

# Atmospheric Lifetime of Fossil Fuel Carbon Dioxide

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## Key Words

climate, warming, ocean chemistry, carbon cycle

## Abstract

CO<sub>2</sub> released from combustion of fossil fuels equilibrates among the various carbon reservoirs of the atmosphere, the ocean, and the terrestrial biosphere on timescales of a few centuries. However, a sizeable fraction of the CO<sub>2</sub> remains in the atmosphere, awaiting a return to the solid earth by much slower weathering processes and deposition of CaCO<sub>3</sub>. Common measures of the atmospheric lifetime of CO<sub>2</sub>, including the e-folding time scale, disregard the long tail. Its neglect in the calculation of global warming potentials leads many to underestimate the longevity of anthropogenic global warming. Here, we review the past literature on the atmospheric lifetime of fossil fuel CO<sub>2</sub> and its impact on climate, and we present initial results from a model intercomparison project on this topic. The models agree that 20–35% of the CO<sub>2</sub> remains in the atmosphere after equilibration with the ocean (2–20 centuries). Neutralization by CaCO<sub>3</sub> draws the airborne fraction down further on timescales of 3 to 7 kyr.

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**Lifetime:** the mean persistence time of a perturbation to the CO<sub>2</sub> concentration of the atmosphere

**Linear kinetics:** the rate of a chemical reaction depends linearly on the concentration of the reactant. Radioactive decay is an example

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## INTRODUCTION

The fate and lifetime of fossil fuel CO<sub>2</sub> released to the atmosphere is not inherently scientifically controversial, but the packaging of this information for public consumption is strewn with such confusion that Pieter Tans proposed in print that the entire concept should be banished (Tans et al. 1990). How long is global warming from CO<sub>2</sub> going to last, policymakers and the public would like to know. If there is a trade-off possible between emissions of CO<sub>2</sub> versus emissions of other greenhouse gases, how shall they be compared? The lifetimes of greenhouse gases are incorporated into the construction of global warming potentials, the time-integrated climate impact of each gas relative to CO<sub>2</sub>.

The question of the atmospheric CO<sub>2</sub> lifetime is also important for predicting the impact of human activity on slowly responding aspects of the climate system, such as the major ice sheets (Archer & Brovkin 2008), permafrost (Gavrilov et al. 2003, Lawrence & Slater 2005), and geochemical impacts of changing the temperature of the deep sea (Archer et al. 2004). Many of the most profound changes in the Earth's surface, such as sea level, take place on timescales of thousands of years and longer.

There is a strong consensus across models of global carbon cycling, as exemplified by the ones presented here, that the climate perturbations from fossil fuel–CO<sub>2</sub> release extend hundreds of thousands of years into the future. This is consistent with sedimentary records from the deep past, in particular a climate event known as the Paleocene-Eocene thermal maximum, which consisted of a relatively sharp increase in atmospheric CO<sub>2</sub> and ocean temperature, followed by a recovery, which took perhaps 150,000 years (Kennett & Stott 1991, Pagani et al. 2006) (see also The Paleocene-Eocene Thermal Maximum Climate Event sidebar).

The gulf between the widespread preconception of a relatively short (hundred-year) lifetime of CO<sub>2</sub> on the one hand and the evidence of a much longer climate impact of CO<sub>2</sub> on the other arguably has its origins in semantics. There are rival definitions of a lifetime for anthropogenic CO<sub>2</sub>. One is the average amount of time that individual carbon atoms spend in the atmosphere before they are removed, by uptake into the ocean or the terrestrial biosphere. Another is the amount of time it takes until the CO<sub>2</sub> concentration in the air recovers substantially toward its original concentration. The difference between the two definitions is that exchange of carbon between the atmosphere and other reservoirs affects the first definition, by removing specific CO<sub>2</sub> molecules, but not the second because exchange does not result in net CO<sub>2</sub> drawdown. The misinterpretation that has plagued the question of the atmospheric lifetime of CO<sub>2</sub> seems to arise from confusion of these two very different definitions.

In the simplest of worlds, the decay of a perturbation might follow first-order linear kinetics. Radioactive decay is an example of this. The resulting concentration trajectory through time

### THE PALEOCENE-EOCENE THERMAL MAXIMUM CLIMATE EVENT

Sediment cores from the deep ocean reveal a climate event 55 million years ago that appears to be analogous to the potential global warming climate event in the future. Isotopes of carbon preserved in CaCO<sub>3</sub> shells reveal an abrupt release of carbon to the atmosphere–ocean system, which took about 150 thousand years to recover. Isotopes of oxygen show a parallel perturbation, reflecting warming of the climate and the deep ocean in response to the carbon release. Although specifics of the event remain uncertain, such as the source, amount, and release timescale of the CO<sub>2</sub>, the event confirms the long timescale for recovery from CO<sub>2</sub> release, as predicted by the models in this study.

**Table 1 Carbon sink reactions**

Seawater buffer	$\text{CO}_2 + \text{CO}_3^{=} + \text{H}_2\text{O} \leftrightarrow 2 \text{HCO}_3^-$ (dissolved in the ocean)
CaCO <sub>3</sub> neutralization	$\text{CO}_2 + \text{CaCO}_3 + \text{H}_2\text{O} \rightarrow 2 \text{HCO}_3^-$ (ocean)
Silicate weathering	$\text{CO}_2 + \text{CaSiO}_3 \rightarrow \text{CaCO}_3 + \text{SiO}_2$ (ocean sediments)

will follow an exponential decay function. In this case, the mean lifetime of the carbon atoms in the atmosphere is closely approximated by the most-of-it-goes-away criterion; the mean lifetime is, in fact, equal to the *e*-folding timescale, the time at which only 1/*e*, ~37%, of the original pulse remains. If fossil fuel CO<sub>2</sub> in the atmosphere was expected to diminish according to linear kinetics, then it would be possible to calculate the lifetime simply using the present-day excess CO<sub>2</sub> concentration in the atmosphere (~100 ppm or 200 Pg C) and the natural uptake rate, currently ~2 Pg C yr<sup>-1</sup> (petagrams of carbon per year) each into the oceans and into the land biosphere. Dividing the inventory by the flux yields an apparent lifetime of 50 to 100 years, depending on whether the terrestrial uptake is counted in addition to the oceanic uptake. This type of calculation has been most recently presented by Jacobson (2005), who determined an atmospheric lifetime of 30 to 95 years. For the nonlinear CO<sub>2</sub> uptake kinetics, as predicted by carbon cycle models, however, this apparent lifetime would increase with time after the CO<sub>2</sub> is released. Some CO<sub>2</sub> from the release would remain in the atmosphere thousands of years into the future, and the atmospheric lifetime of the CO<sub>2</sub>, calculated at that time, would be thousands of years.

The most fundamental problem with the linear-kinetics formulation is that even the simplest atmosphere/ocean carbon cycle models relax to a different atmospheric CO<sub>2</sub> level, higher than the concentration before the spike was released. If the ocean and atmosphere were held in a nonreactive container, that excess CO<sub>2</sub> would remain in the atmosphere forever, an eternal airborne fraction of the CO<sub>2</sub> slug of 15 to 40% (Archer 2005). If the ocean were infinitely large or caustic (high pH), the airborne fraction might approach zero, but given the size and chemistry of the ocean, an airborne fraction of tens of a percent can be calculated from simple thermodynamics, as if the ocean were a bucket of homogeneous seawater in equilibrium with about two bucketfuls of a gas phase.

In the real world, the leftover CO<sub>2</sub> in the atmosphere after ocean invasion interacts with the land biosphere and is taken up by pH-neutralization reactions with calcium carbonate (CaCO<sub>3</sub>) and the CaO component of igneous rocks (**Table 1**). The timescales for these processes range from thousands to hundreds of thousands of years. Keeling & Bacastow (1977) predicted that it would take at least 10,000 years for atmospheric CO<sub>2</sub> to return to preindustrial levels. Walker & Kasting (1992) reached a similar conclusion but extended the duration of the long tail to hundreds of thousands of years. Broecker & Takahashi (1978) described the neutralization reaction with CaCO<sub>3</sub>. Many other carbon cycle models of a variety of configurations and resolutions have found the same result (Sundquist 1990, Caldeira & Kasting 1993, Archer 2005, Lenton & Britton 2006, Montenegro et al. 2007, Ridgwell & Hargreaves 2007, Tyrrell et al. 2007). The mean lifetime of the elevated CO<sub>2</sub> concentration of the atmosphere resulting from fossil fuel combustion has been calculated to be tens of thousands of years (Archer et al. 1997), not at all similar to the 50- to 100-year lifetime calculated using the linear approximation based on fluxes immediately following a release of CO<sub>2</sub> to the atmosphere. Clearly, the linear approximation, using a single characteristic timescale for the removal of CO<sub>2</sub> from the atmosphere, is a poor representation of the way the carbon cycle works. An analogy can be drawn with radioactive waste, for which the decay of its radioactivity as a whole will not conform to any single exponential decay curve because it is composed of a variety of different radionuclides with a wide range of half-lives.

As the ocean is acidified, its ability to hold more CO<sub>2</sub> decreases, so that the airborne fraction of a kilogram of CO<sub>2</sub> is higher if the water has already absorbed a substantial amount of new CO<sub>2</sub>.

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**E-folding timescale:**

the amount of time it would take for a concentration to decrease to a proportion of  $e^{-1}$ , ~37%, of its original

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The change in buffering provided by seawater can be readily calculated and is encapsulated in the Revelle factor (Zeebe & Wolf-Gladrow 2001). For a system in which CO<sub>2</sub> repartitions only between ocean and atmosphere, an analytical expression can be derived for the CO<sub>2</sub> concentration in the air as a function of time (Goodwin et al. 2007). To include ocean-sediment interactions, Kheshgi & Archer (2004) developed an analytical Greens function-type approximation to the results of an ocean and sediment carbon cycle model. These formulations had an explicit accounting of the change in carbon-buffer chemistry with ocean acidification. The buffering capacity and pH of the ocean are ultimately restored by dissolution of CaCO<sub>3</sub> (Broecker & Takahashi 1978, Ridgwell & Zeebe 2005). Even for linear uptake kinetics, the decay timescale from an impulse CO<sub>2</sub> release is different from the decay timescale after ongoing emissions are stopped (Caldeira & Kasting 1993, Gaffin et al. 1995); in other words, the decay time for an additional kilogram of CO<sub>2</sub> depends on the state of the system into which that CO<sub>2</sub> is released.

As more and more CO<sub>2</sub> is emitted, the airborne fraction increases because of the depleted carbon-buffer chemistry of the ocean, while the radiative impact of a further kilogram added to the air decreases, because of the absorption-band saturation effect. Caldeira & Kasting (1992) find that these two effects largely counteract each other, so that the radiative impact of a kilogram of CO<sub>2</sub> is nearly independent of whether that kilogram is released early or late in the fossil fuel era. Eby et al. (2009) find that the logarithmic dependence of radiative forcing on CO<sub>2</sub> concentration acts to extend the mean lifetime of the radiative forcing perturbation to a time scale that is longer than the recovery timescale of the atmospheric CO<sub>2</sub> concentration.

The extent and longevity of the climate impact from CO<sub>2</sub> release will also depend on transient uptake by the terrestrial biosphere, which takes up 2 Pg C year<sup>-1</sup> today and shortens the apparent lifetime of CO<sub>2</sub>, but which could become saturated in the coming decades, leaving pCO<sub>2</sub> to follow the slower uptake kinetics of the ocean (Moore & Braswell 1994). If the terrestrial biosphere, including soil carbon, turned into a new source of CO<sub>2</sub> to the atmosphere at some point in the future (Friedlingstein et al. 2006), then it would act to prolong the apparent lifetime of CO<sub>2</sub>. In addition, high plant primary productivity in a high-CO<sub>2</sub> world may also act to enhance the rate of weathering of soil minerals and bedrock, leading to an acceleration of CO<sub>2</sub> uptake by silicate weathering, the slowest and ultimate sink for fossil fuel CO<sub>2</sub> (Lenton & Britton 2006).

Atmospheric lifetimes of greenhouse gases are considered and compared using time-integrated measures of climate impact called global warming potentials (GWPs) (see Global Warming Potential sidebar). The long tail of the CO<sub>2</sub> lifetime is a problem for calculating the global warming potentials of all gases, because CO<sub>2</sub> is taken to be the reference case against which other gases are compared. Most of the linear-kinetics studies (Caldeira & Kasting 1993, Moore & Braswell 1994, Gaffin 1997) explicitly acknowledge that they are not considering the long lifetime of the CO<sub>2</sub> that remains in the atmosphere after initial equilibration with the ocean. This initial relaxation timescale is a useful and helpful quantity to measure, but it is not by itself appropriate for calculating the GWP because it doesn't represent the entire climate impact of CO<sub>2</sub> release.

## GLOBAL WARMING POTENTIAL

Global warming potential (GWP) is an estimate of the climate impact of greenhouse gas release, which accounts for the longevity of the gas in the atmosphere as well as its instantaneous impact on the radiative energy balance of the Earth. Evaluated on a time horizon of 20 years, the GWP of methane is 73, meaning that a kilogram of methane produces 73 times the radiative forcing of a kilogram of CO<sub>2</sub>. If the gases are compared over a longer time horizon of 100 years, the GWP of methane decreases to 23, because the CO<sub>2</sub> persists through the century.

In practice, the tail is generally thrown out of GWP calculations by truncating the integral at 100 years, a timescale that we argue arises from our own lifetimes rather than anything intrinsic about the carbon cycle.

The 1990 Intergovernmental Panel on Climate Change (IPCC) report included in its “Policymaker Summary” a table showing the properties of various greenhouse gases, including an atmospheric lifetime of CO<sub>2</sub> listed as 50 to 200 years, with a footnote caveat that “the way in which CO<sub>2</sub> is absorbed in the ocean and biosphere is not simple and a single value cannot be given . . .” (Houghton et al. 1990). It was carefully explained in the technical Chapter 1, “Greenhouse Gases and Aerosols,” that, on human timescales, CO<sub>2</sub> really has no sinks, it just equilibrates among the atmosphere, ocean, and biosphere, with some residual remaining in the atmosphere after the equilibration is complete. The timescale of 50 to 200 years was based on the responses of a pair of ocean carbon cycle–models to a pulse CO<sub>2</sub> input (Maier-Reimer & Hasselmann 1987, Siegenthaler & Oeschger 1987). This 50–200 years represents a timescale for equilibration with the ocean, a process that leaves a significant fraction of CO<sub>2</sub> in the atmosphere. Because of the residual fraction in the air after equilibration, the equilibration timescale is no indication of how long the climate impacts of CO<sub>2</sub> release will last. But this distinction was confused in the “Policymaker Summary,” in which the timescale is referred to as an atmospheric lifetime, which is incorrect, and it is used as a rationale to limit the time horizon for calculating global warming potentials of greenhouse gases to 100 years, which is not logical either. The footnote to the table quoted above explains why a range of lifetimes is given rather than a single value, as for methane and other gases, but gives no indication of the new equilibrium or the long tail. On this question, Gaffin (1997, p. 494) wrote that “the door to misinterpretation had been left open. Others have and continue to walk through it.”

Subsequent IPCC reports in 1995 and 2001 compounded the mistake, revising the lower limit of the lifetime estimate down to only five years. Presumably, the lower end of the range was a reflection of CO<sub>2</sub> exchange with the atmosphere or the terrestrial biosphere, although short-term uptake and release, or exchange of one carbon for another, has no impact on the altered climate. Finally, the 2007 IPCC report removed the table from the “Policymaker Summary,” and added in the “Executive Summary” of Chapter 7 on the carbon cycle, “About half of a CO<sub>2</sub> pulse to the atmosphere is removed over a timescale of 30 years; a further 30% is removed within a few centuries; and the remaining 20% will typically stay in the atmosphere for many thousands of years” (Denman et al 2007, page 501).

To summarize, with few exceptions (Jacobson 2005), there has been no disagreement within the carbon cycle scientific community about the long tail to the fossil fuel CO<sub>2</sub> lifetime. But there has been considerable divergence on the way in which the behavior of that carbon pulse should be expressed and evaluated. The result has been an erroneous conclusion, throughout much of the popular treatment of the issue of climate change, that global warming will be a century-timescale phenomenon. Simple thermodynamics of CO<sub>2</sub> dissolved in seawater plus paleo-evidence, in particular the PETM event 55 Mya tell us otherwise.

## A MODEL INTERCOMPARISON EXPERIMENT

We have assessed the range of possibilities for the long-term fate of fossil fuel CO<sub>2</sub> in the atmosphere, ocean, and terrestrial biosphere by means of a model intercomparison experiment that we call LTMIP (Long Tail Model Intercomparison Project). Model intercomparisons have proven extremely useful in climate dynamics (such as the AR4 climate forecast simulations) and paleoclimate modeling (PMIP), (Weber et al. 2007), and this project takes as its template the intercomparison ocean carbon and nutrient cycling models called OCMIP (Orr 1999). In the intercomparison

results we present here, we limit the timescale of our model simulations to 10,000 years, which is arguably not as long as we would like but is still too long for primitive-equation atmosphere circulation codes. Hence, most of the models in our group are streamlined in some computational way.

## Models

CLIMBER-2 consists of a dynamical-statistical two-dimensional (2D) atmosphere coupled to a 2D three-basin dynamic ocean, a terrestrial biosphere model (VECODE), an oceanic biogeochemistry model, and a phosphate-limited model for marine biota (Ganopolski et al. 1998; Brovkin et al. 2002, 2007). The sediment model resolves the diffusive pore-water dynamics, assuming oxic-only respiration and 4.5-order  $\text{CaCO}_3$  dissolution kinetics (Archer 1996, Brovkin et al. 2007). Weathering rates scale to runoff on the land surface grid cells, with separate carbonate and silicate lithological classes.

CC-SED was described by (Archer 2005) and uses the HAMOCC2 stationary annual mean flow field code to advect geochemical tracers. The sediment model is the same as in CLIMBER (Archer 1996). The temperature of the ocean is offset uniformly with a 1000-year response time, relaxing to a target temperature determined by a deep-ocean climate sensitivity of  $3^\circ\text{C}$ . A weathering feedback has been added to the code because its description (Archer 2005), scaled to  $\text{CO}_2$ , according to Berner & Kothavala (2001), with some of the parameters as described by Berner (2004).

GENIE8 (Ridgwell & Hargreaves 2007) consists of a three-dimensional (3D) noneddy-resolving frictional geostrophic-ocean-circulation model and 2D sea-ice and energy-moisture balance atmospheric models (Edwards & Marsh 2005). GENIE8 incorporates a representation of the marine geochemical cycling of carbon and other biologically mediated tracers and a representation of sedimentary stratigraphy and preservation of carbonates in deep-sea sediments (Ridgwell et al. 2007). The sediment diagenesis zone at each ocean grid point is treated as a single (1 cm) box and is coupled to an accumulating column below.  $\text{CaCO}_3$  dissolution in the diagenesis zone is calculated based on the model of Archer (1996) and modified as described in Ridgwell (2007). Carbonate and silicate weathering rates are parameterized as a function of mean global surface air temperature over land, following Berner & Kothavala (2001) and Lenton & Britton (2006) and are initially equal in magnitude. A fixed  $\text{CO}_2$  out-gassing rate balances the baseline silicate weathering rate.

GENIE16 is similar to GENIE8, except it has 16 rather than 8 depth levels in the ocean and is seasonally insolation forced. The climate-model component is described in Singarayer et al. (2008).

GEOCYC is an on-line zero-dimensional descendent of the Berner & Kothavala (2001) GEOCARB III model, with the additions of (a) time-dependent equilibration of the atmosphere and a homogeneous ocean that uses a time constant of  $1 \text{ Pg C yr}^{-1}$  per 300 ppm difference in surface ocean  $\text{pCO}_2$  and (b) time-resolved  $\text{CaCO}_3$  neutralization, where the burial rate of  $\text{CaCO}_3$  is a linear function of ocean  $\text{CO}_3^{=}$  concentration that uses a dependence of  $3 \text{ E-3 Pg C yr}^{-1}$  for every micromolar change in ocean  $\text{CO}_3^{=}$  concentration, which comes from the model results of Archer et al. (1997). GEOCARB III was designed for educational purposes and can be run interactively on the web at <http://understandingtheforecast.org/Projects/geocarb.html>.

LTCM (Cao et al. 2008) is a derivative of the HILDA (Shaffer & Sarmiento 1995) box advection/diffusion model, with the inclusion of an OCMIP-type geochemical component. The sediment model is of similar formulation but of different origin from that used in HAMOCC and CLIMBER. Weathering rates interact with climate, following Berner & Kothavala (2001).

MESMO (Matsumoto et al. 2008) is based on GENIE-1. In the dynamical ocean model, there are 16 vertical levels on a  $36 \times 36$  equal-area horizontal grid. The model also uses an

energy-moisture balance model of the atmosphere and a dynamic-thermodynamic model of sea ice. Ocean production occurs in the two layers within the top 100m and is dependent on, among other factors, the diagnosed mixed-layer depth.

MPI-UW (Mikolajewicz et al. 2007) consists of a coupled coarse-resolution atmospheric general-circulation model, ECHAM3 (Roeckner et al. 1992), an updated version of the large-scale geostrophic-ocean model (LSG) (Maier-Reimer et al. 1993), which drives HAMOCC3 ocean biogeochemistry (Winguth et al. 1994). The land biosphere is simulated using the dynamic vegetation model LPJ (Sitch et al. 2003). To reduce the amount of computer time required, the model has been run in periodically synchronous mode (Mikolajewicz et al. 2007) for the periods without rapid changes in atmospheric CO<sub>2</sub> concentration. Ensemble simulations have been performed for some experiments to average out the effect of natural variability. The simulations have been initialized with different states (1000 years apart) of the control simulation.

UVIC2.8 is the University of Victoria Earth System Climate Model, consisting of a vertically integrated, energy/moisture balance, atmospheric model with dynamic feedbacks, which is coupled to the MOM2 ocean general-circulation model and a dynamic/thermodynamic sea-ice model (Weaver et al. 2001). The horizontal resolution is  $1.8 \times 3.6^\circ$ , and the ocean model has 19 vertical levels. The terrestrial carbon model is a modified version of the MOSES2 land surface model and the TRIFFID dynamic vegetation model (Meissner et al. 2003). Ocean carbon is simulated by means of an OCMIP-type inorganic carbon cycle model and a marine ecosystem model solving prognostic equations for nutrients, phytoplankton, zooplankton, and detritus (Schmittner et al. 2008). Sediment processes are represented using an oxic-only model of sediment respiration (Archer 1996). Isopycnal mixing and flux-corrected transport were used in the ocean model, with diapycnal diffusion specified as a horizontally constant profile. The sea-ice model is a simple, single-layer, thermodynamic version with elastic-viscous-plastic dynamics.

## Methodology

The heart of the intercomparison is a series of fossil fuel-neutralization experiments. The CO<sub>2</sub> release is instantaneous, to simplify the analysis of the resulting atmospheric CO<sub>2</sub> trajectory. Pulses of 1000 and 5000 Pg C are considered, which are round numbers that have been used in the past. For comparison, humankind has already released ~300 Pg C and will surpass 1000 Pg C total release under business-as-usual projections before the end of the century. The entire reservoir of fossil fuel, predominantly coal, totals ~5000 Pg C. Thus, 1000 and 5000 Pg C can be considered moderate (that is to say, at least somewhat moderated by concern about climate change) and large (burning most of the available carbon).

The base case calls for invasion into the ocean with no feedbacks or sediments. This provides a baseline against which the impacts of various feedbacks can be evaluated, and it could be regarded as a control experiment. The chemistry of the ocean responds to the CO<sub>2</sub> invasion, limiting the amount of the released CO<sub>2</sub> slug that the ocean ultimately holds. The CO<sub>2</sub> drawdown trajectories tend to follow quasi-exponential decay functions toward new long-term values within the first millennium of the simulations, representing equilibration between the ocean and the sediments (but not with the minerals). This response time and final state are well resolved by our simulations. We also drove the models with the historical rate of atmospheric CO<sub>2</sub> rise (designated Hist), to obtain a snapshot of CO<sub>2</sub> invasion pathway and to compare the present-day uptake rate of fossil fuel CO<sub>2</sub> with observations.

When possible in each model, we included the impacts of various feedbacks to the CO<sub>2</sub> system, including feedbacks from climate, from sediment dissolution, from enhanced weathering, and from uptake by vegetation and release by soils. The feedbacks were added cumulatively in that order,

and the impact of each feedback is gauged as the difference in the drawdown of atmospheric CO<sub>2</sub> that results. Our definitions of feedback strength are thus experiment-dependent; we might have gotten different answers if we had layered the feedbacks in a different order. The order we chose to some extent reflects the relative certainties of the feedbacks in addition to their expected magnitudes. Not all of the models in our pool have the capacity to do all of the simulations.

First, the climate feedback is added in the C case. The chief impact of changing climate is via the solubility of CO<sub>2</sub> gas in seawater, which decreases with warming. There may also be changes in ocean circulation or ventilation patterns and rates that impact CO<sub>2</sub> invasion.

Sediment feedbacks are neglected in the base case, but they are added in S and CS. The sediment response is composed of two components: a fast pH-neutralizing reaction of CO<sub>2</sub> with CaCO<sub>3</sub> on the sea floor (a process called chemical erosion), followed by a longer timescale reaction of CO<sub>2</sub> with carbonates on land (weathering) (Archer et al. 1997, Ridgwell & Hargreaves 2007). The amount of CaCO<sub>3</sub> on the sea floor available to dissolve is limited by the formation of a clay layer on the sea floor, the nonreactive material remaining after CaCO<sub>3</sub> dissolves, which impedes further CaCO<sub>3</sub> dissolution (Broecker & Takahashi 1978, Archer et al. 1997). The 5000 Pg C CO<sub>2</sub> release is close to enough to deplete the available CaCO<sub>3</sub> stock of the ocean, leaving excess weathering over CaCO<sub>3</sub> burial to drive the CO<sub>2</sub> neutralization.

A weathering feedback W is included in the CSW case, reflecting a climate impact on the rate of chemical weathering on land, which is driven by excess runoff in a wetter, high-CO<sub>2</sub> world in addition to faster rates of dissolution in a warmer, more acidic environment (Berner et al. 1983). The weathering feedback pertains to CaCO<sub>3</sub> dissolution in addition to the dissolution of igneous rocks that ultimately controls atmospheric CO<sub>2</sub>, i.e., the silicate weathering thermostat with its CO<sub>2</sub>-regulating time-constant of hundreds of thousands of years (Walker et al. 1981).

Vegetation feedbacks are indicated by the V in the simulation CSWV. The impact of vegetation is to absorb carbon into biomass and soil organic matter in response to changes in atmospheric CO<sub>2</sub> concentration or climate.

## RESULTS

In the first millennium after the instantaneous CO<sub>2</sub> release, the trajectory of atmospheric CO<sub>2</sub> is dominated by CO<sub>2</sub> dissolution into the ocean and transport to depth. **Figure 1** illustrates this for the ocean-only base case, together with the climate and sediment feedbacks. The equilibration time scale for ocean invasion, calculated by a least-squares fit of an exponential to the CO<sub>2</sub> concentration trajectory, is about  $\sim 250 \pm 90$  years for the 1000 Pg C release spike, and  $\sim 450 \pm 200$  years for the 5000 Pg C release (**Figure 2a**). These equilibration times are conceptually the same as what IPCC called the “atmospheric lifetime,” of 50–200 years, based on the model response to an instantaneous doubling of atmospheric CO<sub>2</sub> (590 Pg C). Our results show that the equilibration timescale depends on the magnitude of the CO<sub>2</sub> slug and is longer for larger slugs. The climate feedback (**Figure 2**, *gray bars*) increases the diagnosed equilibration time in

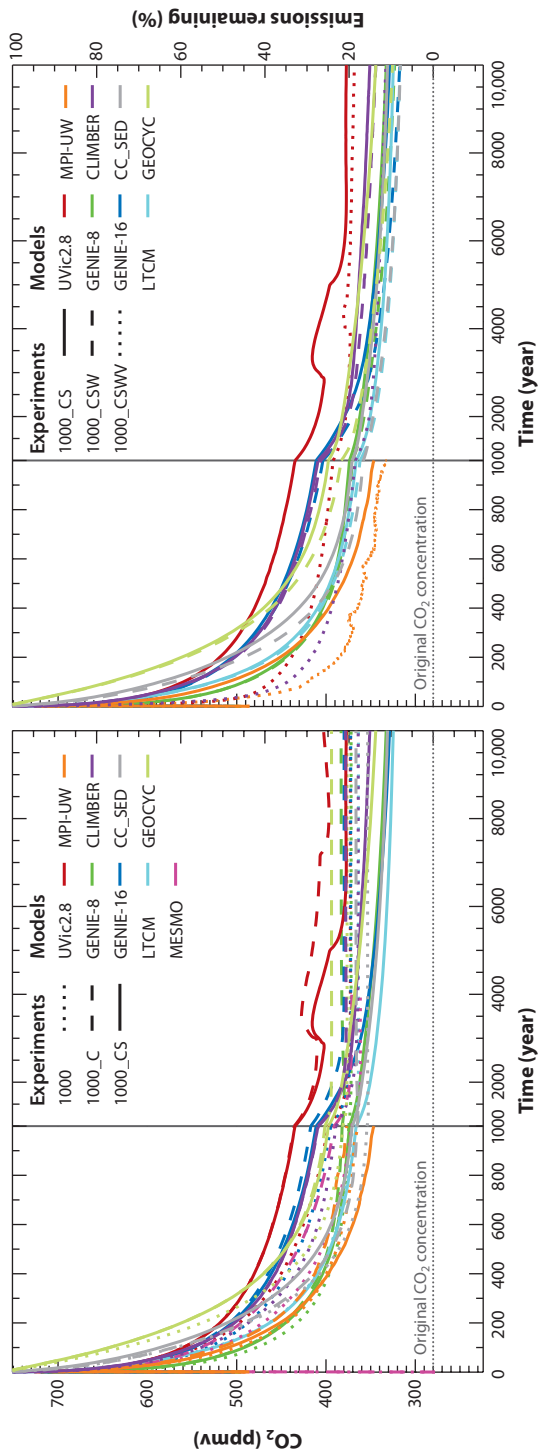
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### Figure 1

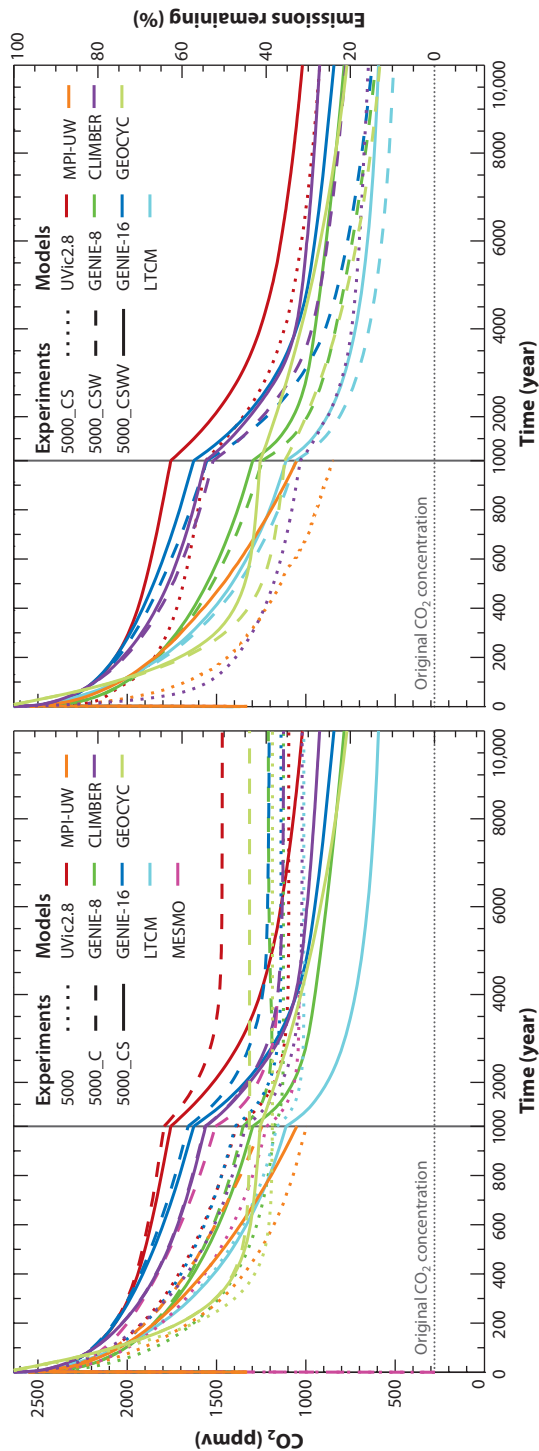
Atmospheric CO<sub>2</sub> trajectories for the 10,000-year duration of the model simulations. Note the split scale that expands the action in the first 1000 years. Colors denote models and line styles denote experiments; the same line style may be used for different experiments in different panels. Experiments for each pulse have been separated into two panels for clarity and the results represented by the solid lines are repeated for reference. Different line types designate the cases of base (ocean only) simulation, the climate feedback (C), the climate plus sediment feedback (CS), the climate plus sediment plus weathering feedback (CSW), and the climate plus sediment plus weathering plus vegetation feedback (CSWV).

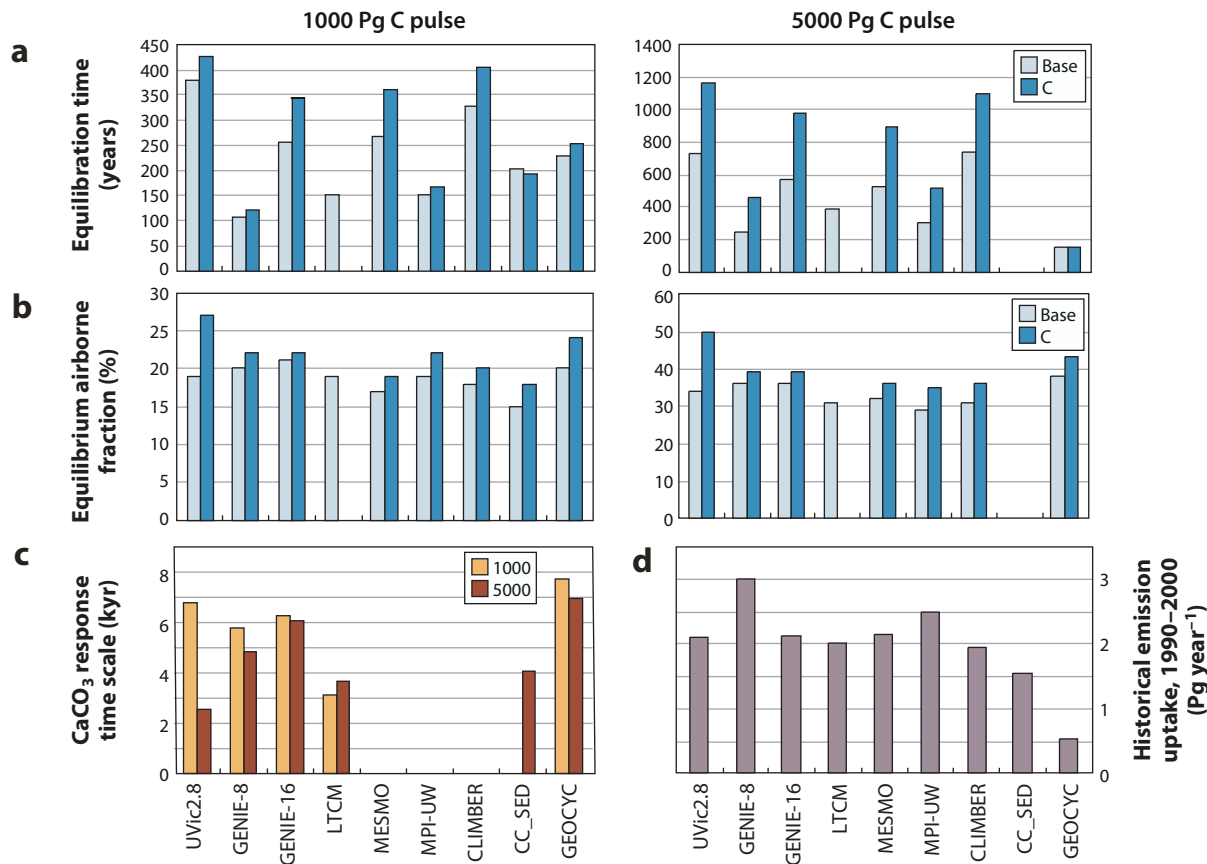


### Atmospheric CO<sub>2</sub> for a 1000 Pg pulse



### Atmospheric CO<sub>2</sub> for a 5000 Pg pulse





**Figure 2**

(a) Equilibration time for CO<sub>2</sub> invasion into the ocean. (b) Equilibrium airborne fraction of CO<sub>2</sub> after ocean invasion, for cases with no sediment interaction. (c) CaCO<sub>3</sub> dissolution response time. (d) Predicted present-day CO<sub>2</sub> uptake rates when the models are subjected to historical atmospheric CO<sub>2</sub>-concentration trajectories. The real ocean took up ~2.2 Pg C year<sup>-1</sup> between the comparable time interval 1990–2000.

the 5000 Pg C release cases but had mixed impact on the 1000 Pg C model runs. In general, there is less model agreement for the base case when the emission slug is large (5000 Pg C) than when it is small (1000 Pg C). The impact of the climate sensitivity, however, is more consistent between models when the emission slug is large.

One check on the predicted rate of CO<sub>2</sub> uptake in the future is to compare the historical forcing (Hist) model runs with the present-day CO<sub>2</sub> uptake rate of ~2.2 Pg C year<sup>-1</sup> observations (Denman et al. 2007). These are presented in **Figure 2d**, showing reasonable values for all models except GEOCYC, which assumes a homogeneous ocean and therefore misses nonlinearity in CO<sub>2</sub> uptake into a more realistic ocean.

The final equilibrium airborne fraction of the released CO<sub>2</sub> is shown for the ocean-only base model and for the climate-feedback C-model runs in **Figure 2b**. Without the climate feedback, the 1000 Pg C release experiments asymptote to an airborne fraction of ~20%, whereas the 5000 Pg C experiments equilibrate with ~35% of the CO<sub>2</sub> released remaining in the atmosphere. For large CO<sub>2</sub> release of 5000 Pg C, the airborne fraction is therefore considerably higher than the 20% claimed in the 2007 IPCC “Policymaker Summary.”

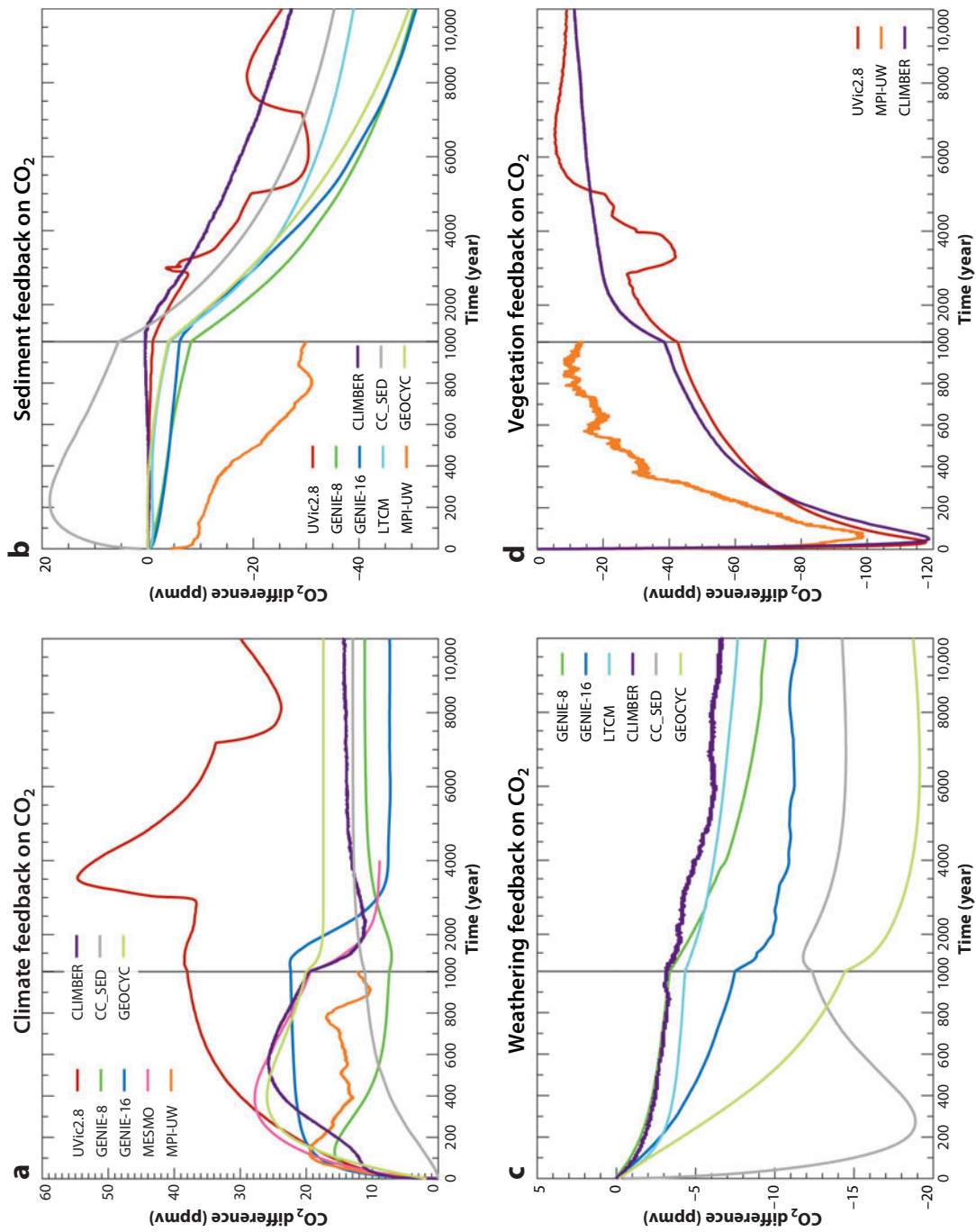
The magnitude of the climate feedback is presented, in **Figures 3a** and **4a**, as the difference in atmospheric CO<sub>2</sub> concentration with the feedback and without it. The climate feedback generally diminishes after the atmosphere equilibrates with the ocean in the first millennium, but persists at a lower level for the entire 10,000-year duration of the runs. The magnitude of the climate feedback varies greatly, with the UVic2.8 model showing by far the largest feedback. Much of this appears to be due to the complex ocean biological response in this model (Schmidtner et al. 2008), with as much as half of the climate feedback attributed to changes in ocean biology (results not shown). The UVic2.8 model also shows abrupt changes in CO<sub>2</sub> for the 1000 Pg C experiment. Abrupt warming and the accompanying increase in CO<sub>2</sub> is caused by flushing events in the Southern Ocean, which, in this model, are shown to be dependent on the level of atmospheric CO<sub>2</sub> (Meissner et al. 2008).

The sediment feedback is primarily driven by excess dissolution of CaCO<sub>3</sub> on the timescales of these model simulations, although imbalance between the dissolution (weathering) of CaCO<sub>3</sub> on land and burial in marine sediments is equally important on slightly longer timescales (Lenton & Britton 2006, Ridgwell & Hargreaves 2007). The impact of the sediment feedback is generally small over the first thousand years, but grows over the 10,000 years of the simulations to result in more drawdown to the equilibrium state than the reduction of drawdown we saw from the climate feedback (**Figures 3b** and **4b**). The CO<sub>2</sub> drawdown from CaCO<sub>3</sub> dissolution follows a trajectory that is similar to an exponential decay, with time constants derived by regression shown in **Figure 2c**. The Lenton and Britton model (2006) had a much faster neutralization response, just 500 years, but none of our models reproduces this. Other previous studies (Archer et al. 1997, Ridgwell & Hargreaves 2007, Tyrrell et al. 2007) predicted CaCO<sub>3</sub> neutralization timescales of millennia and longer, consistent with our results. It should be noted that because of the computational expense of some of the LTMIP ensemble models, the experiments presented here do not reach the steady state where CaCO<sub>3</sub> burial balances weathering, and so the runs are not ideal for determining the equilibrium airborne fraction in the steady state.

The weathering feedback, an acceleration of the dissolution on land of CaCO<sub>3</sub> and calcium-bearing silicate minerals resulting from the warmer climate (Berner et al. 1983, Ridgwell & Zeebe 2005), generally increases its impact on atmospheric CO<sub>2</sub> with time in our model runs, although the CC\_SED model shows a strong peak in CO<sub>2</sub> drawdown in the first millennium, which is when atmospheric CO<sub>2</sub> is highest. By the end of the simulations, the weathering feedback has decreased atmospheric CO<sub>2</sub> by approximately half as much as the sediment feedback alone. The global CaCO<sub>3</sub> burial rates are plotted in **Figure 5**, showing considerable disparity in the model results.

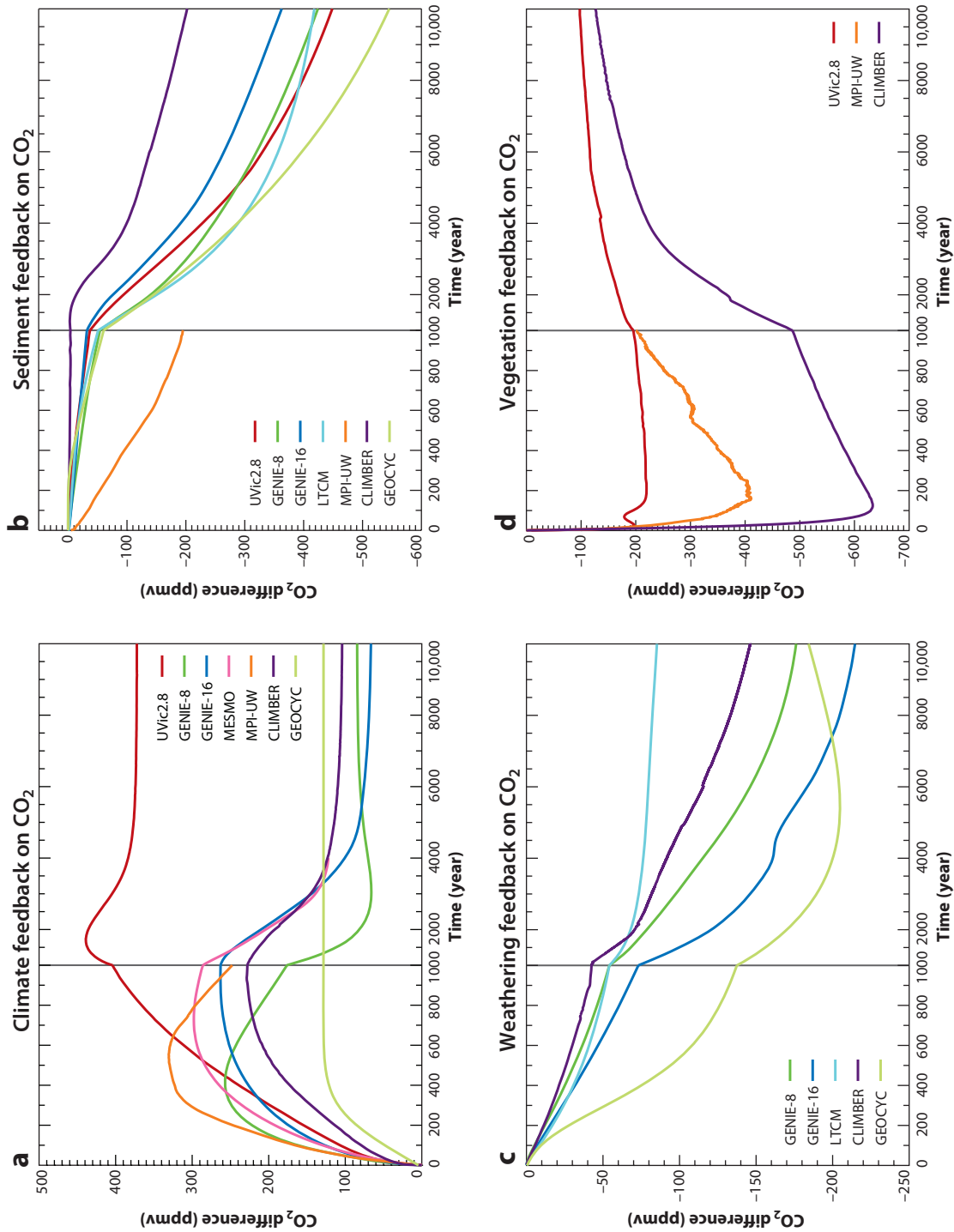
The vegetation feedback operates on annual to century timescales, which is substantially faster than the ocean feedbacks. The productivity of terrestrial plants increases instantaneously with elevated atmospheric CO<sub>2</sub> concentration because a physiological response of the plant stomata leads to higher water-use efficiency and a consequent increase in plant biomass (Denman et al. 2007). Enhanced respiration of plant tissues and accelerated decomposition of soil organic matter owing to elevated temperatures counteract this effect, but a net result of projected changes in CO<sub>2</sub> and climate is an increase of the land carbon storage in most vegetation models (Cramer et al. 2001, Friedlingstein et al. 2006). This is reflected in **Figures 3** and **4**. A presence of vegetation feedback in the simulations substantially reduces an airborne CO<sub>2</sub> fraction, especially during the first hundred years. After this period, the ocean carbon uptake gains control over the atmospheric CO<sub>2</sub> concentration because of the much larger buffering capacity of the ocean in comparison with the land.

Although these results are in line with expected long-term vegetation feedback (Bala et al. 2005, Plattner et al. 2008), many uncertainties in the representation of long-term land biogeochemistry make the land feedback story more comprehensive. Modeling of soil carbon dynamics is still in its



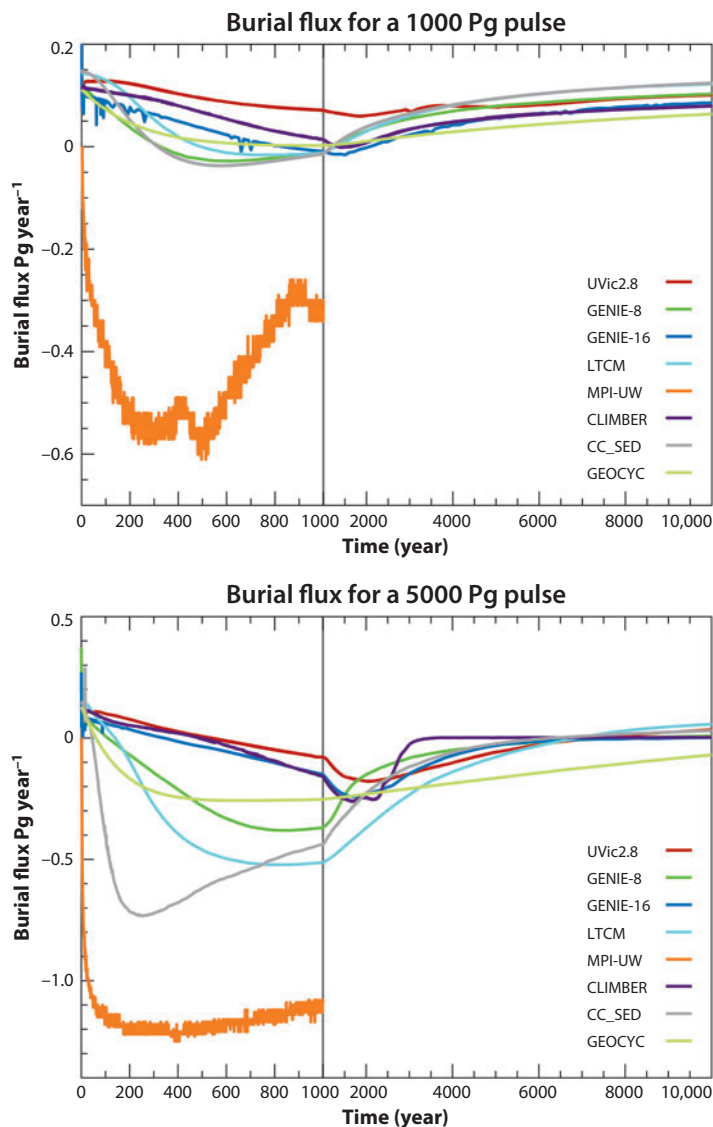
**Figure 3**

Impacts of feedbacks to atmospheric CO<sub>2</sub> concentration, expressed in terms of atmospheric CO<sub>2</sub> difference when the feedback is applied. Results from 1000 Pg C release experiments. Note the split scale that expands the action in the first 1000 years. The physically impossible positive sediment feedback for CC<sub>SED</sub> in the first millennium is an artifact of different initial conditions for the two versions of the model.



**Figure 4**

Impacts of feedbacks to atmospheric CO<sub>2</sub> concentration, expressed in terms of atmospheric CO<sub>2</sub> difference when the feedback is applied. Results from 5000 Pg C release experiments. Note the split scale that expands the action in the first 1000 years.



**Figure 5**

Global rates of  $\text{CaCO}_3$  burial from the models. Negative values indicate net dissolution of  $\text{CaCO}_3$ . Note the split scale that expands the action in the first 1000 years.

infancy; many important mechanisms, the priming effect of the addition of fresh organic material to the soils (Fontaine et al. 2003) and processes of anaerobic decomposition of organic matter, for example (Frolking et al. 2001), are not yet accounted for in the coupled global models. Nitrogen and phosphorus balance is ignored in most of the models (Reich et al. 2006), and changes in carbonate storages in dryland soils are neglected (Lal et al. 2000). Models of vegetation (forest) dynamics on a global scale are extremely simplified and difficult to validate because of the long timescales involved (Purves & Pacala 2008). Finally, changes in the land carbon uptake due to future alteration of land use by humans are almost impossible to foresee. All these limitations of

the land model assumptions make the simulations of the land carbon response to the CO<sub>2</sub> pulse presented here illustrative rather than predictive.

The mean lifetime, or CO<sub>2</sub> transit time back to the solid earth, depends strongly on the assumption of the timescale for the silicate-weathering feedback, which is not resolved by the duration of these model experiments or the primary focus of the modeling exercise. If we assume, however, that 10% of the fossil fuel remains in the atmosphere until it is neutralized by silicate weathering on a timescale of 100,000 years, then the mean lifetime of fossil fuel CO<sub>2</sub> for both CO<sub>2</sub>-release scenarios 1000 and 5000 Pg C is ~12–14 thousand years. Assuming a 400,000-year time constant for the silicate-weathering feedback would result in mean CO<sub>2</sub> lifetimes of ~45 thousand years. These exceed the IPCC-quoted atmospheric lifetime by a factor of 50 or 100 and would change the GWP of the other greenhouse gases by this factor. We do not argue that other greenhouse gases are unimportant by their lack of a comparable geological-timescale tail to their atmospheric residence. It makes sense to base the calculation of GWP on the timescale of the human lifetime. However, the calculation should be transparent and based on the true behavior of CO<sub>2</sub> in the Earth system rather than on a misunderstanding.

## CONCLUSIONS

The models presented here give a broadly coherent picture of the fate of fossil fuel CO<sub>2</sub> released into the atmosphere. Equilibration with the ocean will absorb most of it on a timescale of 2 to 20 centuries. Even if this equilibration were allowed to run to completion, a substantial fraction of the CO<sub>2</sub>, 20–40%, would remain in the atmosphere awaiting slower chemical reactions with CaCO<sub>3</sub> and igneous rocks. The remaining CO<sub>2</sub> is abundant enough to continue to have a substantial impact on climate for thousands of years. The changes in climate amplify themselves somewhat by driving CO<sub>2</sub> out of the warmer ocean. The CO<sub>2</sub> invasion has acidified the ocean, the pH of which is largely restored by excess dissolution of CaCO<sub>3</sub> from the sea floor and on land and, ultimately, by silicate weathering on land. The recovery of ocean pH restores the ocean's buffer capacity to absorb CO<sub>2</sub>, tending to pull CO<sub>2</sub> toward lower concentrations over the next 10,000 years. The land biosphere has its greatest impact within the first few centuries, which is when CO<sub>2</sub> peaks. Nowhere in these model results or in the published literature is there any reason to conclude that the effects of CO<sub>2</sub> release will be substantially confined to just a few centuries. In contrast, generally accepted modern understanding of the global carbon cycle indicates that climate effects of CO<sub>2</sub> releases to the atmosphere will persist for tens, if not hundreds, of thousands of years into the future.

## DISCLOSURE STATEMENT

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