Carbon cycle and climate feedbacks under CO₂ and non-CO₂ overshoot pathways

Irina Melnikova^{1,2}, Philippe Ciais², Katsumasa Tanaka^{1,2}, Hideo Shiogama¹, Kaoru Tachiiri^{1,3}, Tokuta Yokohata¹ and Olivier Boucher⁴

- 5 Earth System Division, National Institute for Environmental Studies (NIES), Tsukuba, 305-8506, Japan,
 - ²Laboratoire des Sciences du Climat et de l'Environnement (LSCE), IPSL, CEA/CNRS/UVSQ, Université Paris-Saclay, Gifsur-Yvette, 91191, France
 - ³Research Institute for Global Change, Japan Agency for Marine-Earth Science and Technology, Yokohama, 236-0001, Japan ⁴Institut Pierre-Simon Laplace, Sorbonne Université / CNRS, Paris, 75005, France
- 10 Correspondence to: Irina Melnikova (melnikova.irina@nies.go.jp)

25

Abstract. Reducing emissions of non-CO₂ greenhouse gases, such as methane and nitrous oxide, complements CO₂ mitigation in limiting global warming. However, estimating carbon-climate feedback for these gases remains fraught with uncertainties, especially under overshoot scenarios. This study investigates the impact of CO₂ and non-CO₂ gases with nearly equal levels of effective radiative forcing on the climate and carbon cycle, using the Earth System Model IPSL-CM6A-LR. We first present a method to recalibrate methane and nitrous oxide concentrations to align with published radiative forcings, ensuring accurate model performance. Next, we carry out a series of idealised ramp-up and ramp-down concentration-driven experiments and show that while the impacts of increasing and decreasing CO₂ and non-CO₂ gases on the surface climate are nearly equivalent (when their radiative forcing magnitudes are set to be the same), regional differences emerge. We further explore the carbon cycle feedbacks and demonstrate that they differ under CO₂ and non-CO₂ forcing. CO₂ forcing leads to both carbon-climate and carbon-concentration feedbacks, whereas non-CO₂ gases give rise to thea carbon-climate feedback only. We introduce a framework, building on previous studies that primarily addressed CO₂ forcing, to separate the carbon-climate feedback into a temperature term and a temperature—CO₂ cross term. Our findings reveal that these feedback terms are comparable in magnitude for the global ocean. This underscores the importance of considering both terms in carbon cycle feedback framework and climate change mitigation strategies.

Plain language summary. Mitigating non- CO_2 greenhouse gases, like methane and nitrous oxide, complements efforts to reduce CO_2 emissions in limiting global warming. In our study, we explore how reducing emissions of these gases compares to CO_2 mitigation using an Earth System Model. Both types of gases contribute to global warming, but their effects on climate vary by region. Additionally, we highlight that the carbon cycle responds differently depending on whether climate change is driven by CO_2 or non- CO_2 gases. This highlights the importance of considering both types of gases within carbon cycle analysis framework and climate change mitigation strategies.

1 Introduction

Increases in the atmospheric concentrations of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), predominantly cause human-induced climate change since the preindustrial period. They contributed nearly 63%, 11% and 6%, respectively, to the total effective radiative forcing (ERF) over 1960–2019 period (Canadell et al., 2021). Anthropogenic CO_2 emissions are dominated by the combustion of fossil fuels (FF) and land-use change (LUC), CH_4 emissions by FF and the agricultural sector, and N_2O emissions by the use of nitrogen fertiliser and manure. CH_4 and N_2O have atmospheric lifetimes of 11.8 ± 1.8 years and 109 ± 10 years and 100-year global warming potentials (CMP_1OO) of 27.9 and 273, respectively (Forster et al., 2021; CM_2OO).

40 Mitigation of non-CO₂ greenhouse gases (GHGs) is an essential strategy to limit global warming in the context of the Paris Agreement's temperature target (Abernethy et al., 2021; Jones et al., 2018; Rao and Riahi, 2006; de Richter et al., 2017; Tanaka et al., 2021). The reduction of CH₄ emissions can lead to a rapid decrease in the radiative forcing and may limit the peak warming (Mengis and Matthews, 2020; Montzka et al., 2011). To facilitate the achievement of the Paris Agreement temperature target, the Global Methane Pledge was adopted to reduce anthropogenic CH₄ emissions by 30% over the 2020–2030 period (CCAC, 2021). Several existing studies have confirmed the technical and socioeconomic capacities to reduce the global methane emissions and benefits for reducing atmospheric pollution (Höglund-Isaksson et al., 2020; Jackson et al., 2021; Malley et al., 2023; Nisbet et al., 2020). Atmospheric methane removal methods have also been discussed in some studies

(Boucher and Folberth, 2010; Jackson et al., 2021; Mundra and Lockley, 2023) but they are still in their infancy.

Numerous studies have investigated the impacts of changes in the emissions / concentrations of CO₂ and non-CO₂ GHGs on the Earth System. The Precipitation Driver and Response Model Intercomparison Project (PDRMIP) focused on the role of different climate change drivers on the mean and extreme precipitation changes using a set of idealised perturbed experiments (Myhre et al., 2017). Richardson et al. (2019) revealed spatial and temporal differences in the surface temperature response to different forcings, such as CO₂ and CH₄, in part due to the physiological CO₂ warming over the densely vegetated regions that is absent under non-CO₂ forcing. This physiological warming occurs because plants close their stomata under elevated CO₂, reducing transpiration and decreasing latent heat loss which causes a local surface warming. Nordling et al. (2021) further demonstrated that the change in long-wave clear-sky emissivity is the key driver forof the differences in temperature response differences—between GHGs forcings. Using intermediate-complexity Earth System Climate Model simulations, Nzotungicimpaye et al. (2023) showed that delaying methane mitigation has implications both for meeting the stringent temperature targets and for the climate over many centuries. Using Earth System model simulations, Tokarska et al. (2018) showed that non-CO₂ forcing reduces the remaining carbon budget due to its direct radiative effects on surface temperature,

causing additional warming. Fu et al. (2020) showed that non-CO₂ GHG₈, which that have a shorter atmospheric lifetime than CO₂-, may have long-term consequences on climate via through their impacts on the carbon cycle. Namely, warming from non-CO₂ GHGs weakens land and ocean carbon sinks.

The weakening of land and ocean carbon sinks due to non-CO₂ GHGs underscores the importance of understanding the differences in carbon cycle feedbacks between CO₂ and non-CO₂ GHGs. Only the changes in CO₂ concentrations give rise to athe carbon-concentration feedback, that is the response of the land and ocean carbon uptake to the changes in CO₂ concentration, mainly via the stimulation of photosynthesis through by the CO₂ fertilisation effect over on land and the solubility pump over in the ocean. The changes in both CO₂ and non-CO₂ concentrations lead to the carbon-climate feedback (γ), that is the response of the land and ocean carbon uptake to climate change, mainly via the increased plant and soil respiration over land and reduction of the CO₂ solubility in the ocean with warming (Arora et al., 2013; Schwinger et al., 2014; Zickfeld et al., 2011). Under changing CO₂ concentrations, land and ocean carbon storages respond to both carbon-concentration and carbon-climate feedbacks. However, the interaction between these feedbacks can introduce a non-linearity into the system, whereby the combined effect is not simply the sum of the individual feedbacks_(Schwinger et al., 2014; Zickfeld et al., 2011). Thus, temperature-mediated feedback can differ under changing versus constant CO₂ levels, an important distinction when comparing CO₂ and non-CO₂ GHG feedback mechanisms. Here, it is also important to acknowledgerecognize that other factors, such as time lags and potential irreversibilities in the climate system, may also contribute to these differences (Boucher et al., 2012; Chimuka et al., 2023; Schwinger et al., 2014).

interactions between changing atmospheric CO_2 and temperatures—can be comparable in size with γ (Schwinger et al., 2014; Zickfeld et al., 2011). They attributed the nonlinearity to the different responses of the land biosphere to the temperature changes, depending on the presence or absence of the CO_2 fertilisation effect, as well as the weakening of ocean circulation and mixing between water masses of different temperatures. However, these studies did not consider non- CO_2 GHGs.

Previous studies have also examined the impact of declining atmospheric CO_2 concentration on the climate and carbon cycle (Boucher et al., 2012; Chimuka et al., 2023; Jones et al., 2016; Koven et al., 2023; Melnikova et al., 2021; Schwinger and Tjiputra, 2018). During the period of decreasing atmospheric CO_2 concentration and temperature (ramp-down), the β and γ feedbacks arise from both the reduction in CO_2 levels and temperature and the inertia of the carbon cycle—specifically, the altered land and ocean carbon pools resulting from prior increases in the CO_2 concentration and temperature (Chimuka et al., 2023; Zickfeld et al., 2016). Melnikova et al. (2021) showed that this leads to an amplification of the β and γ feedbacks under decreasing CO_2 concentration and temperature. The effectiveness of non- CO_2 mitigation has been explored and is an integral part of the integrated assessment models (Ou et al., 2021; Rao and Riahi, 2006; Tanaka et al., 2021). However, few studies investigated the effects of declining non- CO_2 GHG concentrations on the climate and carbon cycle using Earth System Models (ESMs). Abernethy et al. (2021) used an ESM to demonstrate the effectiveness of methane removal in reducing global mean surface temperature, complementing negative CO_2 emissions. Thus, the purpose of this study is twofold:

- to clarify whether the climate and carbon cycle responses to declining ${\rm CO_2}$ and non- ${\rm CO_2}$ GHGs differ globally and regionally

95

 to investigate the nonlinearities of carbon cycle feedbacks under CO₂ and non-CO₂ GHG decrease, and the implications for climate change mitigation. Here, we conduct a se<u>ries</u>t of idealised CO₂ and non-CO₂ (CH₄ and N₂O) concentration-driven ramp-up and ramp-down experiments using the IPSL-CM6A-LR ESM. We then compare the global and spatial impacts of CO₂ and non-CO₂ concentration changes on climate and the carbon cycle under overshoot pathways.

2 Data and Methods

100

110

120

2.1 Recalibration of model's CH4 and N2O concentrations

We use Version 6 of the Institut Pierre-Simon Laplace (IPSL) low-resolution ESM, IPSL-CM6A-LR (Boucher et al., 2020), developed in the runup toof the sixth phase of the Coupled Model Intercomparison Project (CMIP). It comprises the LMDZ atmospheric model Version 6A and the ORCHIDEE land surface model Version 2.0 with a 144×143 spatial resolution, and the NEMO oceanic model Version 3 with a resolution of 1°.

Previous studies showed that IPSL-CM6A-LR can adequately estimate the ERF of CO₂ (Lurton et al., 2020). However, it underestimates the CH₄ radiative forcing for the historical period due to known limitations in the parameterization of gaseous optical properties in the Rapid Radiative Transfer Model (see Fig. 8 in Hogan and Matricardi (2020)). CH4 also absorbs in the shortwave spectrum, an effect that is not accounted for in the radiative transfer code used in IPSL-CM6A-LR and many other climate models. The Intergovernmental Panel on Climate Change (IPCC) Sixth Assessment Report (AR6) (Forster et al., 2021) estimated the CH₄ ERF to be 0.54 [0.43 to 0.65] W m⁻² for the 1750-2019 period. However, the estimated ERF of CH₄ at the top of the atmosphere (TOA) in IPSL-CM6A-LR is only 0.27 Wm⁻² for the 1850–2014 period. Note that in the above estimates, the ERF is defined as the difference in the net TOA flux between a model experiment with perturbed GHG concentration but fixed sea surface and ice temperatures and a control simulation with pre-industrial GHG concentrations. Thus, the estimates include (minimal) effects on the ERF from changes in land surface temperature because, unlike sea surface temperature, the land surface temperature is not prescribed (see Thornhill et al., (2021)). Likewise, the ERF of N2O may not be accurate in the model. This problem may not be specific to IPSL-CM6A-LR: other climate models (e.g., CNRM-CM6) share the same radiative transfer code and most radiative transfer models used in some climate models have some degree of inaccuracy because they are designed to be computationally efficient (Collins et al., 2006; Fyfe et al., 2021; Pincus et al., 2016). Thus, there is a need to represent the ERF of CH₄ and N₂O more accurately in order to better understand the effects of non-CO₂ GHGs mitigation on the Earth system. As developing better parameterizations of the gaseous optical properties is beyond the scope of this study, we have developed an approach that adjusts CH₄ and N₂O concentrations to 'effective' concentrations that generate CH₄ and N₂O ERFs consistent with the reference estimates of IPCC AR6 (see Appendix A). The effective concentrations of CH₄ and N₂O are used as input to the radiative transfer scheme of the climate model throughout the rest of this study. In the text and figures, these are presented as the actual (equivalent) concentrations.

2.2 Experiment design

135

140

We perform and analyse a series of idealised global mean CO₂ and non-CO₂ concentration-driven ensemble experiments as summarised in Table 1 and Fig. 1. Including 50-year ramp-up, ramp-down, and stabilisation periods allows for the exploration of responses to increasing or decreasing CO₂ and non-CO₂ (CH₄ and N₂O) concentrations, as well as the long-term consequences and reversibility of their impacts on the climate and the carbon cycle. The inclusion of CO₂ and non-CO₂ concentration-driven experiments—[CO₂] and [nonCO₂] (with comparable ERF levels), a combined CO₂ and non-CO₂ concentration-driven experiment [CO₂+ nonCO₂], a biogeochemically coupled (BGC) experiment where CO₂ forcing affects only the carbon cycle of land and ocean [CO₂bgc] (with minor temperature effects from CO₂ physiological forcing), and a radiatively coupled (RAD) experiment that includes only CO₂ radiative forcing [CO₂rad] (where CO₂ change does not affect the carbon cycle), enables exploring the impacts of different forcing components on the climate and carbon cycle as well as potential nonlinearities of feedbacks. Additionally, an experiment that combines nonCO₂ radiative forcing with CO₂ physiological forcing [CO₂bgc + nonCO₂] allows for the comparison of nonlinearities arising from combined carbon-concentration feedback and CO₂- and non-CO₂-driven carbon-climate feedback. It serves as the nonCO₂ counterpart of the [CO₂] experiment.

Table 1. Description of experiments. Note that all experiments are analysed relative to their [piControl] counterparts.

Experiment name	Description	Maximum ERF*	Included carbon cycle terms from Eq.2		
[CO ₂]	0.7% CO ₂ concentration increase per year from piControl for 50 years followed by 0.7% CO ₂ decrease for 50 years. After CO ₂ level returns to piControl, 50 years of stable piControl CO ₂ concentrations.	1.88 W m ⁻²	$\Delta U_{eta}, \Delta U_{\gamma,co2}, \Delta U_{\chi,co2}$		
[nonCO ₂]	2% CH ₄ and 2% N ₂ O concentration increase per year from piControl for 50 years followed by 2% CH ₄ and 2% N ₂ O decrease for 50 years. After CH ₄ and N ₂ O levels return to piControl, 50 years of stable piControl CH ₄ and N ₂ O concentrations.	1.83 W m ⁻²	$\Delta U_{\gamma,nonCO2}$		
$[CO_2 + nonCO_2] \\$	Combined [CO ₂] and [nonCO ₂].				
[CO ₂ bgc]	Biogeochemically coupled [CO ₂].		ΔU_eta		
[CO ₂ rad]	Radiatively coupled [CO ₂].		$\Delta U_{\gamma,CO2}$		
[CO ₂ bgc + nonCO ₂]	[CO ₂ bgc] and [nonCO ₂] combined.		$\Delta U_{\beta},\Delta U_{\gamma,nonCO2},\Delta U_{\chi,nonCO2}$		
Combinations of experis	ments				
$[CO_2]-[CO_2bgc] \\$	Combination for comparison with [CO ₂ rad]		$\Delta U_{\gamma,CO2},\Delta U_{\chi,CO2}$		
$\begin{aligned} &[CO_2bgc + nonCO_2] - \\ &[CO_2bgc] \end{aligned}$	Combination for comparison with [nonCO ₂]		$\Delta U_{\gamma,nonCO2}$, $\Delta U_{\chi,nonCO2}$		
$[CO_2] + [nonCO_2]$	Combination for comparison with $[CO_2 + nonCO_2]$		$\Delta U_{\beta},\Delta U_{\gamma,CO2},\Delta U_{\chi,CO2},\Delta U_{\gamma,nonCO2}$		

*according to equations by Etminan et al. (2016), warming from the physiological CO₂ forcing is assumed to be negligible.

145

The experiment design uses a fixed land cover and constant (other than CO₂, CH₄ and N₂O) GHG and aerosol forcings that might otherwise interfere with the interpretation of the results. The maximum ERF in our experiments is 3.69 W m⁻², as estimated from the equations by Etminan et al. (2016) (see also Appendix A), corresponding to (actual) CO₂ concentration of 403 ppm, CH₄ concentration of 2175 ppb and N₂O concentration of 735 ppb. This ERF level (very much in line with the current CO₂ concentration level of ca. 420 ppm) makes our experiments and results relevant to mitigation efforts in the near future. The small differences in ERF between the [CO₂] and [nonCO₂] experiments are not significant when considering rampup, ramp-down and full periods at p < 0.05.

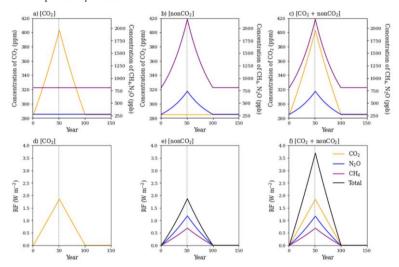


Figure 1: Time series of input (a-c) CO₂, CH₄ and N₂O concentrations and (d-f) their respective radiative ERFs according to Etminan et al. (2016) equations for (a, d) [CO₂], (b, e) [nonCO₂] and (c, f) [CO₂ + nonCO₂] experiments. Note the different scales on the y-axes in panels a-c for CO₂ concentrations in ppm (left) and other GHG concentrations in ppb (right).

We investigate CO_2 and $non-CO_2$ impacts on the climate by looking at the differences between $[CO_2]$ and $[nonCO_2]$ experiments and $[CO_2rad]$ and $[nonCO_2]$ experiments, hereafter referred to as $[CO_2]$ – $[nonCO_2]$ and $[CO_2rad]$ – $[nonCO_2]$, respectively. The experiments manipulate CH_4 and N_2O concentrations simultaneously because our primary focus is to compare the effects of CO_2 with those of $non-CO_2$ gases (i.e., CH_4 and N_2O combined) in this study.

<u>For each experiment, t</u>Three ensemble members are branched from <u>the</u> years 1870, 2020, and 2170 of the CMIP6 piControl experiment, hereafter [piControl]. <u>We note that the use of three members is not ideal, but it is a common compromise between</u>

computational cost and sampling the uncertainty due to climate variability. We estimate the changes relative to the corresponding [piControl] periods in order to avoid the effects of low-frequency internal climate variability from the piControl (Fig. S1), as discussed in Bonnet et al. (2021). When reporting carbon sink/source in the following sections, we refer to the fluxes relative to the [piControl]. For diagnosing Atlantic Meridional Overturning Circulation (AMOC), we utilised the ocean overturning mass streamfunction in depth space (msftyz variable in CMIP6). Specifically, we calculated the maximum annual mean value of the streamfunction in the Atlantic basin north of 20°N through all model's depth layers (up to ca. 5800 m).

2.3 Carbon cycle feedback attribution

Traditionally, carbon cycle feedback analysis relies on fully coupled [CO₂], biogeochemically-coupled [CO₂bgc] and radiatively-coupled [CO₂rad] simulations (Arora et al., 2013, 2020; Friedlingstein et al., 2006; Gregory et al., 2009; Schwinger et al., 2014; Schwinger and Tjiputra, 2018; Williams et al., 2019; Zickfeld et al., 2011). The carbon uptake (ΔU) can then be derived using the well-established carbon cycle feedback framework as a sum of carbon-concentration β parameter (GtC ppm⁻¹) multiplied by the changes in the atmospheric CO₂ concentration ΔC_{CO2} (ppm) and carbon-climate γ feedback parameter (GtC

 $K^{\text{--}1}$ multiplied by the changes in surface temperature ΔT (K), using Eq. (1):

$$\Delta U = \beta \times \Delta C_{CO2} + \gamma \times \Delta T + \varepsilon. \tag{1}$$

Here, term ε refers to a residual term.

The β parameter can be estimated from the [CO₂bgc] - [piControl], using Eq. (2):

$$\beta = \frac{\Delta U_{BGC}}{\Delta C_{CO2}},\tag{2}$$

where ΔU_{BGC} is the carbon uptake in the BGC experiment [CO₂bgc]. The β feedback reflects the strengthening of changes in the land and ocean carbon uptake) pools driven by the changes in CO₂ concentrations. This positive response contributes to negative climate feedback by reducing CO₂ content and dampening climate change.

The γ parameter can be estimated from the $[CO_2 rad]$ - [piControl], using Eq. (3:

$$185 \quad \gamma = \frac{\Delta U_{RAD}}{\Delta T},\tag{3}$$

where ΔU_{RAD} is the carbon uptake in the RAD experiment [CO₂rad]. The γ feedback reflects the weakening of changes in the land and ocean carbon sinks-pools due to the changes in climate globally, albeit with regional variability (negative to the land and ocean). Thus, it contributes to positive climate feedback by increasing CO₂ content and accelerating climate change.

Many existing studies have estimated γ using the difference between the fully-coupled (COU) and BGC experiments, as a proxy for the RAD experiment (Arora et al., 2013, 2020; Asaadi et al., 2024; Friedlingstein et al., 2003, 2006; Melnikova et al., 2021). However, Zickfeld et al. (2011) and Schwinger et al. (2014) have shown that this substitution introduces a residual term ϵ , which can be derived from the difference between [CO₂] – [CO₂bgc] and [CO₂rad] - [piControl], using Eq. (4):

$$\varepsilon = \Delta U_{COU} - \Delta U_{BGC} - \Delta U_{RAD}. \tag{4}$$

Formatted: Subscript

These studies indicate that the residual 'nonlinearity' term depends on both CO₂ concentration and climate change, and it can be of the same order of magnitude as the γ term. Here, we propose that this residual nonlinearity be attributed to a cross term, χ. Although recent studies continue to subsume χ under the γ feedback—partly due to the absence of the [CO₂rad] experiment in some experimental designs, and also because this approach has been widely established in earlier research (Friedlingstein et al., 2003, 2006)—we show that these metrics become less well defined when examining the effects of both CO₂ and non-CO₂ GHGs on the carbon cycle.

200 In order to investigate the associated carbon-climate feedback nonlinearities, we propose the following theoretical framework. The changes in carbon uptake $\Delta U(\Delta C_{co2}, \Delta T)$ (GtC year⁻¹) in the COU simulation can be defined as a function of changes in CO₂ concentration (ΔC_{CO2}) and temperature (ΔT). Following Schwinger et al. (2014), the formulation can be expanded to a Taylor series up to the second-order terms:

205

210

215

$$\Delta U = \Delta U (\Delta C_{co2}, \ \Delta T) = \frac{\partial U}{\partial C_{Co2}} \Delta C_{co2} + \frac{\partial U}{\partial T} \Delta T + \frac{\partial^2 U}{\partial C_{Co2} \partial T} \Delta T \Delta C_{co2} + \frac{1}{2} \frac{\partial^2 U}{\partial C_{Co2}^2} (\Delta C_{co2})^2 + \frac{1}{2} \frac{\partial^2 U}{\partial T^2} (\Delta T)^2 + Res. \tag{5}$$

where ΔC_{CO2} and $\Delta T_{forc.}$ are the respective increments of CO₂ concentration and temperature relative to [piControl]. The third and higher order terms are defined as a residual (*Res.*). We found them to be negligible in our case. We can disentangle the first- and second-order terms of the right-hand side of the Eq. (5) into terms that are purely dependent on CO₂ (ΔU_{β}), on temperature (ΔU_{ν}) and the cross term (ΔU_{ν}), as follows:

 $\Delta U_{\beta} = \frac{\partial U}{\partial C_{CO2}} \Delta C_{CO2} + Res., \tag{6}$

$$\Delta U_{\gamma} = \frac{\partial U}{\partial T} \Delta T + Res., \tag{7}$$

$$\Delta U_{\chi} = \frac{\partial^2 U}{\partial C_{CO2} \, \partial T} \Delta T \Delta C_{CO2} + Res.. \tag{8}$$

For simplicity, the second-order terms of Eqs. (6) and (7) are included in *Res.*. Combining Eqs. (4) and (8), the χ feedback parameter may be quantified from:

$$\chi = \frac{\Delta U_{COU-BGC} - \Delta U_{RAD}}{\Delta T \Delta C_{CO2}}.\tag{9}$$

The carbon-concentration β feedback term ΔU_{β} may be estimated from [CO₂bgc] - [piControl], under the assumption that the physiological CO₂ warming and its impacts on the carbon cycle are negligible, consistent with findings of Asaadi et al., (2024). The γ feedback terms for CO₂ ($\Delta U_{\gamma,CO2}$) and non-CO₂ ($\Delta U_{\gamma,nonCO2}$) gases are estimated from [CO₂rad] - [piControl], and [nonCO₂] - [piControl], respectively. The difference between these terms ($\Delta U_{\gamma,CO2} - \Delta U_{\gamma,nonCO2}$) yields the difference between impacts of CO₂ and non-CO₂ forcing on the carbon-climate feedback. Finally, the cross term ΔU_{χ} , previously referred to as nonlinearity term (Arora et al., 2013, 2020; Gregory et al., 2009; Schwinger et al., 2014; Schwinger and Tjiputra, 2018; Williams et al., 2019; Zickfeld et al., 2011), may be estimated by utilising a combination of experiments. The combination

225 $[CO_2] - [CO_2bgc] - [CO_2rad]$ gives $\Delta U_{\chi,CO2}$, and the combination $[CO_2bgc + nonCO_2] - [CO_2bgc]$ - $[nonCO_2]$ gives $\Delta U_{\chi,nonCO2}$). Analogously, the difference between these cross terms $(\Delta U_{\chi,CO2} - \Delta U_{\chi,nonCO2})$ yields the difference between the CO_2 and $non-CO_2$ forcings on the χ .

3 Results and discussion

3.1 Climate impacts

The analysis of global climate variables as a function of CO₂ concentration and GSAT shown in Fig. 2 follows a previous study by Boucher et al. (2012). Consistent with their findings, our results show that GSAT change lags behind GHG forcing by up to a decade. The lag increases with the increase in the forcing magnitude, so that the largest lag is in the [CO₂ + nonCO₂] experiment. Even after a ramp-down period and 50 years of constant GHG forcing at [piControl] levels, GSAT does not return to preindustrial values in all experiments (Fig. 2a). This can be explained by the inertia of the climate system, apparent in the changes in the ocean heat uptake (OHU). The OHU increases during the ramp-up and decreases during the ramp-down period, being positive, i.e., taking up energy away from the atmosphere, during the ramp-up and first half of the ramp-down period. OHU turns negative by the end of the ramp-down and stays negative during the 50 years of the stabilisation period, releasing energy back into the atmosphere (Fig. 2c-e). The hysteresis of the climate system is evident from the nearly linear relationship between maximum GSAT and mean GSAT during the stabilisation period (Fig. S2). The thermosteric (unrelated to ice sheet melting) sea level increases in all experiments except for [CO₂bgc] and is closely related to the OHU. It does not recover (i.e. it is irreversible) within the time-horizon considered here (Fig. 2 i-k). The AMOC decreases with GSAT (with the strongest decrease reached for GSAT = 2 °C under [CO₂ + nonCO₂]), but fully recovers (Fig. S1b).

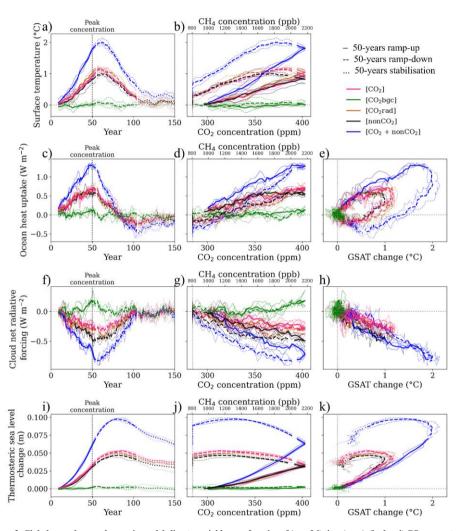


Figure 2: Global annual mean changes in model climate variables as a function of (a, c, f, i) time (year), (b, d, g, j) CO₂ concentration (ppm) / CH₄ concentration (ppb, only for [nonCO2]), (c, e, h, k) GSAT (°C) for (a, b) GSAT (°C), (c-e) ocean heat uptake (W m²), (f-h) cloud net radiative forcing (W m²), and (i-k) thermosteric sea level change (m) under selected scenarios. The ramp-up, ramp-down and stabilisation periods are indicated by different line styles in all panels. Thick lines indicate the ensemble means and thin lines correspond to three ensemble members.

The CO₂ physiological warming that can be quantified by comparing [CO₂bgc] with [piControl] is small (green line in Fig. 2).

Spatially, some differences are ubiquitous over land, e.g., CO₂ physiological warming persists over Eurasia during the rampup period, and over the high latitudes of both land and ocean during the stabilisation period (Fig. S3a). A larger ensemble size of model simulations would be required to investigate these differences more thoroughly. In our following analysis on carbon cycle feedbacks, we assume the CO₂ physiological warming to be negligible, consistent with previous findings of Asaadi et al., (2024).

When comparing CO₂- and non-CO₂-induced forcing ([CO₂] and [nonCO₂] experiments) at a global scale, our results are consistent with Richardson et al. (2019) who show the higher surface temperature response of CO₂ when compared to CH₄. When comparing CO₂- and non-CO₂-induced radiative forcing ([CO₂rad] and [nonCO₂] experiments) at a global scale, the non-CO₂ forcing still leads to a lower GSAT peak and a slightly lower peak of thermosteric sea level rise compared to the CO₂ radiative forcing (brown and black lines of Fig. 2a, significant difference at *p* < 0.05). This cannot be explained just by a slightly higher ERF of the [CO₂rad] compared to [nonCO₂] experiment (Table 1, Figs. 2a, S4). Our results are consistent with Nordling et al. (2021) who show the higher effective temperature response for CO₂ forcing compared to non-CO₂ forcing, attributing it to the changes in clear-sky planetary emissivity.

The combined effects of CO₂ physiological and radiative forcing do not lead to more warming, as the r_R adiative forcing alone ([CO₂rad] experiment) leads to a slightly higher global temperature increase compared to the coupled [CO₂] experiment, which includes the combined effect of CO₂ physiology and radiative forcing (Fig. 2a, b). This temperature difference is particularly evident in the Arctic region (Fig. S3a). Our findings differ from those of a CMIP5 intercomparison study, which reported that CO₂ physiological warming amplifies the Arctic warming (Park et al., 2020). The study showed that the CO₂ physiological effect contributes to high latitude warming by reducing evaporative cooling due to stomatal closure under elevated CO₂ levels. In contrast, we observe higher evapotranspiration in the [CO₂rad] compared to the [CO₂] experiment (Fig. S5), which is probably a consequence of the lower warming in the [CO₂] experiment. In our study, the greater warming in the [CO₂rad] experiment may be driven by increased surface albedo, especially over in the Arctic Ocean (Fig. S3b). While the underlying causes remain unclear, this pattern appears consistent in other experiments conducted with IPSL-CM6A-LR under moderate CO₂ levels (not shown). Because the ensemble size in our study is limited and the effects of the model's internal variability should be considerable, future research should validate the robustness of our findings with larger ensemble simulations.

275 3.2 Carbon cycle feedback

265

270

3.3 Carbon-concentration feedback

The term $\Delta U_{\beta,CO2}$ corresponds to the flux arising from variations in CO₂ concentration ([CO₂bgc] experiment). The $\Delta U_{\beta,CO2}$ dominates the <u>land and ocean</u> carbon uptake changes over the land and ocean through all three considered periods (Fig. 3, Tables 2, S1). Since the considered maximum surface warming levels are below 2 °C, land and ocean carbon fluxes are primarily controlled by the CO₂-induced effects during the ramp-up period, resulting in positive β (Fig. 3 a-d). Yet, these

Formatted: Subscript

effects are expected to decrease with higher warming (Arora et al., 2020; Melnikova et al., 2023). Nearly two-thirds of land and half of ocean carbon accumulated during the ramp-up period due to the atmospheric CO_2 increase is being released during the latter periods, consistent with previous studies (Asaadi et al., 2024; Chimuka et al., 2023). Over In the ocean, β is positive (carbon sink) in all regions during the ramp-up period. However, CO_2 concentration decrease in-induces carbon source over all ocean regions, except for the Atlantic and Southern Oceans (Fig. 4). All regions are carbon sources during the stabilisation period of the [CO₂bgc] experiment.

Table 2. Cumulative CO₂ and climate change-driven changes in the land and ocean carbon fluxes (GtC), shown as three- member ensemble mean. The \pm indicates one standard deviation among the three members. Note that all experiments are analysed relative to their [piControl] counterparts.

Experime	Max. warming	Terms	Years 1-50 (ramp-up)		Years 51-100 (ramp- down)		Years 101-150 (stabilisation)		Total	
nt	(K)*		Land	Ocean	Land	Ocean	Land	Ocean	Land	Ocean
[CO ₂ bgc]	0.1±0.0	$\Delta U_{eta,CO2}$	179.3±2. 2	103.8±0. 7	16.3±6.0	- 19.7±1.0	- 106.3±0.4	32.1±0.8	59.4±1.8	53.4±0.9
[CO ₂ rad]	1.1±0.1	$\Delta U_{\gamma,CO2}$	18.6±2.4	-2.4±0.2	4.5±5.7	0.0 ± 1.4	11.4±4.0	0.3±1.9	-2.2±2.0	-2.1±0.6
$[nonCO_2]$	0.9±0.1	$\Delta U_{\gamma,nonCO2}$	14.8±3.6	-2.3±1.0	1.3±6.0	-0.5±1.3	10.2±2.4	1.3±0.5	-2.5±2.9	-1.6±0.2
$[CO_2] - [CO_2bgc]$	1.0±0.0	$\Delta U_{\gamma,CO2} + \Delta U_{\chi,CO2}$	- 14.7±1.0	-4.5±0.2	3.8±3.0	-1.4±1.0	8.2±1.9	0.6±0.5	-3.8±3.1	-5.5±1.4
		$\Delta U_{\chi,CO2}$	3.9±2.1	-2.2±0.4	-0.7±7.3	-1.4±0.5	-3.2±5.9	0.2 ± 2.3	-1.6±5.0	-3.4±2.0
		$\Delta U_{\chi,nonCO2}$	3.5±1.5	-1.6±1.0	2.8 ± 7.9	0.1 ± 1.1	-1.6±1.5	-1.0±1.3	2.0±4.0	-2.5±1.0
		$\Delta U_{\gamma,CO2} - \Delta U_{\gamma,nonCO2}$	-3.8±5.0	-0.1±0.9	3.2±3.6	0.5±0.9	1.2 ± 2.6	-1.0±2.2	0.3 ± 2.7	-0.5±0.7
		$\Delta U_{\chi,CO2} - \Delta U_{\chi,nonCO2}$	0.4±3.5	-0.6±0.7	-3.5±3.4	-1.6±1.0	-1.6±7.4	1.3±3.3	-3.6±5.5	-1.0±2.8

^{*} defined as the mean $\Delta GSAT$ during years 41-60 in the experiments.

285

290

The transient change of the β feedback over land can be better explained by analysing the gross primary production (GPP) and the autotrophic and heterotrophic respiration (Ra and Rh) fluxes. Land GPP, representing photosynthetic uptake, increases during the ramp-up period under elevated CO_2 concentration and decreases almost linearly with decreasing CO_2 , showing only a small hysteresis (Figs. S6-7). In contrast, both autotrophic and heterotrophic respiration (Ra and Rh) exhibit a larger hysteresis, which leads to an extended period of the carbon release to the atmosphere. This suggests that while there may be initial carbon sequestration benefits gained during elevated CO_2 periods, these benefits are susceptible to being lost as CO_2 concentrations decline due to decreased photosynthesis and increased respiration, albeit at a reduced rate.

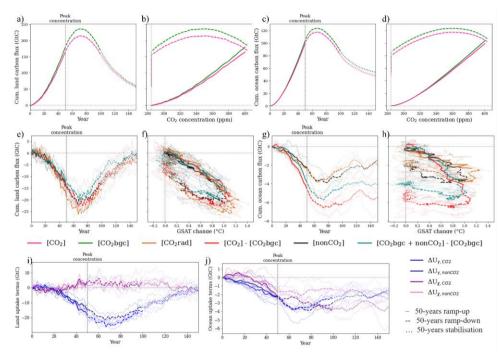
The spatial variation of cumulative net carbon uptake provides further details on the feedback changes (Figs. 4, S8-S10). During the CO_2 ramp-up phase, CO_2 increase triggers a land carbon sink in all regions. However, during the ramp-down phase, it induces a net carbon source over subtropical regions while still driving a land carbon sink in northern high-latitudes, so that global land becomes a carbon source in the middle of the ramp-down phase. Finally, during the stabilisation period, all land regions become net carbon sources.

Formatted Table

305 Subtropical and southern land regions exhibit a shorter hysteresis in response to decreasing CO₂ concentrations. This disparity arises from the larger proportion of carbon accumulated in aboveground vegetation biomass in southern regions, contrasting with the greater fraction stored in soils within northern latitudes (Figs. S9-S11). The extended period of high positive β in northern mid- to high latitudes can be attributed to the longer carbon turnover time, particularly in soils, compared to tropical regions (Fig. S8).

3.4 Carbon-climate feedback

The term ΔU_{γ} corresponds to the carbon flux arising from variations in radiative forcing (such as in the [nonCO₂] and [CO₂rad] experiments). The ΔU_{γ} for CO₂ and non-CO₂ ($\Delta U_{\gamma,CO2}$ and $\Delta U_{\gamma,nonCO2}$, respectively) are equivalent (within one standard deviation uncertainty range) under nearly equivalent levels of ERF (compare panels b and c of Fig. 4, see also $\Delta U_{\gamma,CO2} - \Delta U_{\gamma,nonCO2}$ in Table 2, see also Table S1). The γ is negative on a global scale over in the land and ocean, with greater magnitude but also faster reversibility over land. Spatially, land γ is positive in the mid- to high latitudes and negative in the tropical regions (Figs. 4, S9-10, see also Melnikova et al. (2021)). During the ramp-up, climate change drives carbon sink over in the northern mid- to high- latitudes and carbon source over in the subtropical regions and the Southern Hemisphere, with larger magnitude of changes in the experiments, in which CO₂ concentration change is present (Figs. 4, S9-11).



320 Figure 3. Global cumulative carbon fluxes (GtC) over (a, b, e, f, i) land and (c, d, g, h, j) ocean as a function of (a, c, e, g, i, j) time (year), (b, d) CO₂ concentration (ppm) and (f, h) GSAT changes (°C) under selected scenarios. The ramp-up, ramp-down and stabilisation periods are indicated by different line styles. Thick lines indicate the ensemble means and thin lines correspond to other ensemble members. Note that vertical axes differ between panels e, g and i, j.

3.5 Nonlinearity in carbon cycle feedback

The cross term ΔU_χ induces non-negligible differences between climate change-induced carbon flux when comparing experiments with presence or absence of atmospheric CO₂ concentration change. It reaches ca. 20–23% of the land ΔU_γ and ca. 70–90% of the ocean ΔU_γ, cumulative over the ramp-up period (Table 2). During the ramp-up phase, the ΔU_χ corresponds to a decrease in the climate-driven land carbon source and an increase in the climate-driven ocean carbon sink-source (Fig. 3). The χ feedback is positive (larger carbon sink) onever the land and negative (larger carbon source) inover the ocean (Table S1). There is no significant difference between CO₂ and non-CO₂ χ feedback at similar It is nearly equivalent for CO₂ and non-CO₂ under similar ERF levels of ERF for the land but not for the ocean (compare panels f and g of Fig. 3e-j, Fig. 4 f-g, see also Tables 2 and S1). Over In the ocean, the cross-term differences for CO₂ and non-CO₂ forcing arise already during the ramp-

Formatted: Subscript
Formatted: Subscript

up and propagate during the ramp-down and stabilisation phases, spatially concentrating in the deep mixing region of the Southern Ocean.

OverOn land, the positive χ results from reflects more biomass at high latitudes available for climate change effects, leading to a larger carbon sink (positive γ). During the ramp-down, climate warming through lagged (after GHG concentrations) temperature change (lagged after GHG concentrations change) increases the carbon sink over high latitudes and weakens the carbon source over in the tropics. Here the ΔU_{χ} indicates that reflects more biomass becomes available globally exposed to for the climate change effects, resulting inleading to a larger carbon source (negative γ). The differences (under in the presence / absence of the cross term ΔU_{χ}) diminish for land but not for the ocean during the ramp-down and stabilisation periods.

Over-In the ocean, the contribution from the nonlinearity of carbon cycle feedbacks leads to a greater reduction in the elimate CO_2 -driven carbon sink driven (Fig. 3). Unlike over the land, tT he contribution of the cross term ΔU_{χ} to the total ΔU increases during the ramp-down phases of considered GHG concentration scenarios. Previously, Schwinger and Tjiputra (2018), who considered nonlinearity of carbon cycle feedback, warned that RAD experiments may underestimate the carbon-climate feedback (when compared to COU - BGC experiments), because "the reduction of sequestration of preformed dissolved inorganic carbon under high atmospheric CO_2 is not taken into account." In this study, the nonlinearity effects nearly double climate change-driven carbon loss (compared to the RAD experiment, in which atmospheric CO_2 is constant) relative to the total net 150-year net ocean carbon uptake under the $[CO_2]$ experiment. Spatially, while the Southern Ocean remains the largest ocean carbon sink in all considered experiments involving atmospheric CO_2 changes, it, along with the Atlantic Ocean,

undergoes the largest climate change-driven reduction in carbon sink (Fig. 4).

345

Formatted: Subscript

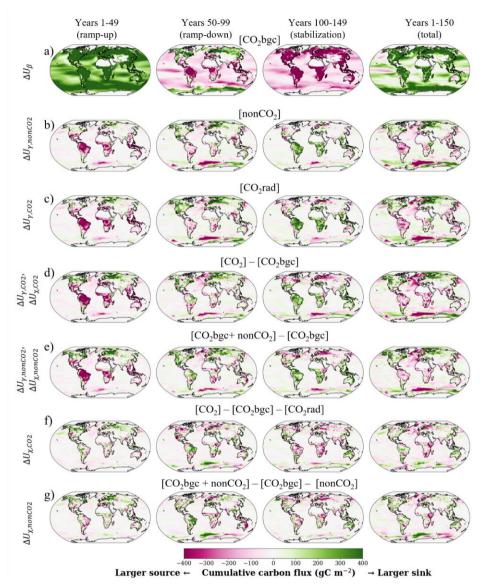


Figure 4. Spatial variation of land and ocean carbon fluxes (GtC, negative to the atmosphere) cumulative over 50 years of (first column) ramp-up, (second column) ramp-down, (third column) stabilisation phases and (last column) full 150-year period. The data for three-member-ensemble mean are used.

Our findings provide evidence on the effectiveness of non-CO₂ GHG mitigation. While it can effectively reduce GSAT peak, non-CO₂ GHG mitigation may also lead to smaller climate change-driven losses in the ocean carbon sink. In the real world, the presence / absence of ΔU_{χ_A} suggests disparities between CO₂ mitigation efforts and non-CO₂ mitigation efforts. The CO₂-and non-CO₂-driven climate change leads to unequal decrease in carbon uptake, especially apparent for the ocean on a global scale (compare red and black lines in Fig. 3, corresponding to $[CO_2] - [CO_2] = [CO_2$

In order to overcome the small signal-to-noise ratio of the considered experiments as well as the regional differences in the

3.6 Radiative forcing and carbon cycle feedback additivity

365

radiative forcing between [CO₂] and [nonCO₂] experiments, we compare the (1) [CO₂ + nonCO₂] experiment that include both CO₂ and non-CO₂ effects with (2) the sum of two [CO₂] and [nonCO₂], that include CO₂ and non-CO₂ effects, accordingly (Figs. 5 and S12-13). The climate effects, defined via temperature change, differ during the ramp-up and ramp-down periods (Fig. S12). These differences imply non-additivity of radiative forcing and can also be attributed to biophysical feedback. As for the carbon cycle feedback, the [CO₂ + nonCO₂] experiment that has all feedback is different from the sum of two experiments [CO₂] + [nonCO₂] both on over the land and in the ocean (Fig. 5). The differences are larger and stay longer inover the ocean. This implies non-additivity of carbon cycle feedback. From the proposed carbon cycle feedback attribution framework, the non-additivity arises from nonequality of $(\Delta U_{\gamma,CO2+nonCO2} + \Delta U_{\chi,CO2+nonCO2})$ and $(\Delta U_{\gamma,CO2} + \Delta U_{\chi,CO2} + \Delta U_{\chi,CO2+nonCO2})$. The significant difference (p < 0.1) onever the land is in the high-latitude region, where "all-effects together" [CO₂ + nonCO₂] experiment yields a larger carbon sink during the ramp-up phase. The largest difference inover the ocean is in the Southern Ocean, followed by the North Atlantic Ocean. The [CO₂ + nonCO₂] experiment has large carbon sink is in the Southern Ocean compared to the sum of two experiments. This might be related to the saturation of the decrease in the mixed layer depth with more warming, but a more thorough study is needed to confirm such phenomena.

Formatted: Font: (Default) +Headings (Times New Roman)
Formatted: Font: (Default) +Headings (Times New Roman)

Formatted: Font: (Default) +Headings (Times New Roman),

Formatted: Font: (Default) +Headings (Times New Roman)

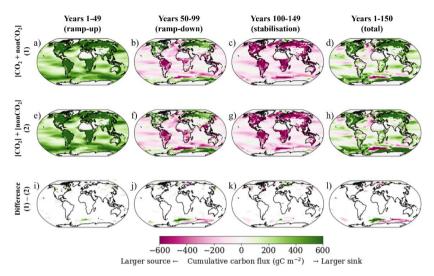


Figure 5. Spatial variation of three-member-ensemble mean land and ocean carbon fluxes (GtC, negative to the atmosphere) cumulative over 50 years of (a, e, i) ramp-up, (b, f, j) ramp-down, (c, g, k) stabilisation phases and (d, h, l) full 150-year period. We draw only significantly different grids between (i-l) $[CO_2 + nonCO_2]$ and $[CO_2] + [nonCO_2]$ experiments using three ensemble members (p < 0.1) based on t-test, N=60).

4 Limitations and future research directions

385

To our knowledge, this is the first study of its kind to compare idealised CO₂ and non-CO₂ ramp-up and ramp-down scenarios for their effects on global temperature change and the carbon-cycle feedbacks. Below we draw attention to the caveats and limitations that should be addressed in future studies.

First, since IPSL-CM6A-LR, like all other ESMs participating in CMIP6, does not have interactive modules of the CH₄ and N_2O cycles, the changes in stratospheric water vapour, aerosols, and tropospheric ozone due to atmospheric CH₄ changes, as well as the effects of nitrogen deposition on the carbon cycle are not considered in this study. Future studies could consider simulations separately for CH₄ and N_2O . However for this study, the use of the model is justified because current changes in CH₄ and N_2O concentrations are primarily driven by anthropogenic sources, suggesting that the absence of interactive modules of natural sink/source processes does not significantly affect the representation of natural variability trends for the CH₄ and N_2O concentration (Nakazawa, 2020; Palazzo Corner et al., 2023; Zhu et al., 2013).

Second, when interpreting the results, it should be kept in mind that some carbon cycle processes in IPSL-CM6A-LR such as permafrost and fire are not considered. Yet, these should have only a limited impact on our results, given the (relatively) small

warming levels in the considered experiments. Previous studies have shown that IPSL-CM6A-LR estimates one of the smallest soil carbon pools among CMIP6 models, which may lead to an underestimation of the carbon-climate feedback (Arora et al., 2020; Melnikova et al., 2021).

Third, the results of the present study are limited by the use of a single ESM and a small number of ensemble members. Conducting similar experiments with other ESMs and using larger ensemble runs, which are particularly valuable in the low warming scenarios, as well as complementing the findings of our study with emission-driven experiments, could contribute to validating and extending our findings.

5 Conclusions

405

410

415

This study first presents a novel approach to recalibrate the ERF of CH₄ and N₂O in ESMs without changing the radiative scheme of the model. We then discuss the effects of increases and decreases in the concentrations of the CO₂ and non-CO₂ GHGs on the surface climate and carbon cycle. We find only small differences between CO₂ and non-CO₂ ramp-up and ramp-down forcing on global and regional climate.

The differences in climate responses can be linked to differences in the carbon cycle feedbacks. We show that CO₂- and non-CO₂-driven carbon-climate feedback are nearly equivalent at a global scale. However, increasing atmospheric CO₂ amplifies the reduction of the climate change-driven carbon sink, especially inover the ocean. We propose a novel framework to disentangle the carbon-climate feedback into a component that is purely driven by climate change i.e., expressed as a temperature term, and a component driven by climate change and rising atmospheric CO₂ at the same time, i.e., a cross term. Since the cross term can be quantified from the difference between COU, BGC and RAD simulations, we advocate for continuing to carry out all three types of experiments in the future phases of CMIP. We further warn that the cross term and non-additivity of feedback should be considered in the simple climate models (emulators).

Finally, this study showcases the additional benefits of non-CO₂ GHG mitigation on a smaller reduction of the ocean carbon sinks under overshoot scenarios. We stress that our findings do not imply that non-CO₂ GHG mitigation should be given-the a priority to over other means to mitigate climate change but they provide an insight on the intricate interplay between the carbon-concentration and CO₂- and non-CO₂-driven carbon-climate feedbacks to inform comprehensive mitigation strategies.

Appendix A

A set of 40-year idealised IPSL-CM6A-LR simulations (840 years in total) has been carried out. In these experiments, the sea surface temperature (SST) and sea ice fractions were fixed to their preindustrial levels. CH₄ concentration levels were kept at 2×CH₄, 3×CH₄, 4×CH₄, 5×CH₄, 8×CH₄, and 12×CH₄, and N₂O concentration levels were kept at 1×N₂O, 1.25×N₂O, and 1.75×N₂O (Table A1).

Table A1. Description of idealised recalibration experiments using IPSL-CM6A-LR

No	Name	Concentration				
		CH ₄ (ppb)	N ₂ O (ppb)	CO ₂ (ppm)		
0	piClim	808.25	273.02	284.32		
1	$2\times CH_4$	1616.50	273.02	284.32		
2	$3\times CH_4$	2424.75	273.02	284.32		
3	$4\times CH_4$	3233.00	273.02	284.32		
4	$5 \times CH_4$	4041.25	273.02	284.32		
5	$8 \times CH_4$	6466.00	273.02	284.32		
6	$12 \times CH_4$	9699.00	273.02	284.32		
7	$1 \times CH_4$ - $1d25N_2O$	808.25	341.28	284.32		
8	$2\times CH_4$ - $1d25N_2O$	1616.50	341.28	284.32		
9	$3\times CH_4$ -1d25N ₂ O	2424.75	341.28	284.32		
10	$4\times CH_4$ -1d25N ₂ O	3233.00	341.28	284.32		
11	$5 \times CH_4$ -1d25N ₂ O	4041.25	341.28	284.32		
12	8×CH ₄ -1d25N ₂ O	6466.00	341.28	284.32		
13	$12 \times CH_4$ -1d25N ₂ O	9699.00	341.28	284.32		
14	$1 \times CH_4$ -1d5N ₂ O	808.25	409.53	284.32		
15	$2 \times CH_4$ -1d5N ₂ O	1616.50	409.53	284.32		
16	$3\times CH_4$ -1d5N ₂ O	2424.75	409.53	284.32		
17	$4\times CH_4$ -1d5N ₂ O	3233.00	409.53	284.32		
18	$5 \times CH_4$ -1d $5N_2O$	4041.25	409.53	284.32		
19	$8 \times CH_4$ -1d5N ₂ O	6366.00	409.53	284.32		
20	12×CH ₄ -1d5N ₂ O	9699.00	409.53	284.32		
21	$1\times CH_4$ -1d75N ₂ O	808.25	477.79	284.32		

⁴³⁰ Fig. A1 shows the set of 40-year time series of global mean radiative forcing based on 21 idealised experiments. The *piClim* experiment holds pre-industrial levels of CO₂, CH₄ and N₂O concentrations. The mean interannual variation of the radiative forcings (one standard deviation) is 0.15 Wm⁻². The first 10 years of the experiments were dropped to allow the climate to adjust to the new radiative equilibrium after an abrupt change from the pre-industrial levels. The last 30 years were used to obtain eighteen data-points of mean global ERF by IPSL-CM6A-LR relative to the levels obtained from the *piClim* experiment.

⁴³⁵ The ERF is estimated as a TOA imbalance difference between each experiment and the piClim.

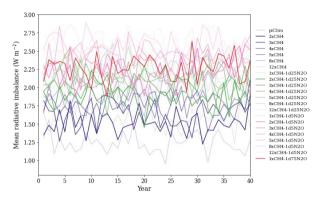


Figure A1. Time series of global mean radiative imbalance of IPSL-CM6A-LR idealised experiments

There are two frequently used sets of equations to derive radiative forcing of well mixed greenhouse gases, e.g., CO₂, CH₄ and N₂O, etc., based on their concentrations. The first set from Myhre et al. (Myhre et al., 1998), thereafter, M98, was used in IPCC AR3:

$$RF(CO2) = 5.35 \times ln \left(CO2/CO2_{t=0}\right)$$

440

445

$$RF(CH4) = 0.036 \times (\sqrt{CH4} - \sqrt{CH4_{t=0}}) - (f(CH4, N2O_{t=0}) - f(CH4_{t=0}, N2O_{t=0}))$$

$$RF(N2O) = 0.12 \times (\sqrt{N2O} - \sqrt{N2O_{t=0}}) - (f(CH4_{t=0}, N2O) - f(CH4_{t=0}, N2O_{t=0}))$$
(A1)

$$f(CH4, N20) = 0.47 \times ln[1 + 2.01 \times 10^{-5} \times (CH4 \times N20)^{0.75} + 5.31 \times 10^{-15} \times CH4 \times (CH4 \times N20)^{1.52}]$$

Here the CO₂, CH₄ and N₂O indicate their concentrations, where the units are ppm, ppb, and ppb.

Etminan et al. (2016), thereafter, E16, improved the IPCC AR3 equations by inclusion of the shortwave (near-infrared) bands of CH₄. Their set of equations was used in IPCC AR6. The radiative forcing of N_2O in the M98 equation depends on the CH₄ and N_2O concentrations, while the radiative forcing of N_2O in the equation by E16 depends on the CO₂, CH₄ and N_2O concentrations:

$$RF(CO2) = [-2.4 \times 10^{-7} \times (\text{CO2} - CO2_{t=0})^2 + 7.2 \times 10^{-4} \times |\text{CO2} - CO2_{t=0}|$$

$$-2.1 \times 10^{-4} \times \frac{1}{2} (\text{N2O} + \text{N2O}_{t=0}) + 5.36] \ln \left(\frac{\text{CO2}}{CO2_{t=0}} \right)$$

$$RF(CH_4) = \left[-1.3 \times 10^{-6} \times \frac{1}{2} (\text{CH4} + CH4_{t=0}) - 8.2 \times 10^{-6} \times \frac{1}{2} (\text{N2O} + \text{N2O}_{t=0}) + 0.043 \right] \times (\sqrt{\text{CH4}} - \sqrt{CH4_{t=0}})$$

$$(A2)$$

$$\begin{split} RF(N_2O) &= [-8.0\times10^{-6}\times\frac{1}{2}(\text{CO2}+CO2_{t=0}) + (4.2\times10^{-6}\times\frac{1}{2}(\text{N2O}+N2O_{t=0})\\ &-4.9\times10^{-6}\times\frac{1}{2}(\text{CH4}+\text{CH4}_{t=0}) + 0.117]\times(\sqrt{N2O}-\sqrt{N2O_{t=0}}) \end{split}$$

In the sets of Eqs. (A2), the radiative forcing due to CH₄ depends not only on the CH₄ concentration but also on that of N₂O (and conversely) because CH₄ and N₂O absorption bands overlap to some extent. These simplified equations are therefore additive: the radiative forcing due to a change in CH₄ and N₂O concentrations (CH_4 , N_2O) relative to reference (preindustrial) values ($CH4_{t=0}$, $N2O_{t=0}$) is equal to:

$$F(CH4, N2O) = RF_{CH4}(CH4, N2O) + RF_{N2O}(H4, N2O)$$
(A3)

The effect of CO_2 on the radiative forcing of N_2O is small (<5%), and thus, for simplicity, is neglected in the rest of this study (Fig. A2).

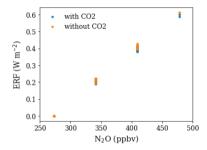


Figure A2.The ERF estimated from E16 with and without accounting for CO_2 impact on the N_2O forcing (including/excluding the term $-8.0 \times 10^{-6} \times 0.5 \times (CO_2 + CO_{2,t=0})$ in the equation) using the concentration values of the idealised experiments described in the Table 1 with preindustrial CO_2 concentration (284.32 ppm) and the same set of the experiments with $3 \times CO_2$ concentration (852.96 ppm).

Figure A3 shows the ERFs simulated by IPSL-CM6A-LR and those estimated from the two equations from the IPCC report and revised by E16. IPSL-CM6A-LR underestimates CH4 ERF and overestimates N₂O ERF, relative to both equations. Considering the improvements introduced by E16 to include the shortwave bands of CH₄, we use their equations (and not of IPCC AR3) to recalibrate the IPSL-CM6A-LR model.

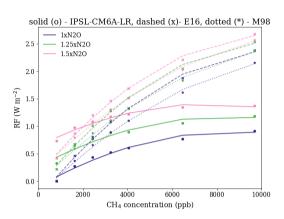


Figure A3. The ERF of CH4 and N2O from IPSL-CM6A-LR idealised experiments (points and fitted solid lines) and estimated from M98 (* and fitted dotted lines) and E16 (x and fitted dashed lines) equations, fitted to polynomial regressions for three levels of N_2O concentrations.

A system of equations can convert the input CH_4 and N_2O concentrations to the effective concentrations so that, if used in the climate model, would yield actual forcing from the equations by E16. Among several linear and nonlinear functions to relate the actual concentrations of CH_4 and N_2O with the concentrations seen by IPSL-CM6A model (effective concentrations) that have been tested, the following set of equations yielded the best fit:

$$\begin{split} CH_4^{effective} &= CH4_{t=0} + a \times (\text{CH4}^{actual} - CH4_{t=0})^c \\ N2O^{effective} &= \text{N2O}_{t=0} + b \times (N2O^{actual} - N2O_{t=0})^d \end{split} \tag{A4}$$

The initial values of CH₄ and N_2O concentrations are fixed at the preindustrial levels (t = 0). Those of the effective CH₄ and N_2O concentrations are also assumed to take the same respective preindustrial levels. The Eq. (A1) is used to relate the effective concentrations of CH₄ and N_2O of IPSL-CM6A-LR to the ERF estimated by E16:

$$\begin{split} RF_{Etm} &= RF_{CH4,Etm} + RF_{N2O,Etm} \\ &= \left(-1.3 \times 10^{-6} \times \frac{1}{2} (\text{CH4}^{effective} + CH4_{t=0}) - 8.2 \times 10^{-6} \times \frac{1}{2} (\text{N2O}^{effective} + N2O_{t=0}) \right. \\ &+ \left. 0.043 \right) \times (\sqrt{\text{CH4}^{effective}} - \sqrt{CH4_{t=0}}) + (4.2 \times 10^{-6} \times \frac{1}{2} (N2O^{effective} + N2O_{t=0}) \right. \\ &- 4.9 \times 10^{-6} \times \frac{1}{2} (\text{CH4}^{effective} + CH4_{t=0}) + 0.117) \times (\sqrt{N2O^{effective}} - \sqrt{N2O_{t=0}}) \end{split}$$

465

470

We optimise the four parameters of Eq. (A4) by minimising the sum of squared residuals between ERF estimated by E16 equations and simulated by IPSL-CM6A-LR using a Python implementation of the Limited-memory Broyden-Fletcher-Goldfarb-Shanno bound-constrained algorithm (L-BFGS-B) that is an algorithm for solving large nonlinear optimization problems with simple bounds (Byrd et al., 1995). The cost function is defined as:

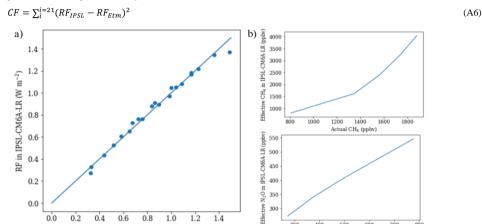


Figure A4. (a) Scatterplot of ERF simulated by IPSL-CM6A and estimated by E16 equations using the corrected effective concentrations from Eq. (A3), and (b) The effective IPSL-CM6A-LR concentrations of CH4 and N₂O as a function of the actual concentrations derived via a system of nonlinear functions.

Actual N₂O (ppbv)

The residual error after fit equals 0.03 (Wm⁻²)² (Fig. A4). The solution of the cost function provides the estimates of four uncertain parameters for Equations 3, indicated in Table A2. The effective IPSL-CM6A-LR concentrations of CH4 and N2O are functions of the actual concentrations (Fig. A4b). Higher effective CH₄ and lower N₂O effective concentrations are needed for IPSL-CM6A-LR to reproduce the ERF in agreement with IPCC estimates.

Table A2. Estimated parameters in Eq. (A4)

0.6 8.0 1.0 1.2

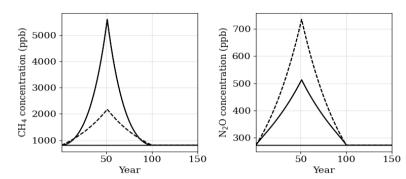
RF E16 with corrected CH₄,N₂O (W m⁻²)

0.0

490

Parameter	Estimate
a	19.44701978
b	0.84856644
c	0.49593024
d	1.13865386

The estimated parameters were applied to derive the effective IPSL-CM6A-LR concentrations for a target scenario (Fig. A5). Higher CH₄ and lower N₂O concentrations are required to reproduce the ERF correctly.



495 Figure A5. The actual (dashed lines) and new recalibrated effective CH₄ and N₂O IPSL-CM6A-LR concentrations (solid lines) for the CH₄-N₂O experiment of this study.

Data and materials availability

The piControl output data from the CMIP6 simulations are available from the CMIP6 archive: https://esgf-node.llnl.gov/search/cmip6 (WCRP, 2022)), additional IPSL-CM6A-LR outputs are available upon request. The Jupyter notebook and data to reproduce the figures are stored in the Zenodo archive: https://doi.org/10.5281/zenodo.12200056.

Acknowledgments

The IPSL-CM6 simulations were performed using the HPC resources of TGCC under the allocation 2022-A0120107732 and 2023-A0140107732 (project genemip6) provided by GENCI (Grand Equipment National de Calcul Intensif). This work benefited from the technical support and discussions with Adriana Sima and Nicolas Lebas at IPSL, Arnaud Caubel and Sébastien Nguyen at LSCE (France) and Michio Kawamiya at JAMSTEC (Japan).

Funding

505

510

This work benefited from State assistance managed by the National Research Agency in France under the Programme d'Investissements d'Avenir under the reference ANR-19-MPGA-0008. We further acknowledge the Program for the Advanced Studies of Climate Change Projection (SENTAN, grant number JPMXD0722681344) from the Ministry of Education, Culture, Sports, Science and Technology, Japan and KAKENHI (JP24K20979) of the Japan Society for the Promotion of Science. We acknowledge the European Union's Horizon Europe research and innovation program under Grant Agreement N° 101056939 (RESCUE – Response of the Earth System to overshoot, Climate neUtrality and negative Emissions) and N° 101081193

(OptimESM – Optimal High Resolution Earth System Models for Exploring Future Climate Changes). P.C. acknowledges support from the CALIPSO (CArbon Losses in Plants, Soils and Ocean) project funded through the generosity of Eric and Wendy Schmidt by recommendation of the Schmidt Futures program.

Author contributions

IM and OB initiated the study. OB, Ktan and IM developed methodology and study ideas. IM and OB performed IPSL-CM6A-LR simulations. IM performed data analysis and wrote the initial draft. All authors contributed to the discussions, writing, and revising the paper.

520 Competing interests

Authors declare that they have no competing interests.

References

Abernethy, S., O'Connor, F. M., Jones, C. D., and Jackson, R. B.: Methane removal and the proportional reductions in surface temperature and ozone, Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences, 379, 20210104, https://doi.org/10.1098/rsta.2021.0104, 2021.

Arora, V. K., Boer, G. J., Friedlingstein, P., Eby, M., Jones, C. D., Christian, J. R., Bonan, G., Bopp, L., Brovkin, V., Cadule, P., Hajima, T., Ilyina, T., Lindsay, K., Tjiputra, J. F., and Wu, T.: Carbon–Concentration and Carbon–Climate Feedbacks in CMIP5 Earth System Models, J. Climate, 26, 5289–5314, https://doi.org/10.1175/JCLI-D-12-00494.1, 2013.

- Arora, V. K., Katavouta, A., Williams, R. G., Jones, C. D., Brovkin, V., Friedlingstein, P., Schwinger, J., Bopp, L., Boucher,
 O., Cadule, P., Chamberlain, M. A., Christian, J. R., Delire, C., Fisher, R. A., Hajima, T., Ilyina, T., Joetzjer, E., Kawamiya, M., Koven, C. D., Krasting, J. P., Law, R. M., Lawrence, D. M., Lenton, A., Lindsay, K., Pongratz, J., Raddatz, T., Séférian, R., Tachiiri, K., Tjiputra, J. F., Wiltshire, A., Wu, T., and Ziehn, T.: Carbon–concentration and carbon–climate feedbacks in CMIP6 models and their comparison to CMIP5 models, Biogeosciences, 17, 4173–4222, https://doi.org/10.5194/bg-17-4173-2020, 2020.
- Asaadi, A., Schwinger, J., Lee, H., Tjiputra, J., Arora, V., Séférian, R., Liddicoat, S., Hajima, T., Santana-Falcón, Y., and Jones, C. D.: Carbon cycle feedbacks in an idealized simulation and a scenario simulation of negative emissions in CMIP6 Earth system models, Biogeosciences, 21, 411–435, https://doi.org/10.5194/bg-21-411-2024, 2024.
- Bonnet, R., Swingedouw, D., Gastineau, G., Boucher, O., Deshayes, J., Hourdin, F., Mignot, J., Servonnat, J., and Sima, A.: Increased risk of near term global warming due to a recent AMOC weakening, Nature Communications, 12, 6108, https://doi.org/10.1038/s41467-021-26370-0, 2021.
 - Boucher, O. and Folberth, G. A.: New Directions: Atmospheric methane removal as a way to mitigate climate change?, Atmospheric Environment, 44, 3343–3345, https://doi.org/10.1016/j.atmosenv.2010.04.032, 2010.

- Boucher, O., Halloran, P. R., Burke, E. J., Doutriaux-Boucher, M., Jones, C. D., Lowe, J., Ringer, M. A., Robertson, E., and Wu, P.: Reversibility in an Earth System model in response to CO 2 concentration changes, Environ. Res. Lett., 7, 024013, https://doi.org/10.1088/1748-9326/7/2/024013, 2012.
 - Boucher, O., Servonnat, J., Albright, A. L., Aumont, O., Balkanski, Y., Bastrikov, V., Bekki, S., Bonnet, R., Bony, S., Bopp, L., Braconnot, P., Brockmann, P., Cadule, P., Caubel, A., Cheruy, F., Codron, F., Cozic, A., Cugnet, D., D'Andrea, F., Davini, P., Lavergne, C. de, Denvil, S., Deshayes, J., Devilliers, M., Ducharne, A., Dufresne, J.-L., Dupont, E., Éthé, C., Fairhead, L., Falletti, L., Flavoni, S., Foujols, M.-A., Gardoll, S., Gastineau, G., Ghattas, J., Grandpeix, J.-Y., Guenet, B., Guez, L., E.,
- Guilyardi, E., Guimberteau, M., Hauglustaine, D., Hourdin, F., Idelkadi, A., Joussaume, S., Kageyama, M., Khodri, M., Krinner, G., Lebas, N., Levavasseur, G., Lévy, C., Li, L., Lott, F., Lurton, T., Luyssaert, S., Madec, G., Madeleine, J.-B., Maignan, F., Marchand, M., Marti, O., Mellul, L., Meurdesoif, Y., Mignot, J., Musat, I., Ottlé, C., Peylin, P., Planton, Y., Polcher, J., Rio, C., Rochetin, N., Rousset, C., Sepulchre, P., Sima, A., Swingedouw, D., Thiéblemont, R., Traore, A. K., Vancoppenolle, M., Vial, J., Vialard, J., Viovy, N., and Vuichard, N.: Presentation and Evaluation of the IPSL-CM6A-LR
 Climate Model, Journal of Advances in Modeling Earth Systems, 12, e2019MS002010,

https://doi.org/10.1029/2019MS002010, 2020.

- Byrd, R. H., Lu, P., Nocedal, J., and Zhu, C.: A Limited Memory Algorithm for Bound Constrained Optimization, SIAM J. Sci. Comput., 16, 1190–1208, https://doi.org/10.1137/0916069, 1995.
- Canadell, J. G., Monteiro, P. M., Costa, M. H., Da Cunha, L. C., Cox, P. M., Alexey, V., Henson, S., Ishii, M., Jaccard, S., and Koven, C.: Global carbon and other biogeochemical cycles and feedbacks., in: Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, vol. Chapter 5. Cambridge University Press. 2021.
 - Chimuka, V. R., Nzotungicimpaye, C.-M., and Zickfeld, K.: Quantifying land carbon cycle feedbacks under negative CO2 emissions, Biogeosciences, 20, 2283–2299, https://doi.org/10.5194/bg-20-2283-2023, 2023.
- 565 Collins, W. D., Ramaswamy, V., Schwarzkopf, M. D., Sun, Y., Portmann, R. W., Fu, Q., Casanova, S. E. B., Dufresne, J.-L., Fillmore, D. W., Forster, P. M. D., Galin, V. Y., Gohar, L. K., Ingram, W. J., Kratz, D. P., Lefebvre, M.-P., Li, J., Marquet, P., Oinas, V., Tsushima, Y., Uchiyama, T., and Zhong, W. Y.: Radiative forcing by well-mixed greenhouse gases: Estimates from climate models in the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4), Journal of Geophysical Research: Atmospheres, 111, https://doi.org/10.1029/2005JD006713, 2006.
- 570 Etminan, M., Myhre, G., Highwood, E. J., and Shine, K. P.: Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing, Geophysical Research Letters, 43, 12,614-12,623, https://doi.org/10.1002/2016GL071930, 2016.
- Forster, P., Storelvmo, T., Armour, K., Collins, W., Dufresne, J.-L., Frame, D., Lunt, D. J., Mauritsen, T., Palmer, M. D., Watanabe, M., Wild, M., and Zhang, H.: The Earth's Energy Budget, Climate Feedbacks, and Climate Sensitivity, edited by: Masson-Delmotte, V., Zhai, P., Pirani, A., Connors, S. L., Péan, C., Berger, S., Caud, N., Chen, Y., Goldfarb, L., Gomis, M. I., Huang, M., Leitzell, K., Lonnoy, E., Matthews, J. B. R., Maycock, T. K., Waterfield, T., Yelekçi, O., Yu, R., and Zhou, B., Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change, 923–1054, https://doi.org/10.1017/9781009157896.009, 2021.
- Friedlingstein, P., Dufresne, J.-L., Cox, P. M., and Rayner, P.: How positive is the feedback between climate change and the carbon cycle?, Tellus B: Chemical and Physical Meteorology, https://doi.org/10.3402/tellusb.v55i2.16765, 2003.
 - Friedlingstein, P., Cox, P., Betts, R., Bopp, L., von Bloh, W., Brovkin, V., Cadule, P., Doney, S., Eby, M., Fung, I., Bala, G., John, J., Jones, C., Joos, F., Kato, T., Kawamiya, M., Knorr, W., Lindsay, K., Matthews, H. D., Raddatz, T., Rayner, P., Reick, C., Roeckner, E., Schnitzler, K.-G., Schnur, R., Strassmann, K., Weaver, A. J., Yoshikawa, C., and Zeng, N.: Climate–Carbon

- Cycle Feedback Analysis: Results from the C4MIP Model Intercomparison, J. Climate, 19, 3337–3353, https://doi.org/10.1175/JCLI3800.1, 2006.
 - Fu, B., Gasser, T., Li, B., Tao, S., Ciais, P., Piao, S., Balkanski, Y., Li, W., Yin, T., Han, L., Li, X., Han, Y., An, J., Peng, S., and Xu, J.: Short-lived climate forcers have long-term climate impacts via the carbon–climate feedback, Nature Climate Change, 10, 851–855, https://doi.org/10.1038/s41558-020-0841-x, 2020.
- Fyfe, J. C., Kharin, V. V., Santer, B. D., Cole, J. N. S., and Gillett, N. P.: Significant impact of forcing uncertainty in a large ensemble of climate model simulations, Proceedings of the National Academy of Sciences, 118, e2016549118, https://doi.org/10.1073/pnas.2016549118.2021.
 - Gregory, J. M., Jones, C. D., Cadule, P., and Friedlingstein, P.: Quantifying Carbon Cycle Feedbacks, J. Climate, 22, 5232–5250, https://doi.org/10.1175/2009JCLI2949.1, 2009.
- Hogan, R. J. and Matricardi, M.: Evaluating and improving the treatment of gases in radiation schemes: the Correlated K Distribution Model Intercomparison Project (CKDMIP), Geosci. Model Dev., 13, 6501–6521, https://doi.org/10.5194/gmd 13-6501-2020, 2020.
 - Höglund-Isaksson, L., Gómez-Sanabria, A., Klimont, Z., Rafaj, P., and Schöpp, W.: Technical potentials and costs for reducing global anthropogenic methane emissions in the 2050 timeframe –results from the GAINS model, Environmental Research Communications, 2, 025004, https://doi.org/10.1088/2515-7620/ab7457, 2020.
- 600 Jackson, R. B., Abernethy, S., Canadell, J. G., Cargnello, M., Davis, S. J., Féron, S., Fuss, S., Heyer, A. J., Hong, C., and Jones, C. D.: Atmospheric methane removal: a research agenda, Philosophical Transactions of the Royal Society A, 379, 20200454, 2021.
 - Jones, A., Haywood, J. M., and Jones, C. D.: Can reducing black carbon and methane below RCP2.6 levels keep global warming below 1.5 °C?, Atmospheric Science Letters, 19, e821, https://doi.org/10.1002/asl.821, 2018.
- 605 Jones, C. D., Ciais, P., Davis, S. J., Friedlingstein, P., Gasser, T., Peters, G. P., Rogelj, J., van Vuuren, D. P., Canadell, J. G., Cowie, A., Jackson, R. B., Jonas, M., Kriegler, E., Littleton, E., Lowe, J. A., Milne, J., Shrestha, G., Smith, P., Torvanger, A., and Wiltshire, A.: Simulating the Earth system response to negative emissions, Environmental Research Letters, 11, 095012, https://doi.org/10.1088/1748-9326/11/9/095012, 2016.
- Koven, C. D., Sanderson, B. M., and Swann, A. L. S.: Much of zero emissions commitment occurs before reaching net zero emissions, Environmental Research Letters, 18, 014017, https://doi.org/10.1088/1748-9326/acab1a, 2023.
 - Lurton, T., Balkanski, Y., Bastrikov, V., Bekki, S., Bopp, L., Braconnot, P., Brockmann, P., Cadule, P., Contoux, C., Cozic, A., Cugnet, D., Dufresne, J.-L., Éthé, C., Foujols, M.-A., Ghattas, J., Hauglustaine, D., Hu, R.-M., Kageyama, M., Khodri, M., Lebas, N., Levavasseur, G., Marchand, M., Ottlé, C., Peylin, P., Sima, A., Szopa, S., Thiéblemont, R., Vuichard, N., and Boucher, O.: Implementation of the CMIP6 Forcing Data in the IPSL-CM6A-LR Model, Journal of Advances in Modeling Earth Systems, 12, e2019MS001940, https://doi.org/10.1029/2019MS001940, 2020.
 - Malley, C. S., Borgford-Parnell, N., Haeussling, S., Howard, I. C., Lefèvre, E. N., and Kuylenstierna, J. C. I.: A roadmap to achieve the global methane pledge, Environmental Research: Climate, 2, 011003, https://doi.org/10.1088/2752-5295/acb4b4, 2023.
- Melnikova, I., Boucher, O., Cadule, P., Ciais, P., Gasser, T., Quilcaille, Y., Shiogama, H., Tachiiri, K., Yokohata, T., and Tanaka, K.: Carbon cycle response to temperature overshoot beyond 2 °C an analysis of CMIP6 models, Earth's Future, 9, e2020EF001967, https://doi.org/10.1029/2020EF001967, 2021.

- Melnikova, I., Ciais, P., Boucher, O., and Tanaka, K.: Assessing carbon cycle projections from complex and simple models under SSP scenarios, Climatic Change, 176, 168, https://doi.org/10.1007/s10584-023-03639-5, 2023.
- Mengis, N. and Matthews, H. D.: Non-CO2 forcing changes will likely decrease the remaining carbon budget for 1.5 °C, npj Climate and Atmospheric Science, 3, 19, https://doi.org/10.1038/s41612-020-0123-3, 2020.
 - Montzka, S. A., Dlugokencky, E. J., and Butler, J. H.: Non-CO2 greenhouse gases and climate change, Nature, 476, 43–50, https://doi.org/10.1038/nature10322, 2011.
 - Mundra, I. and Lockley, A.: Emergent methane mitigation and removal approaches: A review, Atmospheric Environment: X, 100223, https://doi.org/10.1016/j.aeaoa.2023.100223, 2023.
- 630 Myhre, G., Highwood, E. J., Shine, K. P., and Stordal, F.: New estimates of radiative forcing due to well mixed greenhouse gases, Geophysical Research Letters, 25, 2715–2718, https://doi.org/10.1029/98GL01908, 1998.
- Myhre, G., Shindell, D., Bréon, F. M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J. F., Lee, D., and Mendoza, B.: Anthropogenic and natural radiative forcing, in: Climate change 2013: the physical science basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, vol. Chapter 8, Cambridge University Press Cambridge, 2013.
 - Myhre, G., Forster, P. M., Samset, B. H., Hodnebrog, Ø., Sillmann, J., Aalbergsjø, S. G., Andrews, T., Boucher, O., Faluvegi, G., Fläschner, D., Iversen, T., Kasoar, M., Kharin, V., Kirkevåg, A., Lamarque, J.-F., Olivić, D., Richardson, T. B., Shindell, D., Shine, K. P., Stjern, C. W., Takemura, T., Voulgarakis, A., and Zwiers, F.: PDRMIP: A Precipitation Driver and Response Model Intercomparison Project—Protocol and Preliminary Results, Bulletin of the American Meteorological Society, 98, 1185–1198, https://doi.org/10.1175/BAMS-D-16-0019.1, 2017.
 - $Nakazawa, T.: Current \ understanding \ of \ the \ global \ cycling \ of \ carbon \ dioxide, \ methane, \ and \ nitrous \ oxide, \ Proceedings \ of \ the \ Japan \ Academy, Series \ B, 96, 394-419, \ https://doi.org/10.2183/pjab.96.030, 2020.$
- Nisbet, E. G., Fisher, R. E., Lowry, D., France, J. L., Allen, G., Bakkaloglu, S., Broderick, T. J., Cain, M., Coleman, M., Fernandez, J., Forster, G., Griffiths, P. T., Iverach, C. P., Kelly, B. F. J., Manning, M. R., Nisbet-Jones, P. B. R., Pyle, J. A., Townsend-Small, A., al-Shalaan, A., Warwick, N., and Zazzeri, G.: Methane Mitigation: Methods to Reduce Emissions, on the Path to the Paris Agreement, Reviews of Geophysics, 58, e2019RG000675, https://doi.org/10.1029/2019RG000675, 2020.
 - Nordling, K., Korhonen, H., Räisänen, J., Partanen, A.-I., Samset, B. H., and Merikanto, J.: Understanding the surface temperature response and its uncertainty to CO2, CH4, black carbon, and sulfate, Atmos. Chem. Phys., 21, 14941–14958, https://doi.org/10.5194/acp-21-14941-2021, 2021.
- 650 Nzotungicimpaye, C.-M., MacIsaac, A. J., and Zickfeld, K.: Delaying methane mitigation increases the risk of breaching the 2 °C warming limit, Communications Earth & Environment, 4, 250, https://doi.org/10.1038/s43247-023-00898-z, 2023.
 - Ou, Y., Roney, C., Alsalam, J., Calvin, K., Creason, J., Edmonds, J., Fawcett, A. A., Kyle, P., Narayan, K., O'Rourke, P., Patel, P., Ragnauth, S., Smith, S. J., and McJeon, H.: Deep mitigation of CO2 and non-CO2 greenhouse gases toward 1.5 °C and 2 °C futures, Nature Communications, 12, 6245, https://doi.org/10.1038/s41467-021-26509-z, 2021.
- 655 Palazzo Corner, S., Siegert, M., Ceppi, P., Fox-Kemper, B., Frölicher, T. L., Gallego-Sala, A., Haigh, J., Hegerl, G. C., Jones, C. D., Knutti, R., Koven, C. D., MacDougall, A. H., Meinshausen, M., Nicholls, Z., Sallée, J. B., Sanderson, B. M., Séférian, R., Turetsky, M., Williams, R. G., Zaehle, S., and Rogelj, J.: The Zero Emissions Commitment and climate stabilization, Frontiers in Science, 1, https://doi.org/10.3389/fsci.2023.1170744, 2023.

- Park, S.-W., Kim, J.-S., and Kug, J.-S.: The intensification of Arctic warming as a result of CO2 physiological forcing, Nature Communications, 11, 2098, https://doi.org/10.1038/s41467-020-15924-3, 2020.
 - Pincus, R., Forster, P. M., and Stevens, B.: The Radiative Forcing Model Intercomparison Project (RFMIP): experimental protocol for CMIP6, Geosci. Model Dev., 9, 3447–3460, https://doi.org/10.5194/gmd-9-3447-2016, 2016.
 - Rao, S. and Riahi, K.: The Role of Non-CO₃ Greenhouse Gases in Climate Change Mitigation: Long-term Scenarios for the 21st Century, The Energy Journal, 27, 177–200, 2006.
- Richardson, T. B., Forster, P. M., Smith, C. J., Maycock, A. C., Wood, T., Andrews, T., Boucher, O., Faluvegi, G., Fläschner, D., Hodnebrog, Ø., Kasoar, M., Kirkevåg, A., Lamarque, J.-F., Mülmenstädt, J., Myhre, G., Olivié, D., Portmann, R. W., Samset, B. H., Shawki, D., Shindell, D., Stier, P., Takemura, T., Voulgarakis, A., and Watson-Parris, D.: Efficacy of Climate Forcings in PDRMIP Models, J Geophys Res Atmos, 124, 12824–12844, https://doi.org/10.1029/2019JD030581, 2019.
- de Richter, R., Ming, T., Davies, P., Liu, W., and Caillol, S.: Removal of non-CO2 greenhouse gases by large-scale atmospheric solar photocatalysis, Progress in Energy and Combustion Science, 60, 68–96, https://doi.org/10.1016/j.pecs.2017.01.001, 2017
 - Schwinger, J. and Tjiputra, J.: Ocean Carbon Cycle Feedbacks Under Negative Emissions, Geophysical Research Letters, 45, 5062–5070, https://doi.org/10.1029/2018GL077790, 2018.
- Schwinger, J., Tjiputra, J. F., Heinze, C., Bopp, L., Christian, J. R., Gehlen, M., Ilyina, T., Jones, C. D., Salas-Mélia, D., 675 Segschneider, J., Séférian, R., and Totterdell, I.: Nonlinearity of Ocean Carbon Cycle Feedbacks in CMIP5 Earth System Models, Journal of Climate, 27, 3869–3888, https://doi.org/10.1175/JCLI-D-13-00452.1, 2014.
 - Tanaka, K., Boucher, O., Ciais, P., Johansson, D. J. A., and Morfeldt, J.: Cost-effective implementation of the Paris Agreement using flexible greenhouse gas metrics, Sci Adv, 7, eabf9020, https://doi.org/10.1126/sciadv.abf9020, 2021.
- Thornhill, G. D., Collins, W. J., Kramer, R. J., Olivié, D., Skeie, R. B., O'Connor, F. M., Abraham, N. L., Checa-Garcia, R., Bauer, S. E., Deushi, M., Emmons, L. K., Forster, P. M., Horowitz, L. W., Johnson, B., Keeble, J., Lamarque, J.-F., Michou, M., Mills, M. J., Mulcahy, J. P., Myhre, G., Nabat, P., Naik, V., Oshima, N., Schulz, M., Smith, C. J., Takemura, T., Tilmes, S., Wu, T., Zeng, G., and Zhang, J.: Effective radiative forcing from emissions of reactive gases and aerosols a multi-model comparison, Atmos. Chem. Phys., 21, 853–874, https://doi.org/10.5194/acp-21-853-2021, 2021.
- Tokarska, K. B., Gillett, N. P., Arora, V. K., Lee, W. G., and Zickfeld, K.: The influence of non-CO2 forcings on cumulative carbon emissions budgets, Environmental Research Letters, 13, 034039, https://doi.org/10.1088/1748-9326/aaafdd, 2018.
 - World Climate Research Programme (WCRP) Coupled Model Intercomparison Project (Phase 6): https://esgf-node.llnl.gov/search/cmip6/, last access: 6 July 2022.
 - Williams, R. G., Katavouta, A., and Goodwin, P.: Carbon-Cycle Feedbacks Operating in the Climate System, Current Climate Change Reports, 5, 282–295, https://doi.org/10.1007/s40641-019-00144-9, 2019.
- 690 Zhu, X., Zhuang, Q., Gao, X., Sokolov, A., and Schlosser, C. A.: Pan-Arctic land–atmospheric fluxes of methane and carbon dioxide in response to climate change over the 21st century, Environmental Research Letters, 8, 045003, https://doi.org/10.1088/1748-9326/8/4/045003, 2013.
 - Zickfeld, K., Eby, M., Matthews, H. D., Schmittner, A., and Weaver, A. J.: Nonlinearity of Carbon Cycle Feedbacks, Journal of Climate, 24, 4255–4275, https://doi.org/10.1175/2011JCLI3898.1, 2011.

Zickfeld, K., MacDougall, A. H., and Matthews, H. D.: On the proportionality between global temperature change and cumulative CO 2 emissions during periods of net negative CO 2 emissions, Environ. Res. Lett., 11, 055006, https://doi.org/10.1088/1748-9326/11/5/055006, 2016.