



The ‘long tail’ of anthropogenic CO₂ decline in the atmosphere and its consequences for post-closure performance assessments for disposal of radioactive wastes

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ABSTRACT

The extended timescales involved in the decay of radioactive wastes to safe levels mean that geological disposal facilities must continue to function effectively long into the future. It is therefore essential to consider long-term climate evolution in post-closure performance assessments in order to evaluate a geological disposal system's response and robustness to a variety of potential environmental changes, driven by both natural and anthropogenic forcings. In this paper, we illustrate the multiple decay components that characterize the primary driver of climate change – atmospheric CO₂ – in response to fossil fuel carbon emissions. We perform a multi-exponential analysis on a series of atmospheric CO₂ decay curves predicted by an Earth system model and create an empirical response function that encapsulates the long-term (>1 kyr) removal of excess CO₂ from the atmosphere. We present this response function as a simple tool for rapidly projecting the future atmospheric CO₂ concentration resulting from any plausible cumulative release of CO₂. We discuss the implications of the long ‘tail’ to this atmospheric CO₂ decay curve, both in terms of future climate evolution as well as potential impacts on radioactive waste repositories.

KEYWORDS: carbon cycle, climate change, radioactive waste disposal.

Introduction

THE significant timescales involved in the decay to safe levels of radionuclides incorporated in radioactive wastes means that geological disposal facilities containing low- and intermediate-level wastes must continue to function effectively for up to 100,000 years, as in the case of the Low Level

Waste Repository (LLWR) in the UK (LLWR, 2011). For high-level wastes and spent nuclear fuel, repositories such as the proposed KBS-3 facility at Forsmark in Sweden (SKB, 2011) must remain functional for up to 1 million years. It is therefore essential to consider long-term climate evolution in post-closure performance assessments in order to evaluate a geological disposal system's response to and robustness against a variety of potential environmental changes, driven by both natural (e.g. orbital variations) and anthropogenic (e.g. fossil fuel emissions) forcings.

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In order to identify what climate changes may be of relevance to different waste repositories and how they might be impacted by these changes, facilities can be categorized based on various criteria. These categories include the mode of construction (e.g. excavated from the surface or at depth), the geological context, the hydrogeological context (i.e. saturated or unsaturated), the coastal context and the potential for extreme climates. For repositories in glaciated environments for example, the environmental change that is likely to have the most significant impact is the passage of the ice sheet margin across the site (Becker *et al.*, 2014). Whether the accompanying changes in the hydrological regime are likely to have a large impact can be inferred from the hydrogeological and geological context, while the mode of construction will give an indication of how vulnerable the facility is to surface denudation and permafrost formation. For high-level waste repositories the timing and severity of the glaciation may be of particular importance, as this will determine the remaining radionuclide inventory that can be impacted by the altered hydrogeological conditions that occur during passage of the ice sheet. It will also determine the duration of the period of permafrost conditions prior to glaciation and, therefore, be a key factor in determining whether the permafrost develops to repository depth (SKB, 2011). Thus, different repositories have different key timescales that need to be considered, based in part on the climate change processes that are expected to have the most significant impact on the facility. In the case of the UK's LLWR for example, the coming centuries will be key as sea level rise and the resultant coastal erosion are expected to affect the site on these timescales (Fish *et al.*, 2010), whereas permafrost is projected to impact Sweden's shallow disposal facility for operational and decommissioning wastes (SFR) on timescales of tens of millennia (SKB, 2013). In contrast, the key timescale for the proposed KBS-3 repositories in Sweden and Finland is more than 100 kyr. In this context, the timing of the next glacial inception and amplitude of future glacial-interglacial cycles are key.

In general, the primary driver of global and regional climate change on timescales from decades to >1 Myr is variation in the concentration of atmospheric greenhouse gases and principally, carbon dioxide (CO₂). It is in this way that anthropogenic activities are now increasingly impacting the climate system (IPCC, 1990, 1995, 2001, 2007). Between 1750 and 2011 ~555 ±

85 PgC of CO₂ were emitted, raising atmospheric CO₂ concentrations by 40% from 278 to 390.5 ppm in 2011 (Ciais *et al.*, 2013). This concentration is now higher than has occurred at any time in at least the last 800 kyr as recorded in ice cores (Luthi *et al.*, 2008) and equal to the maximum values reached about 2–3 million years ago (Martinez-Boti *et al.*, 2015).

A sound understanding of the global carbon cycle and associated controls on the removal of anthropogenic CO₂ from the atmosphere, particularly geological processes with timescales comparable to the lifetimes of some of the longer-lived major radionuclides, is hence essential. This paper focuses on the range of processes that comprise the long-term carbon cycle and how they affect atmospheric CO₂, and presents a simple empirical representation of the decay of an atmospheric CO₂ perturbation. We also discuss the implications of the projected atmospheric CO₂ response for climate and post-closure performance assessments. This work contributes to that of Working Group 6 of the International Atomic Energy Agency (IAEA) MODARIA Programme, which has been tasked with developing a common framework for addressing climate change in post-closure radiological assessments of solid radioactive waste disposal.

The fate of fossil fuel CO₂

As CO₂ is emitted to the atmosphere via the combustion of fossil fuels and land-use change, its concentration is elevated above the long-term average which, for the last ten thousand years of the Holocene, lies in the range ~260–280 ppm. This excess anthropogenic CO₂ is subsequently removed by a range of carbon cycle processes operating on a variety of timescales, summarized in Fig. 1. These are:

- (1) On the shortest timescales of months to years (Fig. 1-I), gaseous CO₂ is mixed between the two hemispheres of the atmosphere and transferred across the air–sea interface of the oceans, dissolving in surface waters and decreasing seawater pH (Turley *et al.*, 2010). The net outcome of this process is approximated by reaction 1 in Fig. 1-I. CO₂ fertilization of terrestrial primary productivity also results in a further uptake of CO₂ from the atmosphere (Sitch *et al.*, 2008). Climate feedbacks modify the rate of uptake by reducing the solubility of CO₂ in seawater (warming) as well as mixing (ocean stratification).

- (2) Over decades to centuries, surface ocean waters rich in dissolved CO₂ are transported down into the ocean interior, primarily through the formation of intermediate and deep waters at high latitudes (Archer and Brovkin, 2008; Archer *et al.*, 1998). In terrestrial environments, a new equilibrium will be established with enhanced primary productivity driving a larger soil carbon pool, illustrated in Fig. 1-II. Climate feedbacks again modify the strength of carbon uptake from the atmosphere for instance via soil warming, with increased metabolic activity driving an increased rate of return of CO₂ back to the atmosphere.
- (3) The chemical impact of dissolving excess CO₂ in seawater results in a reduction in carbonate (CO₃²⁻) concentration in ocean waters causing a reduction in the stability of biogenic calcium carbonate (CaCO₃) minerals (principally calcite). Sedimentary CaCO₃ that was originally formed at the surface and transported by gravitational settling to the seafloor may now start to dissolve. This process (reaction 2 in Fig. 1-III) is known as seafloor neutralization (Archer *et al.*, 1997) and has the effect of partially regenerating CO₃²⁻ concentrations and hence the buffering capacity of the ocean, allowing additional draw-down of atmospheric CO₂ via reaction 1 (Ridgwell and Hargreaves, 2007). This all occurs on timescales of millennia and can be modified by any changes in ocean circulation induced by a warming climate.
- (4) At the same time as rates of CaCO₃ burial in marine sediments are reduced (by process #3 above), the supply of solutes from terrestrial rock weathering continues. This creates an imbalance in the supply *vs.* removal of Ca²⁺ and HCO₃⁻ to the ocean that on multi-millennial timescales results in further uptake of CO₂ (Lenton and Britton, 2006; Ridgwell and Hargreaves, 2007). This is known as 'terrestrial CaCO₃ neutralization' and is illustrated in Fig. 1-IV. Here climate feedbacks, by enhancing the weathering of carbonate rocks (e.g. via more acidic rainwater), will act to increase the recovery of ocean carbonate chemistry.
- (5) The final process which occurs on a timescale of hundreds of thousands to millions of years – the weathering of terrestrial silicate rocks – will act to remove the remaining atmospheric CO₂ perturbation and restore atmospheric CO₂

back to its pre-industrial state (Lenton and Britton, 2006) via reaction 3 in Fig. 1-V.

A helpful parallel can be drawn between the decay of an atmospheric CO₂ perturbation via the action of the various processes described above, and the radioactive decay of waste. Both types of decay can be characterized by a series of decay curves, each with a different characteristic timescale – each either associated with individual radionuclides or different carbon cycle processes. For CO₂, the combined operating timescale and uptake capacity of each of these carbon cycle processes controls the atmospheric lifetime of the carbon perturbation, and hence the timescale and magnitude of the resultant climate changes. The radioactive isotopes of most interest in waste disposal exhibit a similar range of decay timescales to that of excess atmospheric CO₂, ranging from decades (e.g. Cs-137 – 30.1 yr) to hundreds of years (e.g. Am-241 – 458 yr) to tens of thousands of years (e.g. Pu-239 – 24.1 kyr) to hundreds of thousands of years or more (e.g. U-234 – 245.5 kyr). This comparative similarity highlights the importance of consideration of long-term atmospheric CO₂ in the future integrity of waste repositories.

Characterizing the timescales of atmospheric CO₂ decay

The decay of an atmospheric CO₂ perturbation, and hence the long-term carbon cycle processes, can be captured in the form of an empirical function comprising a series of exponential decay curves, fitted to a composite CO₂ decay curve that is generated by means of a mechanistic numerical model. This approach has been undertaken in a number of past studies which produced response functions for specific sized CO₂ releases (Archer *et al.*, 1997; Colbourn, 2011), but is extended here by including a number of climate-carbon feedbacks on ocean circulation and terrestrial weathering and by using a larger range of CO₂ emissions.

To characterize the timescales of CO₂ decay, we utilize an Earth system model (*cGENIE*). This is based on the efficient climate model of Edwards and Marsh (2005) and incorporates a 2-D Energy-Moisture Balance atmosphere (EMBM), coupled to a 3-D frictional geostrophic ocean circulation model together with a dynamic-thermodynamic sea-ice component. The version of the *cGENIE* model used here also includes a representation of the global carbon cycle, including ocean cycling of

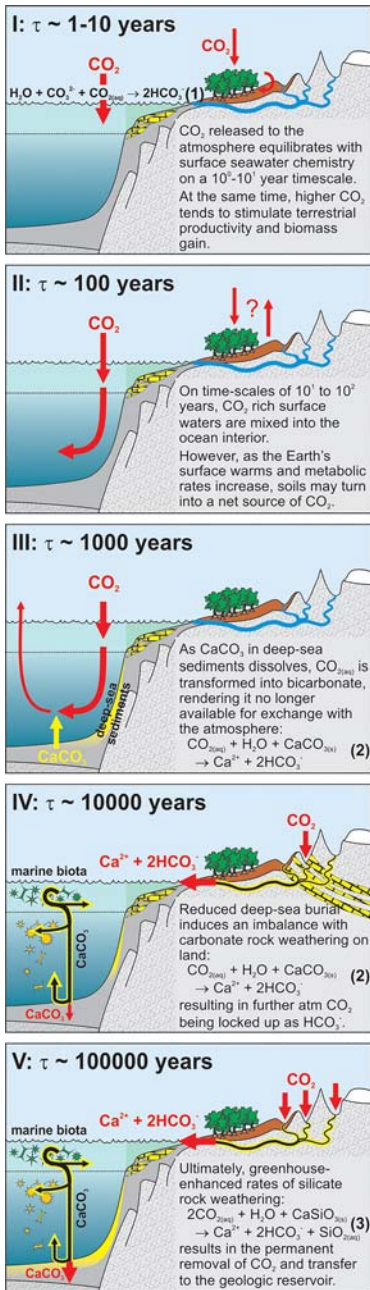


FIG. 1. Illustration of the primary mechanisms of sequestration of CO_2 from the atmosphere. Panel (I) represents air-sea gas exchange and ‘ CO_2 fertilization’ of the terrestrial biosphere, (II) shows ocean invasion and soil carbon inventory adjustment, (III) represents sea-floor CaCO_3 neutralization, (IV) illustrates terrestrial CaCO_3 neutralization, and (V) represents the silicate weathering carbon sink.

carbon and nutrients (Ridgwell *et al.*, 2007), geochemical interactions with marine sediments (Ridgwell and Hargreaves, 2007) and the weathering of carbonate and silicate rocks on land (Colbourn *et al.*, 2013). The model is configured on a 36×36 equal horizontal grid with 8 vertical levels in the ocean. It is non-seasonally insolation forced (i.e. with an annual average latitudinal distribution of solar insolation) and because of the absence of a dynamical atmospheric circulation model component, requires that the spatial (and annual average) distribution of wind stress on the ocean surface and transport by circulation patterns, be prescribed (see Edwards and Marsh, 2005). The time step of the ocean circulation model component is approximately 7 days, with ocean and terrestrial carbon cycle processes updated a little less frequently (see Ridgwell *et al.*, 2007) and deep-sea sediment composition only annually (Ridgwell and Hargreaves, 2007).

The Earth system model is sufficiently computationally efficient to undertake simulations of the order of 1 Myr, yet capable of representing to a first-order the basic responses of: (a) CO_2 uptake by the ocean and interaction with marine sediments, (b) the global climate response to elevated CO_2 and primary feedbacks of climate on ocean circulation and carbon cycling, and (c) weathering response to climate. However, there is no inter-annual variability in climate in this model and nor is the interaction of the terrestrial biosphere with CO_2 and climate included. There is currently a great deal of uncertainty associated with the overall response of the terrestrial biosphere to future climate changes, such that even the sign of the change is not known for certain. There is also no representation of ice sheets in the model and associated feedbacks such as were important during the past glacial-interglacial cycles (Kohfeld and Ridgwell, 2009).

We perform a series of ten experiments, each of 1 Myr duration, in which idealized pulse emissions of CO_2 are applied to the model. In these, all input of CO_2 to the atmosphere occurs at a uniform rate and over a single model year. The total carbon release spans 1000 to 10,000 PgC, deemed to be a reasonable range of fossil fuel CO_2 emissions that could be expected to be released in the next few hundred years. It is estimated that approximately 1000–2000 PgC remain in fossil fuel reserves that are presently economically exploitable (Ciais *et al.*, 2013). However, fossil fuel resources (where economic extraction may be feasible in the future) are estimated to total 8543–13,649 PgC, while non-

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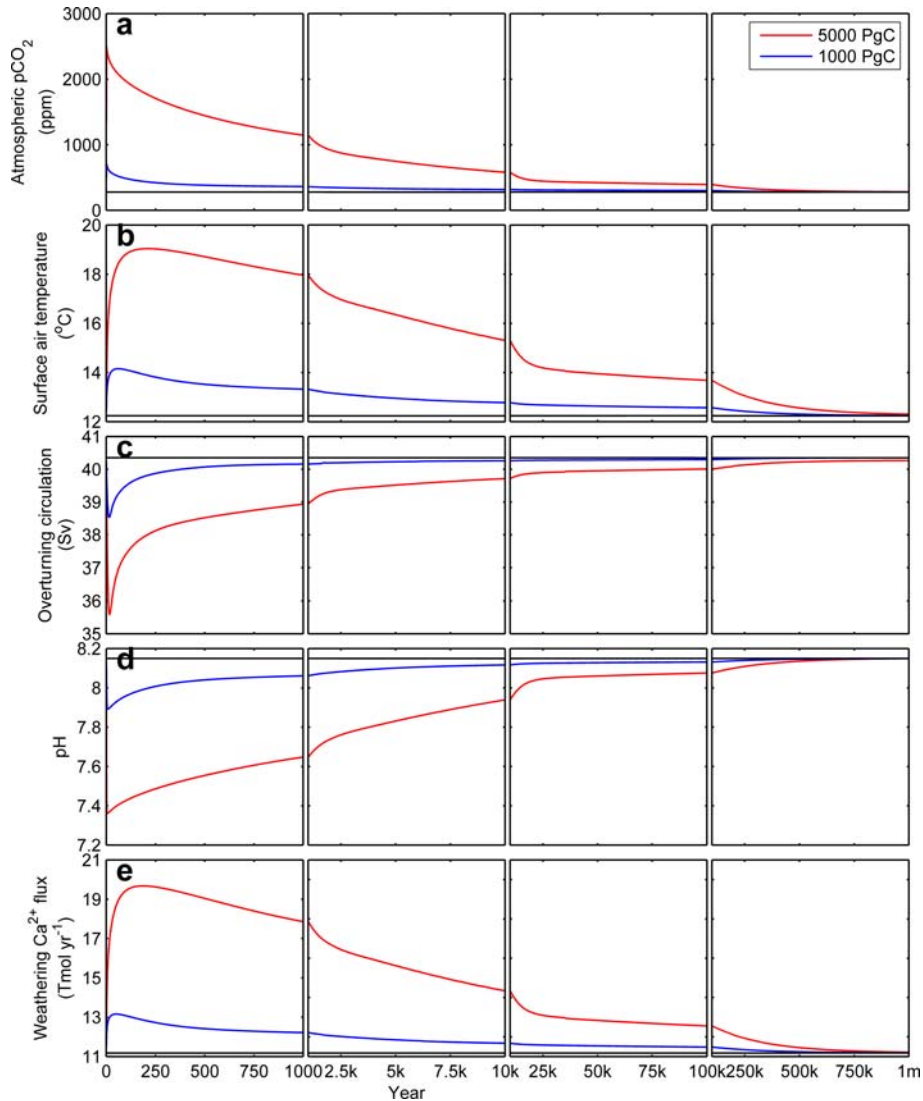


FIG. 2. Climatic consequences (mean global) for anthropogenic emissions (1000 and 5000 PgC), as simulated by the *cGENIE* model. Pre-industrial levels are shown in black. (a) Atmospheric p_{CO_2} . (b) Global mean surface air temperature. (c) Global meridional overturning circulation. (d) Ocean surface pH. (e) Global flux of Ca^{2+} to the ocean from terrestrial weathering.

conventional resources such as methane clathrates could be as high as 20–25,000 PgC (Rogner, 1997). Pulse rather than time-dependent emissions have been used as previous studies have found that total emissions, rather than the rate of release, is the dominant control on the long-term response of atmospheric CO₂, assuming emissions occur over no longer than about the next 300 years (Eby *et al.*,

2009). This also appears to be true for the evolution of the ice sheets in response to fossil fuel emissions (Charbit *et al.*, 2008). However, whilst a year-long pulse release facilitates the extraction of the underlying process timescales, our results will not be applicable to the evolution of atmospheric CO₂ over the coming centuries as emissions continue and evolve in rate.

The long-term response of the climate to CO₂ emissions of 1000 and 5000 PgC is shown in Fig. 2, as predicted by cGENIE. While the initial atmospheric CO₂ peak subsides relatively quickly (Fig. 2a), it is clear that the overall perturbation is long-lived, with atmospheric p_{CO₂} taking hundreds of thousands of years to return to pre-industrial levels, particularly for the 5000 PgC scenario. As a consequence, the wider issues associated with anthropogenic CO₂ emissions, including warming (Fig. 2b), weakening of ocean circulation (Fig. 2c), ocean acidification (Fig. 2d) and increased terrestrial weathering rates expected due to an overall warmer and wetter climate (Fig. 2e) also last for hundreds of thousands of years.

In order to explore the different timescales of decay of the atmospheric CO₂ perturbations, a multi-exponential function was fitted to the decay curves for the pulse experiments, taking the form:

$$CO_2(t) = 278.0 + 0.469 \times E \sum_{i=1}^n A_i \exp^{-t/\tau_i} \quad (1)$$

where CO₂(t) is the atmospheric CO₂ concentration (ppm) at a given time (t), E is the total emissions (PgC) which are assumed to be released at time t = 0, and n is the number of exponentials used in the fit. The factor 0.469 simply converts PgC of instantaneous CO₂ emissions into the equivalent atmospheric concentration in ppm, and 278.0 is the assumed pre-industrial CO₂ concentration. For each (i) of the exponentials; τ_i is the timescale of decay and A_i represents the fraction of total emissions removed from the atmosphere over this timescale.

An equation consisting of the sum of five exponentials was chosen as the Akaike Information Criterion (AIC) value, a measure of the relative quality of the fit which favours models with fewer parameters (a low value is best), indicated that this provided a better fit (had a lower value) than the four exponential model.

Adding more exponentials does not improve the overall fit, as generally five different exponential curves are fitted, with the additional curve(s) being very similar to one of the other curves or else not having meaningful coefficient values.

The fitting coefficients (A_i and τ_i) were made linear functions of the total amount of carbon released (μ PgC) via a regression analysis, hence the full empirical function took the form:

$$CO_2(t) = 278.0 + 0.469 \times E \sum_{i=1}^n (\alpha_i + \beta_i \mu) \exp^{-t/(\gamma_i + \delta_i \mu)} \quad (2)$$

Using the parameters listed in Table 1, this empirical response function is capable of projecting the atmospheric decay of CO₂ for a range of total emissions. For further details of the above methodology please see Colbourn *et al.* (2015).

Long-term future CO₂

The dynamics of the global climate and carbon cycle system with its myriad of processes and attendant feedbacks cannot be accurately partitioned into just a few idealized decay curves. However, each of the five exponentials that were fitted to the CO₂ decay curves will tend to have some correspondence with one or more of the global carbon cycle processes described earlier (and summarized in Fig. 1) as the individual processes span a wide range of timescales at which they will tend to dominate. The following section discusses the values of the fitting coefficients for each of the five exponentials, giving the median values and range (minimum to maximum) for each parameter. It is likely that the first and second exponentials, with median lifetimes of 6.5 and 50.37 yr (5.3 to 7.4 yr and 39.6 to 57.7 yr) respectively, represent invasion of CO₂ into the upper water column (I). The first exponential

TABLE 1. Linear regression scaling parameters.

Fitting coefficient	Parameter	i				
		1	2	3	4	5
A _i	α _i	0.1372	0.184	0.4207	0.0938	0.07413
	β _i	-1.26E-05	-1.33E-05	-1.47E-06	3.32E-05	-3.12E-07
τ _i	γ _i	6.62	50.87	220.96	2678.17	242092.1
	δ _i	-3.03E-05	-0.00012	0.05723	0.8447	2.41367

captures processes with shorter timescales, such as dissolution of gaseous CO₂ into the surface ocean, removing 5% (3 to 16%) of the total CO₂ emitted. The second represents mixing of dissolved CO₂ into the upper water column, resulting in the removal of 10% (7 to 20%) of CO₂, meaning that the process(es) of ocean invasion removes 15% (10 to 36%) of the total CO₂ emitted to the atmosphere in this model. The third exponential captures the transport of CO₂ into the ocean interior (II). It has a timescale of 580 yr (214 to 734 yr), which is comparable to that of ocean mixing, and accounts for the removal of a further 41% (35 to 45%) of the CO₂ perturbation. The fourth exponential has an *e*-folding timescale of 7 kyr (4 to 11 kyr), and represents a combination of seafloor and terrestrial CaCO₃ neutralization (III and IV). 28% (13 to 42%) of total CO₂ emissions are removed by these processes. Finally, enhanced silicate (and carbonate) weathering (V) results in the uptake of the remaining 7% (7 to 8%) of the excess CO₂, operating on an average timescale of 256 kyr (243 to 267 kyr).

Our empirical response function (equation 2) represents a useful tool for rapidly projecting the long-term response of atmospheric CO₂ to emissions of any realistic size, removing the need for long simulations using computationally expensive models. However, it should be borne in mind that the first several hundred years of the simulation cannot be considered as realistic, as the emissions are released as a pulse rather than as a time-dependent flux, which would occur in reality. Despite this, atmospheric CO₂ concentrations are predicted to within $\pm 11\%$ by year 1000 compared to the equivalent simulation by the Earth system model. The response function is hence suitable for CO₂ projections on timescales of a millennium and beyond.

The empirical model can be applied to help further illustrate the evolution of atmospheric CO₂ by means of a continuous (interpolated) graphical function of both time and total emissions size, shown in Fig. 3. Readily apparent in this analysis is that while the initial large perturbations subside relatively early on, the remaining atmospheric CO₂ anomaly decreases very slowly. For emissions of more than 3000 PgC, even after 1 Myr atmospheric p_{CO_2} not yet equilibrated at natural concentrations (*c.* 280 ppm). The long atmospheric lifetime of perturbations is in good agreement with the findings of previous studies (Archer, 2005; Colbourn, 2011; Colbourn *et al.*, 2015) and has important implications for future climate evolution, both over the coming centuries but also hundreds of thousands of years from now.

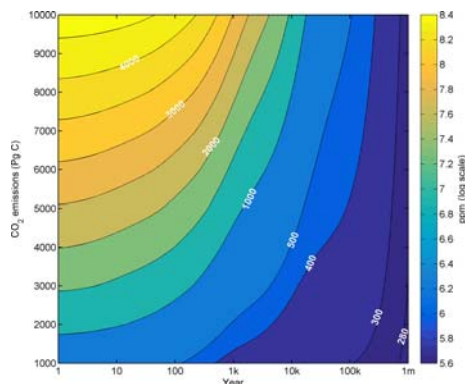


FIG. 3. Evolution over time of atmospheric p_{CO_2} (see colour bar) for emissions of 1000–10,000 PgC predicted using the response function (equation 2). Atmospheric p_{CO_2} (ppm) for selected contours is indicated (white labels). Note the logarithmic scale on the *y*-axis.

Potential implications for post-closure performance assessments

Glacial-interglacial cycles, largely driven by the waxing and waning of ice sheets in the Northern Hemisphere, have been a dominant feature of the Earth's climate system over the past several million years. The timing and periodicity of Northern Hemisphere glaciation is closely linked to the geometry of the Earth's orbit and its rotation through its impact on insolation (solar radiation received at the top of the atmosphere). Summertime Northern Hemisphere is known to be particularly important (Hays *et al.*, 1976; Kawamura *et al.*, 2007; Milankovitch, 1941; Ruddiman, 2006) as it exerts a strong control on whether winter snow cover persists into the next year and hence accumulates, or completely melts. However, on its own, an insolation-only description of the glacial-interglacial cyclicality is incomplete (Ridgwell *et al.*, 1999) and changes in carbon dioxide (CO₂) and hence the degree of greenhouse gas warming also appear intimately linked (Petit *et al.*, 1999). This raises the potential for future anthropogenic activities to impact on the glacial-interglacial cycles by modifying atmospheric CO₂ and also, because the current or proposed locations of many nuclear waste sites in the Northern Hemisphere are often at latitudes higher than or close to glacial maximum ice margins, to impact on waste repositories.

Variations in orbital parameters and insolation for the last and next 500 kyr are shown in Fig. 4. In the

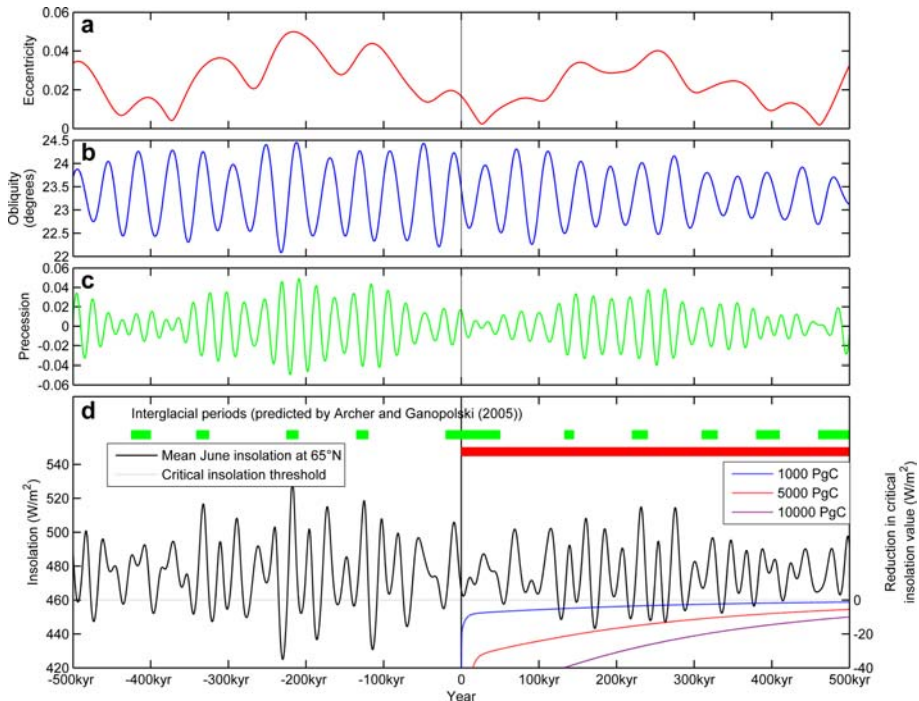


FIG. 4. Orbital parameters for the last and next 500 kyr calculated using the Laskar *et al.* (2004) solution, and the impact of anthropogenic emissions on the future evolution of climate. (a) Eccentricity. (b) Obliquity. (c) Precession. (d) Top panel: Interglacial periods (solid blocks) for natural climate evolution at 280 ppm (green) and emissions of 5000 PgC (red). Left y-axis: Insolation, and critical insolation threshold at 280 ppm that triggers glacial inception, adapted from Archer and Ganopolski (2005). Right y-axis: Reduction in critical insolation threshold resulting from radiative forcing following anthropogenic emissions (dashed lines). Capped at -40 W/m^2 as this encompasses the minimum insolation values for the illustrated period. Scaled similar to Archer and Ganopolski (2005).

absence of elevated atmospheric CO_2 concentrations, the relatively muted cyclical minima in 65°N June insolation (Fig. 4d) suggests that the next glacial inception may not occur for some tens of thousands of years in the future. For instance, Archer and Ganopolski (2005) found that glacial inception was initiated in the ‘CLIMBER-2’ coupled climate-ice sheet model when Northern Hemisphere insolation fell below a critical value (Fig. 4d). Under natural conditions of pre-industrial CO_2 (~ 280 ppm) glacial inception occurs periodically with the first occurrence not until after 50 kyr (Fig. 4d; top panel), in good agreement with previous studies (Berger and Loutre, 2002; Paillard, 2001).

The picture changes if CO_2 emissions are taken into account. Archer and Ganopolski (2005) used the same model to examine the potential timing of the next glaciation following anthropogenic CO_2 emissions of 300, 1000 and 5000 PgC released over 150 years. They found that CO_2 emissions had the

capacity to affect the future evolution of climate for hundreds of thousands of years, increasing radiative forcing and so decreasing the critical insolation threshold from its natural value. The 300 PgC scenario glaciated after 50 kyr, similar to an experiment forced with natural CO_2 evolution, although glacial termination in the emissions scenario took several millennia longer. A release of 1000 PgC prevented glaciation until 130 kyr from present, due to the long atmospheric lifetime of emissions. Anthropogenic emissions of 5000 PgC resulted in a termination of the glacial-interglacial cycles, with the current interglacial continuing for at least the next 500 kyr (Fig. 4d). A different study by Berger and Loutre (2002), forcing a different coupled climate-ice sheet model but with simplified assumptions regarding CO_2 , also predicted an exceptionally long current interglacial, lasting for a further 5 kyr (for constant p_{CO_2} of 210 ppm) to 50 kyr (with an initial p_{CO_2} of

750 ppm) after present, with the next glacial maximum occurring in 100 kyr. An atmospheric concentration of less than 220 ppm was required to force an early glacial inception.

Although influential and including a representation of coupled climate-ice sheet dynamics not available here, these previous studies did not include a fully explicit description of the long-term decline of fossil fuel CO₂. For instance, Berger and Loutre (2002) assumed that atmospheric p_{CO_2} returned to its pre-industrial state following emissions within 1000 years. Archer and Ganopolski (2005) considered different timescales of CO₂ decay, but made assumptions regarding the timescale of the silicate weathering feedback. They also fixed the shorter timescales and characteristic removal fractions according to a single magnitude of CO₂ release rather than allowing them to become functions of the CO₂ release, as is encapsulated in equation 2.

Using our empirical response function, in Fig. 4d we illustrate the potential reduction in the critical insolation threshold due to radiative warming for the 1000, 5000 and 10,000 PgC emissions scenarios, and its decline as a function of time compared to future variations in June 65°N insolation. This was calculated based on the relationship between global radiative forcing from CO₂ and the model-predicted critical insolation value found by Archer and Ganopolski (2005). The addition of anthropogenic CO₂ to the atmosphere reduces the threshold such that the next glaciation may be delayed until ~130 kyr from now for the 1000 PgC scenario, and beyond 500 kyr for the 5000 and 10,000 PgC scenarios. Positive feedbacks with carbon release from methane hydrates and thawing permafrost, not included in our analysis here but included in the case of hydrates by Archer and Ganopolski (2005), may act to increase the CO₂ maximum reached and/or delay the overall rate of decay, hence further displacing future glacial-interglacial cycles. It should also be noted that a significant amount of uncertainty remains in our understanding of the long-term carbon cycle. In particular, past atmospheric CO₂ has varied naturally by 80–100 ppm in association with the glacial-interglacial cycles (Petit *et al.*, 1999), but the driving mechanisms for these changes are not yet known (Kohfeld and Ridgwell, 2009).

Our analysis illustrating the importance of CO₂ emissions on glacial inception has important implications for waste repositories. During glacial episodes, the advance and retreat of ice sheets can lead to surface erosion (Clayton, 1994), deposition and isostatic effects. Major changes in the hydrological and hydrogeological regime close to the ice

margin may also occur (SKB, 2011). Permafrost may develop during glacial or periglacial conditions, reaching depths of tens or hundreds of metres (French, 2007). Hence, a significantly extended interglacial or a delayed glaciation with an extended prior period of periglacial conditions can have important implications, which may be either detrimental or advantageous to safety, for the long-term performance of repositories located in previously glaciated regions.

Other, both natural and anthropogenically-driven environmental changes may also be important. For example, sea-level rise is expected to have a significant impact on the safety of the LLWR (LLWR, 2011). This is affected by the atmospheric lifetime of CO₂ emissions through its impact on radiative forcing, which in turn affects thermal expansion of the oceans (Williams *et al.*, 2012) and the risk of melting and/or collapse of the Greenland and West Antarctic ice sheets (Church *et al.*, 2013). In contrast, the proposed Yucca Mountain facility for deep geological disposal of spent fuel and high-level waste, which is located in the unsaturated zone in fractured rock, is thought to be vulnerable to high-intensity fluvial episodes in a warmer wetter climate through their impact on infiltration (Thorne, 2013). This highlights that, while regional changes in temperature and precipitation are important, modifications to atmospheric and oceanic circulation regimes are also of interest due to their influences on local and regional climates.

Conclusions

The comparable decay timescales of radioactivity and atmospheric CO₂ emissions highlight the importance of consideration of the latter in post-closure performance assessments for radioactive waste repositories. The characteristic response times of the various processes of the global carbon cycle together with their respective carbon uptake capacities, control the overall timescale and severity of the CO₂ perturbation. This can have important implications for future climate changes, including potentially affecting the timing of the future glacial-interglacial cycles, the occurrence (and extent and depth) of permafrost, and the hydrological and hydrogeological regime at different spatial scales. We provide an empirical response function as a simple tool for rapidly projecting the long-term response of atmospheric CO₂ to emissions of any realistic size, removing the need for long simulations using computationally expensive models.

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References

- Archer, D. (2005) Fate of fossil fuel CO₂ in geologic time. *Journal of Geophysical Research – Oceans*, **110**. doi: 10.1029/2004JC0026252625.
- Archer, D. and Brovkin, V. (2008) The millennial atmospheric lifetime of anthropogenic CO₂. *Climatic Change*, **90**, 283–297.
- Archer, D. and Ganopolski, A. (2005) A movable trigger: Fossil fuel CO₂ and the onset of the next glaciation. *Geochemistry Geophysics Geosystems*, **6**. doi: 10.1029/2004GC00891.
- Archer, D., Kheshgi, H. and Maier-Reimer, E. (1997) Multiple timescales for neutralization of fossil fuel CO₂. *Geophysical Research Letters*, **24**, 405–408.
- Archer, D., Kheshgi, H. and Maier-Reimer, E. (1998) Dynamics of fossil fuel CO₂ neutralization by marine CaCO₃. *Global Biogeochemical Cycles*, **12**, 259–276.
- Becker, J. K., Lindborg, T. and Thorne, M. (2014) Influence of climate on landscape characteristics in safety assessments of repositories for radioactive wastes. *Journal of Environmental Radioactivity*, **138**, 192–204.
- Berger, A. and Loutre, M.F. (2002) An exceptionally long interglacial ahead? *Science*, **297**, 1287–1288.
- Charbit, S., Paillard, D. and Ramstein, G. (2008) Amount of CO₂ emissions irreversibly leading to the total melting of Greenland. *Geophysical Research Letters*, **35**, 33472.
- Church, J.A. *et al.* (2013) Sea level change. Pp. 1137–1216 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* (T.F. Stocker *et al.*, editors). Cambridge University Press, Cambridge, UK and New York, USA.
- Ciais, P. *et al.* (2013) Carbon and other biogeochemical cycles. Pp. 465–570 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* (T.F. Stocker *et al.*, editors). Cambridge University Press, Cambridge, UK and New York, USA.
- Clayton, K. (1994) Glaciation of the British Isles: An Approach Seeking to Determine the Role of Glaciation in Landform Development over the Last Million Years. *Nirex Safety Studies Report NSS/R337*, Available from Radioactive Waste Management Limited, UK.
- Colbourn, G. (2011) *Weathering effects on the carbon cycle in an Earth System Model*. Unpublished PhD thesis, University of East Anglia, UK.
- Colbourn, G., Ridgwell, A. and Lenton, T. M. (2013) The Rock Geochemical Model (RokGeM) v0.9. *Geoscientific Model Development*, **6**, 1543–1573.
- Colbourn, G., Ridgwell, A. and Lenton, T. (2015) The time scale of the silicate weathering negative feedback on atmospheric CO₂. *Global Biogeochemical Cycles*, **29**, 583–596.
- Eby, M., Zickfeld, K., Montenegro, A., Archer, D., Meissner, K.J. and Weaver, A.J. (2009) Lifetime of anthropogenic climate change: millennial time scales of potential CO₂ and surface temperature perturbations. *Journal of Climate*, **22**, 2501–2511.
- Edwards, N. and Marsh, R. (2005) Uncertainties due to transport-parameter sensitivity in an efficient 3-D ocean-climate model. *Climate Dynamics*, **24**, 415–433.
- Fish, P. *et al.* (2010) LLWR Environmental Safety Case: Forecasting the Development of the Cumbrian Coastline in the Vicinity of the LLWR Site. *Quintessa Report QRS 1443X 1*, Version 1.
- French, H.M. (2007) *The Periglacial Environment*. John Wiley & Sons, Chichester, UK.
- Hays, J.D., Imbrie, J. and Shackleton, N.J. (1976) Variations in the earth's orbit: Pacemaker of the Ice Ages. *Science*, **194**, 1121–1132.
- IPCC (1990) Contribution of Working Group I to the First Assessment Report of the Intergovernmental Panel on Climate Change. *Climate Change: The IPCC Scientific Assessment* (J.T. Houghton, G.J. Jenkins, and J.J. Ephraums, editors), 410 pp. Cambridge University Press, Cambridge, UK, and New York, USA.
- IPCC (1995) Contribution of Working Group I to the Second Assessment Report of the Intergovernmental Panel on Climate Change. *Climate Change 1995: The Science of Climate Change* (J.T. Houghton, L.G. Meiro Filho, B.A. Callander, N.R.P. Harris, A. Kattenberg and K. Maskell, editors), 532 pp. Cambridge University Press, Cambridge, UK, and New York, USA.
- IPCC (2001) Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change. *Climate Change 2001: The Scientific Basis* (J.T. Houghton *et al.*, editors), 786 pp. Cambridge University Press, Cambridge, UK, and New York, USA.
- IPCC (2007) Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. *Climate Change 2007: The Physical Science Basis* (S. Solomon *et al.*, editors) 940 pp. Cambridge University Press, Cambridge, UK, and New York, USA.

- Kawamura, K. *et al.* (2007) Northern Hemisphere forcing of climatic cycles in Antarctica over the past 360,000 years. *Nature*, **448**, 912–914.
- Kohfeld, K.E. and Ridgwell, A. (2009) Glacial-interglacial variability in atmospheric CO₂. Pp. 251–286 in: *Surface Ocean-Lower Atmosphere Processes* (C. Quere and E.S. Saltzman, editors). American Geophysical Union, Washington DC, USA.
- Laskar, J., Robutel, P., Joutel, F., Gastineau, M., Correia, A.C.M. and Levrard, B. (2004) A long-term numerical solution for the insolation quantities of the Earth. *Astronomy & Astrophysics*, **428**, 261–285.
- Lenton, T.M. and Britton, C. (2006) Enhanced carbonate and silicate weathering accelerates recovery from fossil fuel CO₂ perturbations. *Global Biogeochemical Cycles*, **20**. doi: 10.1029/2005GB002678.
- LLWR (2011) Environmental Safety Case – Main Report. *LLWR/ESC/(R11)10016*.
- Luthi, D. *et al.* (2008) High-resolution carbon dioxide concentration record 650,000–800,000 years before present. *Nature*, **453**, 379–382.
- Martinez-Boti, M.A. *et al.* (2015) Plio-Pleistocene climate sensitivity evaluated using high-resolution CO₂ records. *Nature*, **518**, 49.
- Milankovitch, M. (1941) Canon of Insolation and the Ice-Age Problem. *Royal Serbian Academy Special Publication*, **132**. [Israel Program for Scientific Translations, Jerusalem (1969)].
- Paillard, D. (2001) Glacial cycles: Toward a new paradigm. *Reviews of Geophysics*, **39**, 325–346.
- Petit, J.R. *et al.* (1999) Climate and atmospheric history of the past 420,000 years from the Vostok ice core, Antarctica. *Nature*, **399**, 429–436.
- Ridgwell, A. and Hargreaves, J.C. (2007) Regulation of atmospheric CO₂ by deep-sea sediments in an Earth system model. *Global Biogeochemical Cycles*, **21**. doi:10.1029/2006GB002764.
- Ridgwell, A. *et al.* (2007) Marine geochemical data assimilation in an efficient Earth System Model of global biogeochemical cycling. *Biogeosciences*, **4**, 87–104.
- Rogner, H.H. (1997) An assessment of world hydrocarbon resources. *Annual Review of Energy and the Environment*, **22**, 217–262.
- Ruddiman, W.F. (2006) Orbital changes and climate. *Quaternary Science Reviews*, **25**, 3092–3112.
- Sitch, S. *et al.* (2008) Evaluation of the terrestrial carbon cycle, future plant geography and climate-carbon cycle feedbacks using five Dynamic Global Vegetation Models (DGVMs). *Global Change Biology*, **14**, 2015–2039.
- SKB (2011) Long-term safety for the final repository for spent nuclear fuel at Forsmark. Main report of the SR-Site project. *SKB Report TR-11-01*.
- SKB (2013) Climate and climate-related issues for the safety assessment SR-PSU. *SKB Report TR-13-05*.
- Thorne, M.C. (2013) Book review of J.S. Stuckless (editor). *Annals of Nuclear Energy*, **53**, 545–549.
- Turley, C. *et al.* (2010) Carbon uptake, transport and storage by oceans and the consequences of change in carbon capture and storage (CCS). Pp. 240–284 in: *Issues in Environmental Science and Technology (IEST)* (R.M. Harrison and R.E. Hester, editors). Royal Society of Chemistry.
- Williams, R.G., Goodwin, P., Ridgwell, A. and Woodworth, P.L. (2012) How warming and steric sea level rise relate to cumulative carbon emissions. *Geophysical Research Letters*, **39**. doi: 10.1029/2012GL052771.