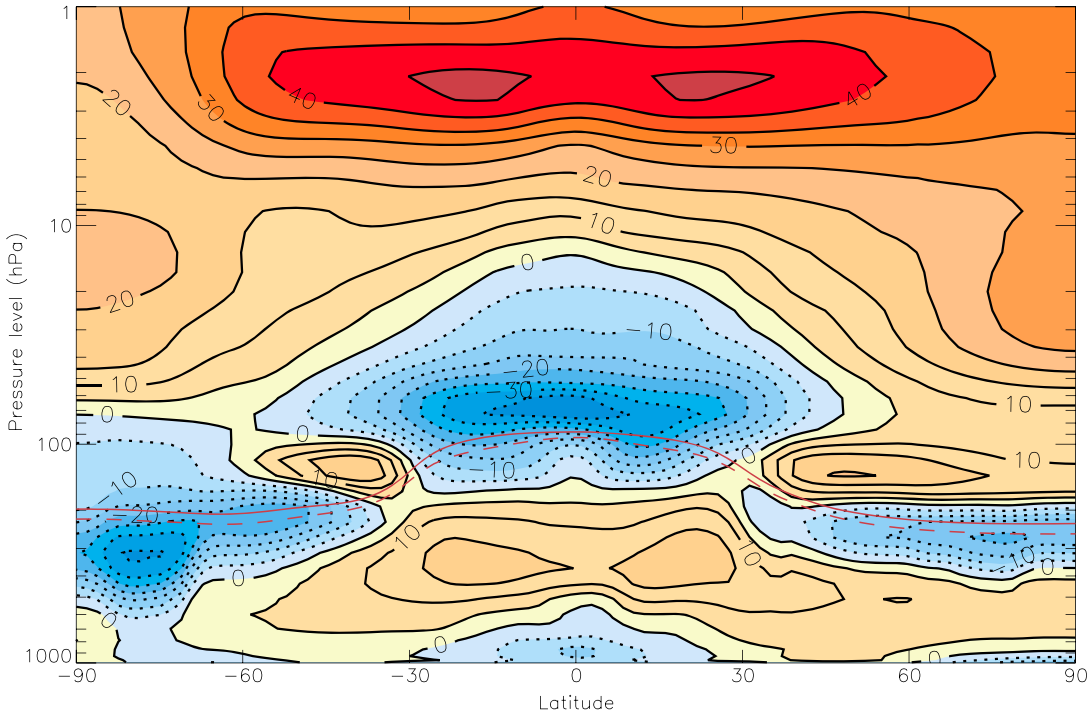

Figure 3. Percentage changes in zonal mean ozone averaged over years 100–149 in $4\times CO_2$ experiment relative to PI values in simulations with interactive chemistry. Solid red line is $4\times CO_2$ tropopause level and dashed red line PI tropopause level.

of CO_2. The ozone differences over the last 50 years of the interactive chemistry simulation relative to the fixed ozone fields used in the SC-WACCM simulation (themselves taken from the interactive PI control) are presented in Figure 3. In the upper stratosphere at latitudes less than 50° one can see a large (>40%) increase in ozone. The causes of this increase are the ~15 K cooling of the upper stratosphere due to increased CO_2 (shown in the supporting information), and the resulting slowdown of the ozone recombination reaction $O + O_3 \rightarrow 2O_2$, the rate constant of which is proportional to $\exp(-2060/T)$ [*Sander et al.*, 2006]. Additionally, cooler temperatures reduce the catalytic losses with nitrogen species and increase the conversion of atomic oxygen to ozone. In the lower stratosphere, one can see a decrease in ozone, approximately 30%, throughout the tropics; this is caused by an increase in tropical upwelling rates associated with an acceleration of the Brewer-Dobson circulation [see, e.g., *Hardiman et al.*, 2014]. We note here that the vertical offset of the lines in Figure 2 is consistent with the rapid adjustment of the distribution of stratospheric ozone in the WACCM model and the resulting change in RF, rather than a slow adjustment which would tend to change slope of the line rather than its $\Delta T = 0$ intercept. Radiative forcing calculations from ozone changes were performed using the Parallel Offline Radiative Transfer (PORT) model and the methodology of *Conley et al.* [2013] (see also *Gilford et al.* [2016] for a comparison of PORT with a line by line calculation). In these calculations, the temperatures above the tropopause are allowed to adjust under the assumption of fixed dynamical heating. As indicated in Table 1, the net (combined shortwave and longwave) all-sky forcing associated with the ozone changes from Figure 3 is approximately -0.08 W m^{-2}, which is slightly larger than the total forcing. For clear-sky conditions the ozone forcing is estimated to be -0.13 W m^{-2}, consistent with the offsets of the lines found in Figure 2 at $\Delta T = 0$.

4. Discussion

The pattern of differences in ozone shown in Figure 3 is remarkably similar to the one presented by *Nowack et al.* [2014] (i.e., decreases in the tropical lower stratosphere and increases near the stratopause). In our model the magnitude of the reduction in ozone in the lower stratosphere is smaller than in *Nowack et al.* [2014] who found a decrease in excess of 50%. In contrast, the increase in the upper stratosphere is slightly larger in WACCM. Both models predict on the order of 10% increases in the middle tropical troposphere and midlatitude upper stratosphere and lower stratosphere (UTLS) ozone. The pattern of the ozone response also agrees with the one presented by *Dietmüller et al.* [2014] for the case of $2\times CO_2$, albeit with a much larger amplitude

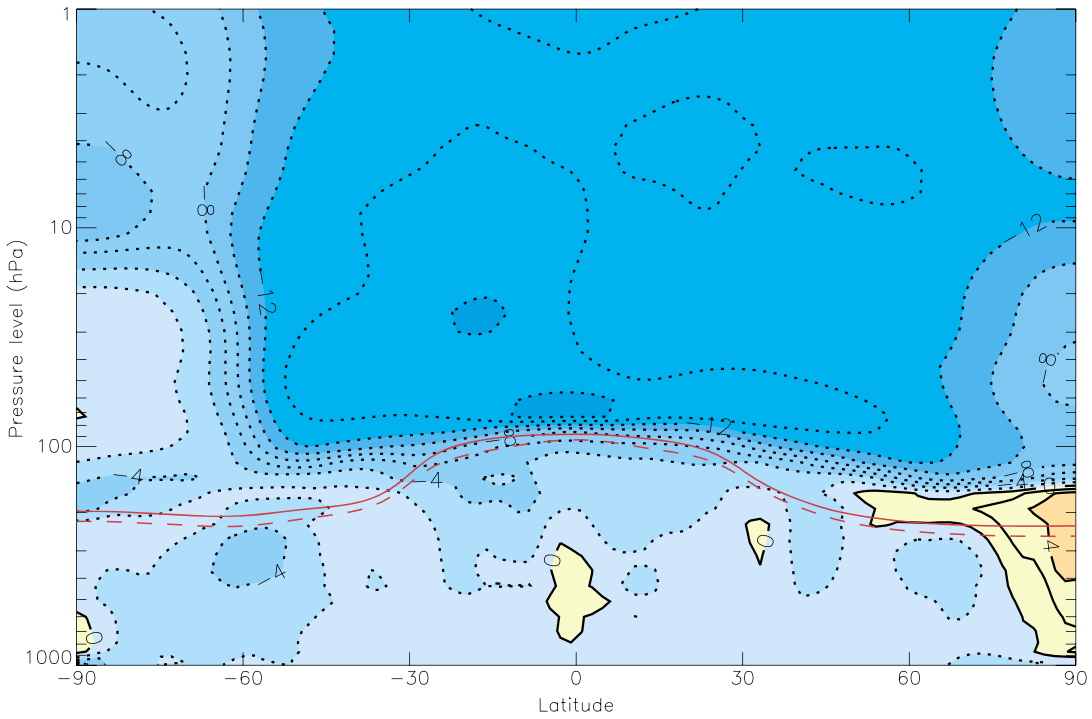

Figure 4. Percentage difference relative to the WACCM PI values in the zonal mean annual mean water vapor response to an abrupt 4 times increase in CO_2 between simulations with WACCM and SC-WACCM. Solid red line is $4 \times CO_2$ tropopause level and dashed red line PI tropopause level.

as we might expect from the larger change in CO_2. This agreement indicates that the chemical reaction schemes and the acceleration of the Brewer-Dobson circulation are similar in these models.

Given this agreement, why then the strikingly different negative chemistry feedbacks between these models? First, the amount of radiative forcing calculated from the ozone change is almost an order of magnitude less in this study than that of *Nowack et al.* [2014], who report −0.68 W m^{-2}. Using a $4 \times CO_2$ total ozone feedback of −0.015 W m^{-2} K^{-1} estimated by *Dietmüller et al.* [2014], along with their coupled-chemistry model's ΔT of 7.52 K, we calculate an ozone RF of −0.11 W m^{-2}. This value is much closer to our off-line calculation using ozone from the WACCM model (−0.08 W m^{-2}), but the EMAC value still represents a 38% stronger negative feedback than WACCM. Considering the apparently good agreement in the ozone differences, it is perplexing to see such a range in the strengths of the ozone feedbacks across the three models.

Second, there are large differences across the models in the water vapor responses to increased CO_2 with and without interactive chemistry. *Nowack et al.* [2014] differenced the water vapor distributions during the latter part of their model simulations and found greater than 30% differences in the tropical lower stratosphere, with 10 to 20% decreases over most of the middle to upper troposphere (see their supplemental Figure 3). For the case of CO_2 doubling, *Dietmüller et al.* [2014] predicts 10–15% differences in the stratosphere, and up to 3% in the troposphere. The differences in the water vapor distributions, averaged over the last 50 years of the two $4 \times CO_2$ WACCM simulations, are shown in Figure 4. In the lower tropical stratosphere, water vapor is 10 to 14% smaller in the simulation with interactive chemistry. This is likely due to cooler tropopause temperatures (shown in Figure S1 in the supporting information) leading to stronger dehydration of the air entering the stratosphere in the tropics. The amount of cooling below the tropopause is less in our model than shown by *Nowack et al.* [2014] and is the likely cause of the smaller differences in WACCM water vapor compared to their model. To put the WACCM differences into perspective, in the lowermost stratosphere (~85 hPa), the change in WACCM equatorial water vapor from PI to $4 \times CO_2$ is 1.52 ppmv, while in SC-WACCM it is 1.79 ppmv (i.e., the inclusion of chemistry reduces the water vapor increase by only 0.27 ppmv). The globally averaged forcing from the stratospheric water vapor difference was estimated to be −0.78 W m^{-2} by *Nowack et al.* [2014] and −0.32 W m^{-2} by *Dietmüller et al.* [2014], both of which are larger than their calculated ozone change forcing. Again using PORT, we calculate the net adjusted forcing from stratospheric water vapor differences

to be −0.10 W m^{-2}, with a small shortwave warming of 0.03 W m^{-2} partially canceling a longwave cooling of −0.13 W m^{-2}.

For most of the troposphere the changes in WACCM water vapor are less than 2%. Using radiative kernels, *Shell et al.* [2008] showed that TOA longwave radiative flux is most sensitive to water vapor in the tropical upper troposphere (~250 hPa, see their Figure 1a). Changes in this region in WACCM are drastically smaller than those reported by *Nowack et al.* [2014], and even smaller than from *Dietmüller et al.* [2014] (calculated for a factor of 3 smaller change CO_2). These changes are all broadly consistent with the tropospheric water vapor response to surface temperature change (e.g., with a water vapor integrated column response of 7.5% K^{-1} [see *Held and Soden*, 2006]). In the case of WACCM, the less than 2% water vapor differences are expected, since the changes in global surface temperature are almost identical. We note that these small differences in water vapor between WACCM and SC-WACCM are dwarfed by the tropospheric water vapor changes in WACCM between the PI state and $4 \times CO_2$ state; the general warming of the troposphere leads to a water vapor increases of over 100% in the upper tropical troposphere (shown in Figure S2). Hence, it is not surprising then that the water vapor differences between the run with and without chemistry do not lead to a significantly different ECS.

5. Summary and Conclusions

We have conducted abrupt $4 \times CO_2$ increase experiments in the CESM, using WACCM and SC-WACCM as the atmospheric model component, in order to investigate the effects of including interactive chemistry within an ESM on equilibrium climate sensitivity (ECS). We found that the distribution of ozone is significantly affected by the larger CO_2 values, with strong increases in the upper stratosphere and decreases in the lower stratosphere. A cooling of the UTLS leads to a drying of the stratosphere, but changes in tropospheric water vapor are relatively small. Using the methodology of *Gregory* [2004], we estimate that the magnitude of the chemistry feedback effect on the RF, ECS, and net feedback is on the order of 1%. Off-line radiative transfer calculations were performed to calculate the change in forcing expected from the perturbations in ozone and water vapor that result from including interactive chemistry under $4 \times CO_2$. The changes in RF were similarly found to be small, consistent with a weak chemistry feedback effect on ECS.

In comparison to studies of the chemistry feedback effects in other ESMs, the CESM chemistry feedback is significantly lower than previously reported (e.g., up to a 20% reduction in ECS in *Nowack et al.* [2014]). While the changes in ozone from the abrupt increase in CO_2 appear to be consistent across the various models, the subsequent RF and water vapor changes and its forcing can be radically different. Understanding why these models respond so differently to ozone changes will be critical in the evaluation of model spread in simulations used to estimate the response of our climate to increasing greenhouse gases. In particular, the differences will need to be considered when interpreting the results of the proposed DECK (Diagnostic, Evaluation and Characterization of Klima) experiments conducted as part of the next Coupled Model Intercomparison Project (CMIP) [*Eyring et al.*, 2015] that include both 1% yr^{-1} until doubling and abrupt $4 \times CO_2$ increase simulations.

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