

## State Dependence of CO<sub>2</sub> Effective Radiative Forcing from 1/16× to 16×CO<sub>2</sub>

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**ABSTRACT:** Most research on the state dependence of climate sensitivity has focused on radiative feedbacks, with less attention given to radiative forcing. However, recent studies show that the carbon dioxide (CO<sub>2</sub>) radiative forcing depends not only on the CO<sub>2</sub> concentration but also on the base state, particularly the stratospheric temperature profile. Hence, we here carry out atmosphere-only experiments with prescribed sea surface temperatures using Community Earth System Model, version 1, Large Ensemble (CESM1-LE), broadband radiative transfer calculations, and a one-dimensional radiative-convective equilibrium model to thoroughly investigate the dependence of effective radiative forcing (ERF) on varying levels of CO<sub>2</sub> forcing and base-state stratospheric temperatures from 1/16× to 16×CO<sub>2</sub>. Using both the CESM1-LE and a radiative-convective equilibrium model, we demonstrate that ERF strongly depends on the CO<sub>2</sub> value of the underlying base state, deviating significantly from a simple logarithmic relationship with CO<sub>2</sub> concentration. Specifically, doubling CO<sub>2</sub> from a base state of 8×CO<sub>2</sub> results in an ERF value that is 50% higher than doubling CO<sub>2</sub> from a 1/16×CO<sub>2</sub> state. By decomposing ERF into instantaneous radiative forcing (IRF) and radiative adjustments, we show that the IRF is largely responsible for the state dependence of ERF. We attribute the increase in IRF with CO<sub>2</sub> concentrations to the stratospheric cooling at 10 hPa. Furthermore, we find that the radiative adjustments are not constant with each CO<sub>2</sub> doubling and halving, and their magnitude depends on the method used to compute them, be it via radiative kernels or via offline radiative transfer calculations. A significant implication of our findings is that the state dependence of ERF needs to be taken into account when studying climate sensitivity under large CO<sub>2</sub> perturbations within the feedback-forcing framework.

**KEYWORDS:** Atmosphere; Forcing; Climate; Greenhouse gases; Radiative forcing

### 1. Introduction

Estimating equilibrium climate sensitivity (ECS) is a crucial component in understanding the potential impacts of climate change and interpreting the mechanisms of past climate changes. ECS is the global mean surface warming after doubling carbon dioxide (CO<sub>2</sub>) concentration from preindustrial (PI) conditions. ECS has been approximated with effective climate sensitivity (EffCS), the surface warming calculated from 150-yr abrupt 4×CO<sub>2</sub> runs and divided by 2, assuming a logarithmic dependence on the radiative forcing with each doubling of CO<sub>2</sub>. EffCS is directly proportional to the radiative forcing imposed on the system and inversely proportional to the feedback parameter  $\lambda$  (EffCS =  $F/\lambda$ ).

Efforts to determine EffCS have involved various approaches, including the examination of paleoclimatic evidence (Anagnostou et al. 2016, 2020; Farnsworth et al. 2019; Friedrich et al. 2016; Shaffer et al. 2016; Zhu et al. 2019) and modeling studies (Hansen et al. 2005; Bloch-Johnson et al. 2021; Meraner et al. 2013; Mauritzen et al. 2019; Sherwood et al. 2020; Mitevski et al. 2021; Zhu and Poulsen 2020). However, a major assumption in estimating EffCS is that it remains constant under any CO<sub>2</sub> doubling, neglecting the potential for variation due to

nonlinear temperature dependence of radiative feedbacks caused by changes in base state and/or CO<sub>2</sub> perturbations (Bloch-Johnson et al. 2021; Sherwood et al. 2015). Additionally, small contributions from nonlogarithmic CO<sub>2</sub>-dependent radiative forcing have been reported (Mitevski et al. 2022).

A recent study by He et al. (2023) has provided new insights into the state dependence of ECS, highlighting not only the influence of feedbacks but also the role of the radiative forcing  $F$ . The study revealed that the instantaneous radiative forcing (IRF), which constitutes the bulk of  $F$ , strongly depends on the climatological base state. Specifically, the IRF increases when computed from a higher CO<sub>2</sub> base state, and this increase is primarily related to the colder stratospheric temperatures associated with higher CO<sub>2</sub> concentrations. These findings align with earlier analytical work (Jeevanjee et al. 2021), emphasizing the importance of stratospheric-surface temperature difference in determining CO<sub>2</sub> IRF. Furthermore, the stratospheric-surface temperature difference was previously shown to explain the spatial and temporal inhomogeneity of the IRF from CO<sub>2</sub> in an Earth system modeling study, despite the CO<sub>2</sub> concentration being uniformly distributed (Huang et al. 2016; Chen et al. 2023).

Most of the previous work on the state dependence of the CO<sub>2</sub> radiative forcing has primarily centered on the IRF (Jeevanjee et al. 2021; He et al. 2023; Pincus et al. 2020), with less attention given to the state dependence of adjustments.

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Previous studies have emphasized the IRF due to it being better understood (Mlynczak et al. 2016) and significant contribution (around two-thirds) to the effective radiative forcing (ERF), where ERF is the sum of the IRF and the radiative adjustments. Additionally, the IRF has been shown to be the primary driver of the adjustments (Sherwood et al. 2015; Forster et al. 2016; Ramaswamy et al. 2018; Hansen et al. 2005; Smith et al. 2018, 2020a). However, ERF is the most widely adopted definition of radiative forcing (Forster et al. 2016; Ramaswamy et al. 2018; Sherwood et al. 2015) because it has been found to be the most comprehensive measure of its impact on the surface climate (Hansen et al. 2005; Richardson et al. 2019). Hence, the state dependence of ERF in response to CO<sub>2</sub> forcing bears further examination.

Our previous study (Mitevski et al. 2022) demonstrated that ERF exhibits asymmetry across CO<sub>2</sub> doublings and halvings, leading to asymmetric surface temperature responses. Specifically, we found that the ERF increases slightly more than the logarithm of CO<sub>2</sub> concentration, and hence, the temperature response in the CO<sub>2</sub> doubling cases (up to 8×CO<sub>2</sub>) is stronger than the response in the corresponding CO<sub>2</sub> halvings (down to 1/8×CO<sub>2</sub>). While other ECS studies have also demonstrated a higher ERF increase than the expected logarithmic dependence (Hansen et al. 2005; Colman and McAvaney 2009; Caballero and Huber 2013), the underlying mechanisms behind this ERF increase, and its decomposition into IRF and adjustments, have yet to be thoroughly explored. To understand this, we here conduct simulations using the Community Earth System Model, version 1, Large Ensemble (CESM1-LE), model, calculating ERF from atmosphere-only experiments with prescribed sea surface temperatures and sea ice starting from different CO<sub>2</sub> base states. Additionally, we carry out offline radiative transfer calculations and use a one-dimensional radiative-convective equilibrium model to calculate IRF. By separately quantifying the IRF and the adjustments, we extend previous work that studied only the state dependence of IRF (He et al. 2023) and elucidate the mechanisms behind the state dependence of ERF. Specifically, we investigate whether the stratospheric temperature response due to a change in CO<sub>2</sub> causes ERF to increase more than the logarithm of the CO<sub>2</sub> concentration and whether the adjustments change as the logarithm of the CO<sub>2</sub> concentration.

## 2. Methods

### a. Models

We utilize the original CESM1-LE (Kay et al. 2015). This model consists of the Community Atmosphere Model, version 5 (CAM5), and the Parallel Ocean Program, version 2 (POP2). The model components have a horizontal resolution of approximately 1°, with 30 vertical levels in the atmospheric model and a model top of 3.6 hPa, and with 60 vertical levels in the ocean model. Additionally, we incorporate some results from the GISS-E2.1-G model (Miller et al. 2021; Kelley et al. 2020). To estimate the IRF from the CESM1-LE runs, we use radiative kernels (Soden et al. 2008) and the radiative transfer model suite of community radiative transfer codes based on Edwards

and Slingo (SOCRATES) on monthly model output (Edwards and Slingo 1996; Manners 2015).

To isolate the role of the radiative-convective response in the ERF increase with CO<sub>2</sub>, we use a one-dimensional radiative-convective equilibrium model—Konrad (Dacie et al. 2019; Kluft et al. 2019). We run the default configuration of Konrad, which comes with clear skies, fixed relative humidity, fixed ozone, and a saturated isentropic lapse rate in the troposphere. We run Konrad with 100 vertical levels from 1000 to 0.1 hPa. The reference CO<sub>2</sub> concentration is set to 284.7 ppm, the same as in the CESM1-LE experiments. We run Konrad with time steps of 24 h for 150 days. Note that Konrad uses the same radiation code as CESM1-LE, Rapid Radiative Transfer Model for general circulation models (GCMs) (Iacono et al. 2008), to calculate radiative fluxes.

It is important to note that SOCRATES does not use RRTMG for its radiation calculations. Previous work (Pincus et al. 2020) has investigated the differences between SOCRATES and RRTMG and found them to be small. Nonetheless, the use of SOCRATES here contributes some small differences in radiative forcing calculations, compared to CESM1-LE and Konrad, but these do not affect our results or conclusions.

### b. Experiments

We conduct 20-yr-long abrupt forcing experiments using the CESM1-LE model with prescribed sea surface temperature and sea ice to preindustrial values, referred to as “prescribed-SST” runs, to examine the effects of stratospheric temperature on the ERF in abrupt CO<sub>2</sub> doublings and halvings. For the doubling experiments, we start from PI conditions and, every 20 years, abruptly double the CO<sub>2</sub> concentration up to 16×CO<sub>2</sub> (indicated in red in Fig. 1a). We chose to do the step experiments because the stratospheric temperature adjusts (cools) at each doubling step, allowing us to elucidate its impact on the ERF more cleanly. The halving experiments follow a similar approach; starting from PI conditions, we decrease the CO<sub>2</sub> concentration by half every twenty years. The stratospheric temperature adjusts (warms) at each step, and we continue the steps until 1/16×CO<sub>2</sub> (blue in Fig. 1a). Similarly, with Konrad, we first equilibrate to a reference climate, and then, keeping the surface temperature fixed and allowing the atmospheric temperature to adjust, we perturb the CO<sub>2</sub> concentration from 1/16× to 16×CO<sub>2</sub>.

### c. ERF

The ERF in the CESM1-LE runs is calculated as the global mean net top-of-the-atmosphere (TOA) radiation between two adjacent CO<sub>2</sub> steps, incorporating both stratospheric and tropospheric adjustments (Sherwood et al. 2015). While the SSTs and sea ice concentrations (SICs) are fixed at PI values, the land temperature in CESM1-LE responds to the radiative forcing, as prescribing fixed land surface conditions in a climate model presents technical challenges (Andrews et al. 2021; Hansen et al. 2005; Ackerley et al. 2018). The land warms and cools with increasing and decreasing CO<sub>2</sub> levels, respectively. This land surface temperature response introduces a radiative effect and some adjustments, which we separately quantify as

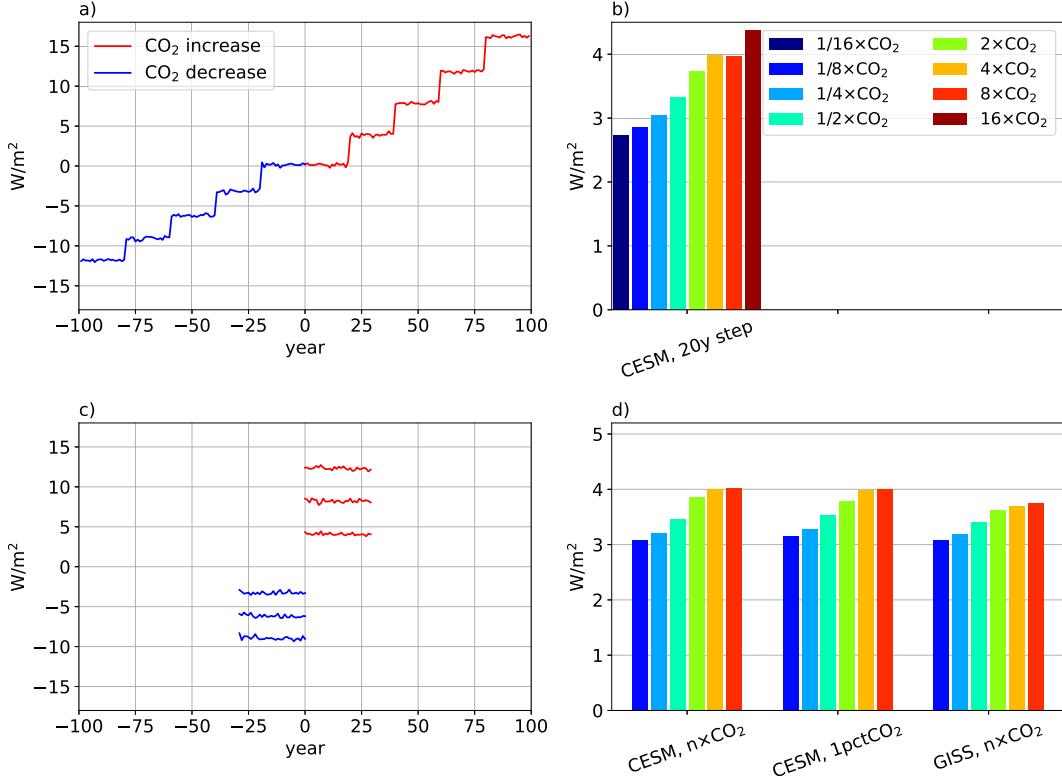


FIG. 1. (a) The radiative imbalance at the TOA in the 20-yr step experiments with CESM1-LE, where  $\text{CO}_2$  is doubled (red) or halved (blue) starting from the PI condition (years from -20 to 20). (b) The ERF, calculated as the difference between the two levels shown in (a). (c) The radiative imbalance at the TOA in the  $n \times \text{CO}_2$  prescribed-SST runs, starting from the PI  $\text{CO}_2$  concentration [distinct from the step experiments in (a) and (b), previously documented in Mitevski et al. (2022)]. (d) Normalized ERF (divided by  $\log_2 n$ ) obtained from the (left) abrupt  $n \times \text{CO}_2$  run with CESM1-LE, (center) 1pctCO<sub>2</sub> runs with CESM1-LE, and (right) abrupt  $n \times \text{CO}_2$  runs with the GISS-E2.1-G model, with adjustments made for land warming, previously documented in Mitevski et al. (2022).

surface temperature adjustment  $A_{T_s}$  by multiplying the surface temperature radiative kernel with the surface temperature. Since the surface temperature is fixed in Konrad, we simply compute the ERF as the difference in net TOA radiation between each successive  $\text{CO}_2$  doubling/halving run.

#### d. IRF and adjustments

To calculate the IRF, we employ two different approaches. First, using the double call method, we estimate the IRF using offline radiative transfer calculations with the SOCRATES broadband model (Edwards and Slingo 1996; Manners 2015). In the double call method, we first compute the net TOA radiative flux using SOCRATES with the initial atmospheric state. We then repeat the computation with a perturbed  $\text{CO}_2$  concentration (e.g., doubled) and find the IRF as the difference in the net TOA radiative flux. We do these calculations twice, once as a forward calculation with the unperturbed atmospheric state from each  $\text{CO}_2$  level and doubled the  $\text{CO}_2$  concentration and second as a backward calculation with the perturbed atmospheric state for each  $\text{CO}_2$  level and keeping the  $\text{CO}_2$  concentration the same. We then take the average of both forward and backward calculations to find the IRF.

Nonetheless, the forward and backward calculations yield similar results.

Second, we estimate IRF as a residual by subtracting the rapid adjustments from the ERF. Rapid adjustments occur on a much shorter time scale than the radiative feedbacks and are decoupled from the surface response. Following the approach of Soden et al. (2008), we express the ERF resulting from a climate perturbation as a sum of IRF and several rapid adjustments:

$$\text{ERF} = \text{IRF} + A_T + A_{T_s} + A_q + A_\alpha + A_c, \quad (1)$$

where  $A_x$  are the rapid adjustments,  $x$  is the atmospheric temperature  $T$ ,  $T_s$  is the surface temperature,  $q$  is the specific humidity,  $\alpha$  is the surface albedo, and  $c$  represents the clouds. We further separate the rapid adjustments into stratospheric and tropospheric contributions using a tropopause that varies linearly from 100 hPa at the equator to 300 hPa at the poles. The radiative kernel  $K_x$  approximates the change in TOA shortwave or longwave radiation  $\Delta R$  resulting from a unit change in the state variable  $\Delta x$ , such that  $K_x \approx \partial R / \partial x$  and  $A_x = K_x \Delta x$ . The cloud kernels  $A_c$  are determined following the method of Soden et al. (2008) by computing the difference

between all-sky kernel decomposition and clear-sky kernel decomposition. The radiative kernels are calculated by making small perturbations in the model's base climatology and running the resulting atmosphere through an offline radiative transfer code. TOA fluxes were then compared between the perturbed and base states. Since stratospheric adjustments have been shown to depend on the vertical resolution of the kernel (Smith et al. 2020b), we perform adjustment calculations using three different kernels that have the highest pressure level of 1 hPa or more from the following models: HadGEM3-GA7.1 (Smith et al. 2020a), ECMWF-RRTM (Huang et al. 2017), and ECHAM6 (Block and Mauritsen 2013). The results in all figures represent the mean of these three kernels. While the kernel method offers advantages for computing adjustments—such as eliminating the need to run offline radiative transfer models and enabling easier decomposition of individual adjustments—it also introduces uncertainties. These include the choice of the kernel base state, the kernel model top, and, most importantly, the linearity of the kernel, particularly when exploring adjustments at very high or low CO<sub>2</sub> concentrations. A more detailed discussion of these uncertainties, along with a comparison of adjustment computations using kernels versus offline calculations with SOCrates, is provided later in the text.

We calculate the stratospheric adjustment with SOCrates by doing two offline calculations with the same CO<sub>2</sub> concentrations but a different atmospheric state (e.g., temperature profile). In the Konrad simulations, we calculate the adjustments simply as a residual between the ERF and the IRF.

### 3. Results

#### a. Nonlogarithmic ERF increase with CO<sub>2</sub> concentrations

We start with the step experiments with CESM1-LE (Fig. 1a) where CO<sub>2</sub> is increased from PI to 16×CO<sub>2</sub> values in 20-yr steps (red) and similarly decreased to 1/16×CO<sub>2</sub> (blue). We calculate the resulting ERF as the difference between two consecutive CO<sub>2</sub> steps. While a purely logarithmic relationship would imply equal ERF for each step, we observe that the absolute value of the “blue step” at year −80 is considerably smaller than the absolute value of the “red step” at year 80. Figure 1b displays the calculated ERF from each step, revealing that the ERF does not remain constant but increases more than the expected logarithmic dependence of the CO<sub>2</sub> concentration, with the ERF from CO<sub>2</sub> doubling rising from 2.8 W m<sup>−2</sup> at 1/16×CO<sub>2</sub> to 4.5 W m<sup>−2</sup> at 8×CO<sub>2</sub>.

In our previous work (Mitevski et al. 2021, 2022), we calculated the ERF from abrupt  $n \times \text{CO}_2$  forcing (from 1/8× to 8×CO<sub>2</sub>) as the net TOA radiation between the  $n \times \text{CO}_2$  states and PI, which we reproduce those results here in Fig. 1c, for the sake of completeness. The ERF calculated this way with the same CESM1-LE model (Fig. 1d) exhibits an increase larger than the logarithmic relationship with CO<sub>2</sub> concentration. Specifically, the ERF increases from 3 W m<sup>−2</sup> at 1/8×CO<sub>2</sub> to 4 W m<sup>−2</sup> at 8×CO<sub>2</sub>. Note that these experiments differ from the 20-yr step experiments in Figs. 1a and 1b in that they always start from the PI atmospheric state and yet yield similar ERF increases with CO<sub>2</sub>. Furthermore, we obtain similar results with

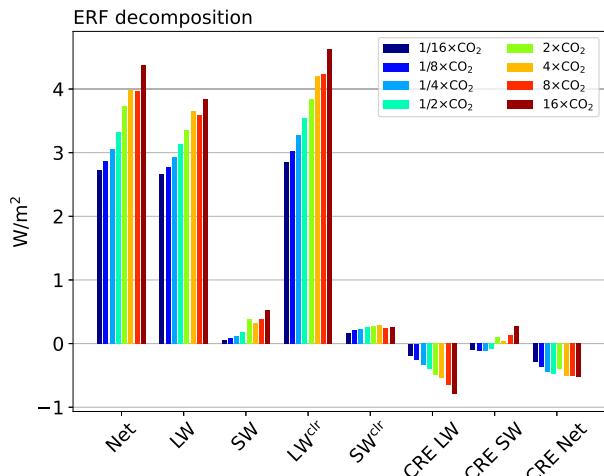


FIG. 2. Net TOA radiation for the runs in Figs. 1a and 1b decomposed into LW, SW, all sky, clear sky (clr), and CREs. CRE is the difference between all sky and clear sky.

increasing and decreasing CO<sub>2</sub> by 1% yr<sup>−1</sup> (1pctCO<sub>2</sub> runs) with CESM1-LE and abrupt  $n \times \text{CO}_2$  runs with the GISS model (Fig. 1d). These ERF increases exceed both the logarithmic relationship and the simplified expressions proposed in the literature (Byrne and Goldblatt 2014; Etminan et al. 2016) (see Fig. 2 in Mitevski et al. 2022). The similarity between the abrupt  $n \times \text{CO}_2$ , the 20-yr abrupt step, and 1% yr<sup>−1</sup> CO<sub>2</sub> forcing experiments demonstrates that the ERF increase with CO<sub>2</sub> concentration is independent of the method used to perturb CO<sub>2</sub> in prescribed-SST runs with a general circulation model. Furthermore, the GISS-E2.1-G model results confirm that the ERF increase is not limited to a particular model—we hypothesize that the result will be found in other GCMs. Last, our findings indicate that the ERF increases with CO<sub>2</sub> irrespective of whether we start from the PI or an  $n \times \text{CO}_2$  atmospheric state.

Next, we decompose the net TOA radiation imbalance (which, by definition, is the ERF) in the “step” experiments from Fig. 1b into longwave (LW) and shortwave (SW) components, as well as the clear sky (clr), all sky, and cloud radiative effects (CREs) in Fig. 2. As one would expect, most of the increase in net TOA radiation with CO<sub>2</sub> comes from the all-sky LW component (second set of bars), with a smaller contribution from the SW all-sky component. Further decomposition of the LW component shows that most of the increase with CO<sub>2</sub> comes from the clear-sky LW, with the CRE LW opposing the rise. We find that the net CRE (CRE Net) has a relatively small impact on the overall ERF increase, and it actually opposes the ERF increase.

More importantly, we find that the nonlogarithmic ERF increase with CO<sub>2</sub> concentration coincides with an increase in the IRF (Fig. 3). First, we compute the IRF (Fig. 3a) as the residual between the ERF and the total adjustments, which in turn are calculated with radiative kernels (see methods). Our analysis reveals that the IRF, calculated as a residual, increases with CO<sub>2</sub>, particularly showing a pronounced increase between

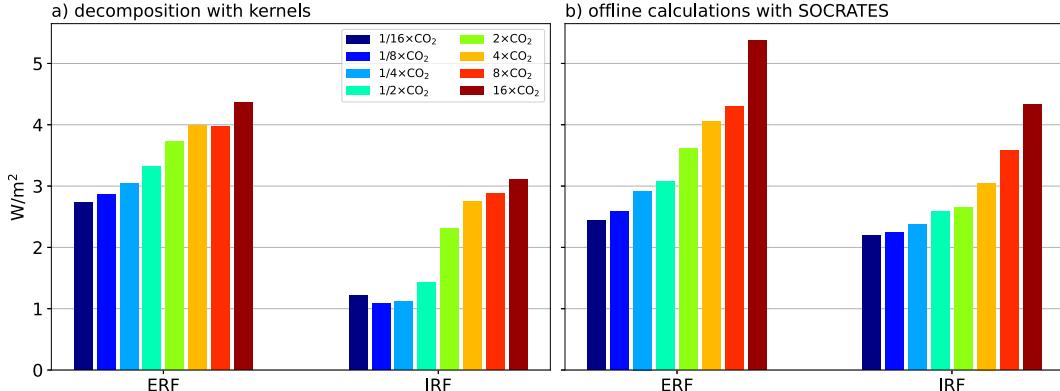


FIG. 3. ERF and IRF with (a) kernel decomposition and (b) offline calculations with SOCrates.

1/2 $\times$  and 2 $\times$ CO<sub>2</sub>. In addition, we carry out offline radiative transfer calculations with SOCrates using the same atmospheric profiles from the 20-yr step experiments to compute both ERF and IRF (Fig. 3b). These radiative transfer calculations confirm our findings with the CESM1 prescribed-SST runs and clearly show a similar monotonic increase from 1/16 $\times$ CO<sub>2</sub> to 16 $\times$ CO<sub>2</sub>. The ERF increase here also corresponds with the IRF's increase, although the IRF has a more gradual increase from 1/16 $\times$ CO<sub>2</sub> to 16 $\times$ CO<sub>2</sub> with no step-like behavior between 1/2 $\times$  and 2 $\times$ CO<sub>2</sub> like the IRF calculated as a residual in Fig. 3a. In the next section, we investigate the IRF discrepancy between both calculations.

The IRF's state dependence, specifically its increase with CO<sub>2</sub>, has been investigated in prior studies (Huang et al. 2016; Jeevanjee et al. 2021; He et al. 2023; Chen et al. 2024), which have demonstrated that the emission temperatures within the troposphere and stratosphere can account for the IRF's state dependence. Specifically, when the surface temperature is fixed, a decrease in stratospheric temperature is anticipated to correspond with an increase in IRF. In our experimental setup with CESM1-LE, we vary CO<sub>2</sub> concentrations from 1/16 $\times$  to 16 $\times$ CO<sub>2</sub>, consequently altering the stratospheric temperature near CO<sub>2</sub> emissions (near 10 hPa,  $T_{10\text{hPa}}$ ) from 260 to 205 K, while the near-surface temperature responds very little

compared to  $T_{10\text{hPa}}$  (see Fig. 4a). It is well established in the literature that the addition of CO<sub>2</sub> leads to surface and tropospheric warming but induces stratospheric cooling (Manabe and Wetherald 1967; Ramaswamy et al. 2006). The opposite occurs when CO<sub>2</sub> is removed: The surface and troposphere cool, while the stratosphere warms. Hence, we analyze the relationship between  $T_{10\text{hPa}}$  and ERF, and consistent with the previous literature, we find a strong negative correlation ( $R^2 = 0.99$ ) in Fig. 4b. As the stratospheric temperatures decrease, ERF increases. Moreover, whether the IRF is computed directly with SOCRA-TES or indirectly as a residual, its correlations with  $T_{10\text{hPa}}$ ,  $R^2 = 0.81$ , and  $R^2 = 0.95$ , are very high. Therefore, we conclude that the IRF increase with CO<sub>2</sub> associated with stratospheric cooling near 10 hPa largely explains the observed rise in ERF. This direct link between stratospheric temperature states and the increase in ERF largely explains the state dependence of ERF on CO<sub>2</sub> concentration.

#### b. Nonlogarithmic CO<sub>2</sub> dependence of radiative adjustments

To gain further insight into the difference in IRF values when calculated from SOCrates or from the kernels shown in Fig. 3, we now consider the radiative adjustments in Fig. 5. The total of the adjustments calculated from the kernels  $\sum A_i$

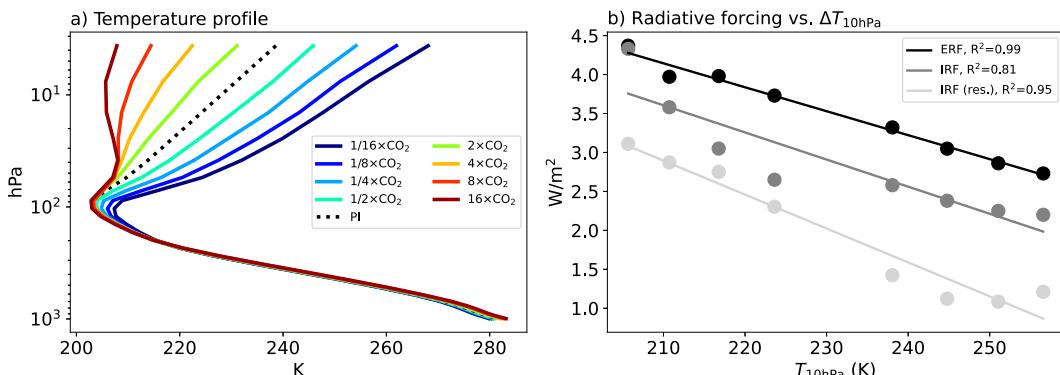


FIG. 4. (a) Global mean atmospheric temperature profiles for the 20-yr step experiments from Fig. 1a. (b) ERF, IRF calculated from SOCrates (IRF), and IRF calculated as a residual from the kernel adjustment calculations (IRF (res.)) on the y axis correlated with temperature response at 10 hPa on the x axis.

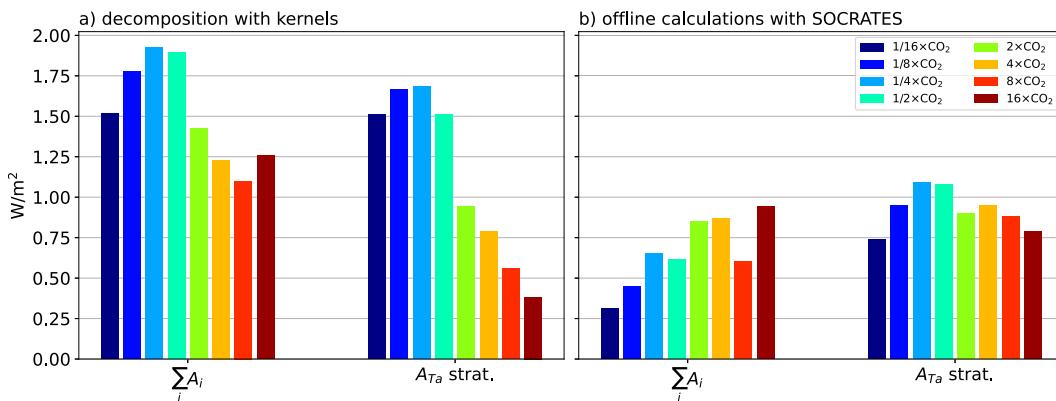


FIG. 5. Total adjustments  $\sum_i A_i$  and stratospheric temperature adjustments  $A_{T_a}$  strat. calculated with (a) kernels and (b) offline radiative transfer calculations with SOCRAVES.

(Fig. 5a) is found to be much larger in the cases with decreasing CO<sub>2</sub> levels compared to those with increasing CO<sub>2</sub> levels, resulting in an overall decreasing but nonmonotonic dependence with CO<sub>2</sub> concentration. This decrease with CO<sub>2</sub> concentration, which is largest between 1/2× and 2×CO<sub>2</sub>, explains why the IRF calculated as the residual increases the most between 1/2× and 2×CO<sub>2</sub>. Additionally, we compute the adjustments using offline calculations with SOCRAVES (Fig. 5b; also see methods) and find that the total adjustments increase nonmonotonically with CO<sub>2</sub>, which is opposite to what we find with the kernel calculations. Although both SOCRAVES and the kernel method show nonconstant adjustments, they exhibit opposite overall changes from 1/16× to 16×CO<sub>2</sub>. Therefore, caution is needed when studying the adjustments across multiple CO<sub>2</sub> doublings and halvings using the kernel method.

An important caveat regarding the kernel calculations is that kernels are derived for small perturbations in warming scenarios and do not account for any state dependency, as reported by previous studies (Jonko et al. 2012), which likely has an impact on the wide CO<sub>2</sub> range we explore here. To check for any state dependency in the kernel calculations, we repeat the adjustment calculations with offline radiative transfer calculations with SOCRAVES. The SOCRAVES calculations exhibit some similarities with the kernel calculations (Fig. 5): Total adjustments increase between 1/16× and 1/2×CO<sub>2</sub> and decrease between 2× and 8×CO<sub>2</sub>, while showing an increase at 16×CO<sub>2</sub>. The change in stratospheric adjustment largely follows this pattern from 1/16× to 1/2×CO<sub>2</sub>. The largest difference between kernels and SOCRAVES calculations occurs between 1/2× and 2×CO<sub>2</sub>, where there is a large decrease of almost 0.5 W m<sup>-2</sup> in the kernel calculations, likely attributable to state dependence. Further exploration of the state dependence of kernels, particularly in CO<sub>2</sub> decrease scenarios, is clearly needed but lies beyond the scope of our work here.

Surprisingly, we find that the stratospheric adjustment is not constant with each CO<sub>2</sub> doubling/halving perturbation calculated with both kernels and SOCRAVES, with the strongest response when calculated with kernels (Fig. 5a). Since the stratospheric temperature adjustment is simply the spatial and vertically integrated product between the stratospheric temperature response

and the radiative kernel, we can further clarify whether the temperature response or the radiative kernel itself causes the stratospheric adjustment to not be constant with each CO<sub>2</sub> doubling/halving. Since the kernels are constant across all the CO<sub>2</sub> perturbations, they do not impact the stratospheric adjustment across CO<sub>2</sub> levels. In fact, we find that the stratospheric temperature response has a stronger response when CO<sub>2</sub> concentration is lower (e.g., 1/4×CO<sub>2</sub>) and monotonically increases with CO<sub>2</sub> up to 16×CO<sub>2</sub> (Fig. 6). Hence, the temperature response explains the asymmetry in the stratospheric adjustment ( $A_{T_a}$  strat. in Fig. 5a): It increases between 1/16× and 1/4×CO<sub>2</sub> and then decreases up to 8×CO<sub>2</sub>.

One possible explanation for the stronger stratospheric temperature response in the CO<sub>2</sub> decrease cases compared to the CO<sub>2</sub> increase cases (Fig. 6) is the anticipated changes in emissivity at different CO<sub>2</sub> levels. Emissivity is directly proportional to the CO<sub>2</sub> concentration (Hartmann 2015), so it is smaller for the CO<sub>2</sub> decrease cases and larger for the CO<sub>2</sub> increase cases up to 16×CO<sub>2</sub>. Hence, owing to the higher emissivity in the CO<sub>2</sub> increase scenarios (assuming a constant radiative flux divergence in the stratosphere with each CO<sub>2</sub> perturbation), a smaller stratospheric cooling is enough to restore radiative equilibrium in the stratosphere. The opposite occurs when lowering CO<sub>2</sub> concentrations, when a larger stratospheric temperature change is needed to reestablish radiative equilibrium. The qualitative dependence of emissivity on CO<sub>2</sub> concentration at least accounts for the sign of the change in the stratospheric temperature adjustment: a stronger temperature response at low CO<sub>2</sub> values to compensate for the low emissivity and a weaker temperature response at high CO<sub>2</sub> levels. However, because stratospheric temperature is itself a state-dependent variable that influences radiative flux divergence, it could potentially offset the impact of emissivity changes. Nonetheless, a more detailed investigation, including a careful examination of the role of the vertical lapse rate and a thorough analysis of the spectral structure, is necessary to fully comprehend the nonconstant adjustments. However, these lie beyond the scope of our work here.

Another possible explanation for why the stratospheric temperature response in the CO<sub>2</sub> decrease cases is stronger

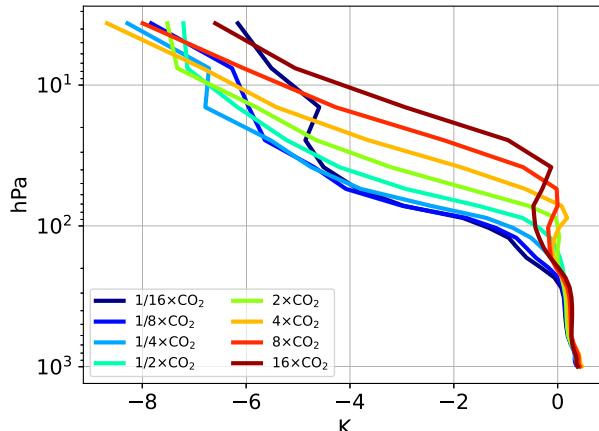


FIG. 6. Stratospheric temperature response between each successive  $\text{CO}_2$  doubling and halving in the 20-yr step experiments from Fig. 1a.

than in the  $\text{CO}_2$  increase cases is the change in the pressure of the emission level. In a previous study by Jeevanjee et al. (2021), the emission pressure level was found to be inversely proportional to the  $\text{CO}_2$  concentration. Therefore, with each  $\text{CO}_2$  doubling, the emission pressure level shifts below 10 hPa, effectively raising the stratospheric temperature profile (Fig. 6), while the stratospheric lapse rate remains unchanged. Similarly, with each  $\text{CO}_2$  halving, the emission pressure level shifts above 10 hPa, thereby lowering the stratospheric temperature profile (Fig. 6). Hence, from the shift in emission pressure level, when integrating over the stratosphere, we can deduce a stronger stratospheric temperature response upon halving than upon doubling  $\text{CO}_2$  (see schematic in Fig. A1 in the appendix).

We further decompose the total adjustments  $\sum_i A_i$  into the individual adjustments (Fig. A2) and find that the largest contribution comes from the stratospheric temperature adjustment ( $A_{T_{\text{strat}}}$ ), while the tropospheric adjustments predominantly exhibit negative values and do not contribute to the overall  $\sum_i A_i$  decrease with  $\text{CO}_2$ . Furthermore, within the stratospheric adjustments, the temperature response is the primary driver, with minimal contributions from stratospheric water vapor in the longwave and shortwave. Regarding the tropospheric adjustments, the largest contribution to the increase in ERF with  $\text{CO}_2$  is the cloud adjustment  $A_c$ ; however, the tropospheric temperature adjustment  $A_{T_{\text{trop}}}$  opposes it (Fig. A2).

#### c. Results with an radiative–convective equilibrium (RCE) model

The stratospheric temperature response to  $\text{CO}_2$  doubling/halving is due to radiative, convective, and large-scale circulation adjustments (Wang and Huang 2020). In the CESM1-LE calculations, sea surface temperatures remain fixed, allowing radiative, convective, and atmospheric circulation to adjust at each  $n \times \text{CO}_2$  level. Given the potential impact of circulation adjustment on the stratospheric temperature, it is crucial to isolate this effect to determine its significance for the stratospheric temperature response. To separate the influence of

stratospheric dynamics on temperature adjustment, we utilize a one-dimensional radiative–convective equilibrium model, Konrad (see methods). Since Konrad lacks any horizontal circulation (and thus any adjustment thereof), it allows us to isolate the radiative–convective response in the increase in ERF with  $\text{CO}_2$ . We find that ERF calculated with Konrad also increases from  $1/16 \times$  to  $16 \times \text{CO}_2$ ; from  $3.2$  to  $5.1 \text{ W m}^{-2}$  (Fig. 7a), further corroborating our previous results with CESM1-LE (Fig. 3). Furthermore, we find that the IRF calculation with Konrad is also the primary driver of the ERF increase, consistent with CESM1-LE. Konrad also produces a similar response in the total adjustments as the SOCRATES calculations from CESM1-LE model output: an increase from  $1/16 \times$  to  $1/2 \times \text{CO}_2$  and a decrease from  $2 \times$  to  $16 \times \text{CO}_2$  ( $\sum_i A_i$  in Fig. 7a). Last, we find that Konrad exhibits the same stratospheric temperature response as the CESM1-LE model: higher response at low  $\text{CO}_2$  values and weaker response at high  $\text{CO}_2$  values (Figs. 7b,c). Hence, the agreement between CESM1-LE and Konrad leads us to conclude that the adjustment of the stratospheric circulation is unlikely to play an important role in determining the ERF dependence on  $\text{CO}_2$ .<sup>1</sup> Moreover, the consistency in the response of the total adjustments between offline calculations with SOCRATES (based on CESM1-LE data) and on the 1D Konrad simulations, in spite of the different broadband radiation codes, indicates that our findings are robust and likely not radiation model dependent.

#### 4. Discussion and conclusions

We have investigated the state dependence of effective radiative forcing (ERF) using prescribed-SST runs with Earth System Model CESM1-LE and the radiative–convective equilibrium model Konrad. We have found that ERF increases by over 50% more than the value expected from the logarithmic dependence on  $\text{CO}_2$  concentration in the range from  $1/16 \times$  to  $16 \times \text{CO}_2$ . We have decomposed the ERF into instantaneous radiative forcing (IRF) and adjustments and attributed the ERF increase primarily to an increase in IRF. This increase in clear-sky IRF, in turn, was linked to the cooling of stratospheric temperatures with increasing  $\text{CO}_2$  concentration. Furthermore, we have found a nonconstant stratospheric temperature adjustment across  $\text{CO}_2$  perturbations, with larger temperature responses at lower  $\text{CO}_2$  values and smaller responses at higher  $\text{CO}_2$  values. We do not currently have a simple and compelling explanation for why the stratospheric temperature response has a larger magnitude at low  $\text{CO}_2$  concentrations, but we suggest that this tendency arises from changes in emissivity and emission pressure as  $\text{CO}_2$  concentration rises from  $1/16 \times$  to  $16 \times \text{CO}_2$ . However, a more rigorous attribution should also consider the spectral variations of the radiative forcing.

It is important to note that our IRF calculations for CESM1-LE using SOCRATES were performed using the

<sup>1</sup> One caveat is worth noting: CESM1-LE is a notoriously low-top model, and its representation of the Brewer–Dobson circulation is probably quite biased. It would thus be valuable to confirm (or possibly invalidate) our conclusion with a high-top model with realistic Brewer–Dobson circulation.

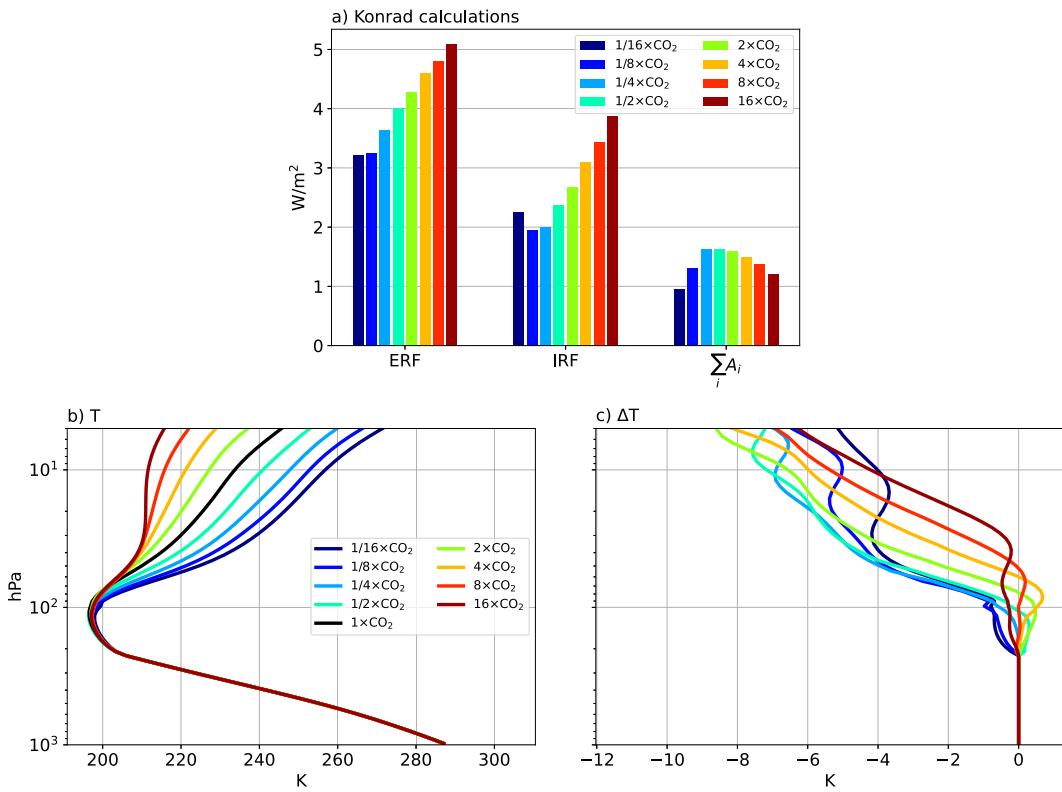


FIG. 7. Results with one-dimensional RCE model Konrad for (a) ERF decomposition into IRF and adjustments, (b) temperature profiles, and (c) temperature response per CO<sub>2</sub> doubling.

base state corresponding to  $n \times \text{CO}_2$ . As a result, the nonlogarithmic behavior of IRF was attributed to changes in the atmospheric state, primarily the stratospheric temperature, in each  $n \times \text{CO}_2$  run, along with the change in CO<sub>2</sub> concentration. By doubling the CO<sub>2</sub> concentration from each  $n \times \text{CO}_2$  base state, we did not specifically explore the extent to which IRF depends solely on the CO<sub>2</sub> forcing. To isolate the importance of the CO<sub>2</sub> concentration from changes in the base state, we additionally calculate IRF with Konrad while maintaining the base state at 1×CO<sub>2</sub> and only perturbing the CO<sub>2</sub> concentration from 1/16× to 16×CO<sub>2</sub>. Comparing this with IRF calculated starting from different  $n \times \text{CO}_2$  states (Fig. 8), we find that IRF remains nearly constant when computed from the same 1×CO<sub>2</sub> base state (the second set of bars in Fig. 8) compared to when computed by doubling CO<sub>2</sub> from  $n \times \text{CO}_2$  state (the first set of bars in Fig. 8). Hence, the majority of the IRF increase stems not from the actual CO<sub>2</sub> concentration but from the base state at which it is computed. The negligible impact of the CO<sub>2</sub> concentration compared to the base state has been noted by other studies (He et al. 2023; Pincus et al. 2020).

One limitation to our adjustment calculations for CESM1-LE (Fig. 6) is the uncertainty arising from the choice of kernel base state, kernel model top, and kernel linearity at very high/low CO<sub>2</sub> perturbations. First, Pincus et al. (2020) demonstrated that radiative transfer calculations across different line-by-line models yield consistent results when performed on an identical base state. Therefore, the base state is the primary

source of uncertainty across different line-by-line models, and this base-state dependence is also critical for kernel calculations. By performing calculations with three different kernels, we attempt to estimate this uncertainty (Fig. A2). Second, the uncertainty across kernels is also caused by the different model tops, as described in Smith et al. (2020b). Variations in stratospheric adjustments derived from kernels can exhibit nearly a twofold difference, as depicted in Fig. 5 of Smith et al. (2020b). Third, the kernel method relies on the assumption of linearity between top-of-the-atmosphere (TOA)

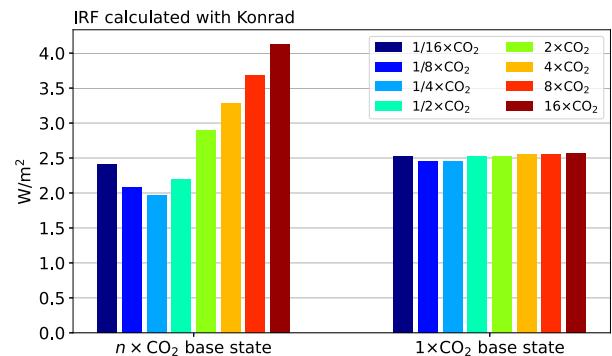


FIG. 8. Normalized clear-sky IRF (divided by  $\log_2 n$ ) calculations using base state of each (left bars)  $n \times \text{CO}_2$  run (same as IRF in Fig. 7a) and (right bars) 1×CO<sub>2</sub> base state.

radiative fluxes and atmospheric variables. However, this linearity assumption is limited to small climate perturbations and may break down under higher forcing scenarios (e.g.,  $16\times\text{CO}_2$ ), as shown by Jonko et al. (2012), particularly when  $\text{CO}_2$  levels are decreasing, as we found between the  $1/2\times$  and  $2\times\text{CO}_2$  in our calculations (Fig. 5a). Hence, these issues ought to be taken into account when utilizing kernels to quantify adjustments.

An additional limitation of our experiments is the absence of fixed land temperatures. Land warming/cooling has the potential to induce feedbacks and alter the atmospheric circulation, which, in turn, can impact cloud adjustment. We can incorporate the surface temperature adjustments calculated from kernels to address this issue. However, we find that the adjustment due to land surface temperatures is roughly constant across the  $\text{CO}_2$  experiments and does not impact the ERF increase ( $A_{T_s}$ , Fig. A2). In addition, our results with CESM1-LE are clearly corroborated by the results from Konrad, which lacks land surface interactions, indicating that our findings are not sensitive to the radiative adjustment due to land temperature response.

Our key finding, that ERF is larger at larger  $\text{CO}_2$  concentrations, underscores the importance of carefully considering radiative forcing when studying the response of the climate system to increases in  $\text{CO}_2$ . Furthermore, our results demonstrate that the radiative adjustments can also vary considerably with different  $\text{CO}_2$  forcings. Thus, when studying the state dependence of  $\text{CO}_2$ 's forcing, it is crucial not only to

calculate the IRF but also to incorporate the adjustments and compute the entire ERF.

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**Data availability statement.** The experiments with CESM1-LE and GISS-E2.1-G models were previously analyzed in Mitevski et al. (2021, 2022, 2023). The CESM1-LE model data can be obtained at <https://doi.org/10.5281/zenodo.5725084>, and the GISS-E2.1-G model data can be obtained at <https://doi.org/10.5281/zenodo.3901624>.

## APPENDIX

Figures A1 and A2 are supplementary figures to the main text.

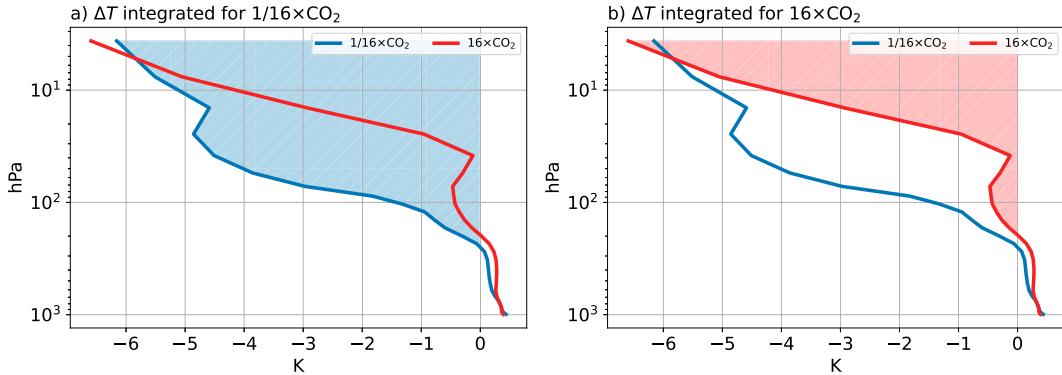


FIG. A1. Schematic of integrating the temperature response profile from (a)  $1/16\times\text{CO}_2$  (blue) and (b)  $16\times\text{CO}_2$  (red). The stratospheric temperature response associated with  $\text{CO}_2$  halving between  $1/16\times$  and  $1/8\times\text{CO}_2$  is shown in the blue line, and  $\text{CO}_2$  doubling between  $8\times$  and  $16\times\text{CO}_2$  in the red line from the 20-yr step experiments (same as in Fig. 6).

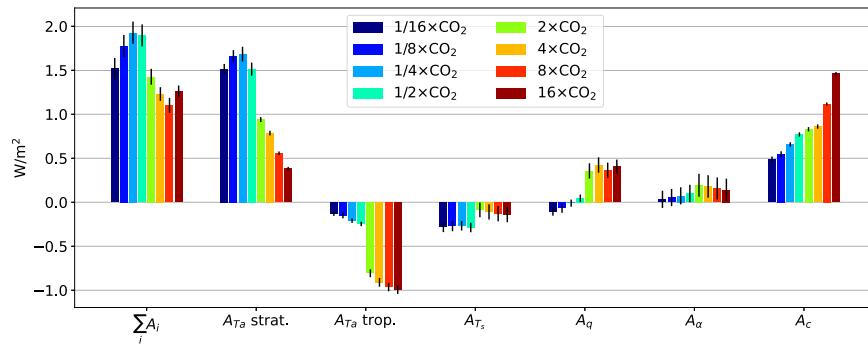


FIG. A2. Total radiative adjustments  $\sum_i A_i$  decomposed into stratospheric temperature ( $A_{T_a}$  strat.), tropospheric temperature ( $A_{T_a}$  trop.), surface temperature ( $A_{T_s}$ ), water vapor ( $A_q$ ), albedo ( $A_a$ ), and cloud ( $A_c$ ) adjustments. The vertical lines show one standard deviation of the three kernels, and the bars show the mean. See the methods section for details on how the calculations are done.

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