

Long-term effectiveness and consequences of carbon dioxide sequestration

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One proposal for the mitigation of ongoing global warming is the sequestration of carbon dioxide extracted at combustion sites or directly from the air^{1,2}. Such sequestration could help avoid a large rise in atmospheric CO₂ concentration from unchecked use of fossil fuels, and hence extreme warming in the near future^{3,4}. However, it is not clear how effective different types of sequestration and associated leakage are in the long term, and what their consequences might be. Here I present projections over 100,000 years for five scenarios of carbon sequestration and leakage with an Earth system model⁵. Most of the investigated scenarios result in a large, delayed warming in the atmosphere as well as oxygen depletion, acidification and elevated CO₂ concentrations in the ocean. Specifically, deep-ocean carbon storage leads to extreme acidification and CO₂ concentrations in the deep ocean, together with a return to the adverse conditions of a business-as-usual projection with no sequestration over several thousand years. Geological storage may be more effective in delaying the return to the conditions of a business-as-usual projection, especially for storage in offshore sediments. However, leakage of 1% or less per thousand years from an underground stored reservoir, or continuous re-sequestration far into the future, would be required to maintain conditions close to those of a low-emission projection with no sequestration.

Massive application of carbon dioxide capture and storage (CCS) has become a prominent proposal for mitigating global warming^{1,2}. Recent discussions of CCS have been centred on its motivation, feasibility and cost compared with other mitigation strategies^{2,6–8}. Concerns have also been raised about possible consequences of storing CO₂ in the ocean and of leakage of CO₂ stored in geological formations^{1,9}. Here I compare the effectiveness as well as Earth system consequences of CO₂ leakage for sequestration in the ocean and in onshore and offshore geological formations. For this purpose I make long projections with the reduced-form, Danish Center for Earth System Science (DCESS) model⁵.

As a target for desirable future emissions, I designed an emission scenario for avoiding strong global warming (Fig. 1, AG scenario; see the Methods section). For a projection forced with AG emissions, atmospheric *p*CO₂ peaks at 408 ppm by AD 2038 and mean atmosphere warming peaks at 2.03 °C by AD 2102 (as compared with AD 1765; Fig. 2a,c). As actual fossil-fuel emissions may continue to rise during the next decade, these and related results¹⁰ indicate that greater global emission reductions than those assumed here may be needed during the rest of this century to avoid warming greater than 2 °C above pre-industrial conditions. Still greater reductions would be required to meet this goal if actual climate sensitivity is greater than the model value of 3 °C. In the AG

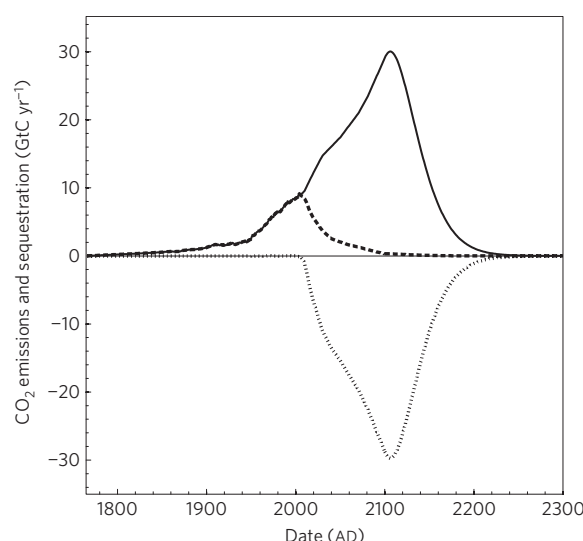


Figure 1 | Carbon dioxide emission and sequestration scenarios. The solid and dashed lines are total anthropogenic CO₂ emissions from fossil fuels and land-use change for the A2 and AG scenarios, respectively (see the Methods section). The dotted line is the CO₂ sequestration scenario calculated such that A2 emissions together with CO₂ sequestration yield AG emissions.

projection, atmosphere warming does not fall below 1 °C until AD 4100, owing mainly to slow ocean CO₂ uptake.

As an example for possible future, ‘business-as-usual’ emissions, I use the extended A2 emission scenario¹¹ (Fig. 1; see the Methods section). A projection forced with A2 emissions shows atmosphere warming above 5 °C by AD 2130 (Fig. 2c), ocean warming above 3 °C by AD 4010 (Fig. 2e) and tenfold expansion of ocean ‘dead zone’ volume by AD 3000 (Fig. 2f; ref. 11). In the A2 projection, greater warming from higher emissions induces positive carbon feedbacks, such as less CO₂ dissolution in the ocean and more soil remineralization¹¹, that further extend the duration of strong warming. As a result, atmosphere warming does not fall below 3 °C until AD 9550. Three million years ago, mean global temperature was about 2–3 °C warmer than pre-industrial conditions and sea level was about 20–30 m higher¹². Such a large sea level rise can be explained only by partial melting of the Greenland and Antarctic ice sheets. Given the size and duration of global warming in the A2 projection, it is likely that large sea level rise from ice-sheet melting would also be a consequence of the A2 emissions. The extended atmospheric warming of the A2 projection also promotes greater ocean warming, leading to enhanced ‘dead zones’ from decreased

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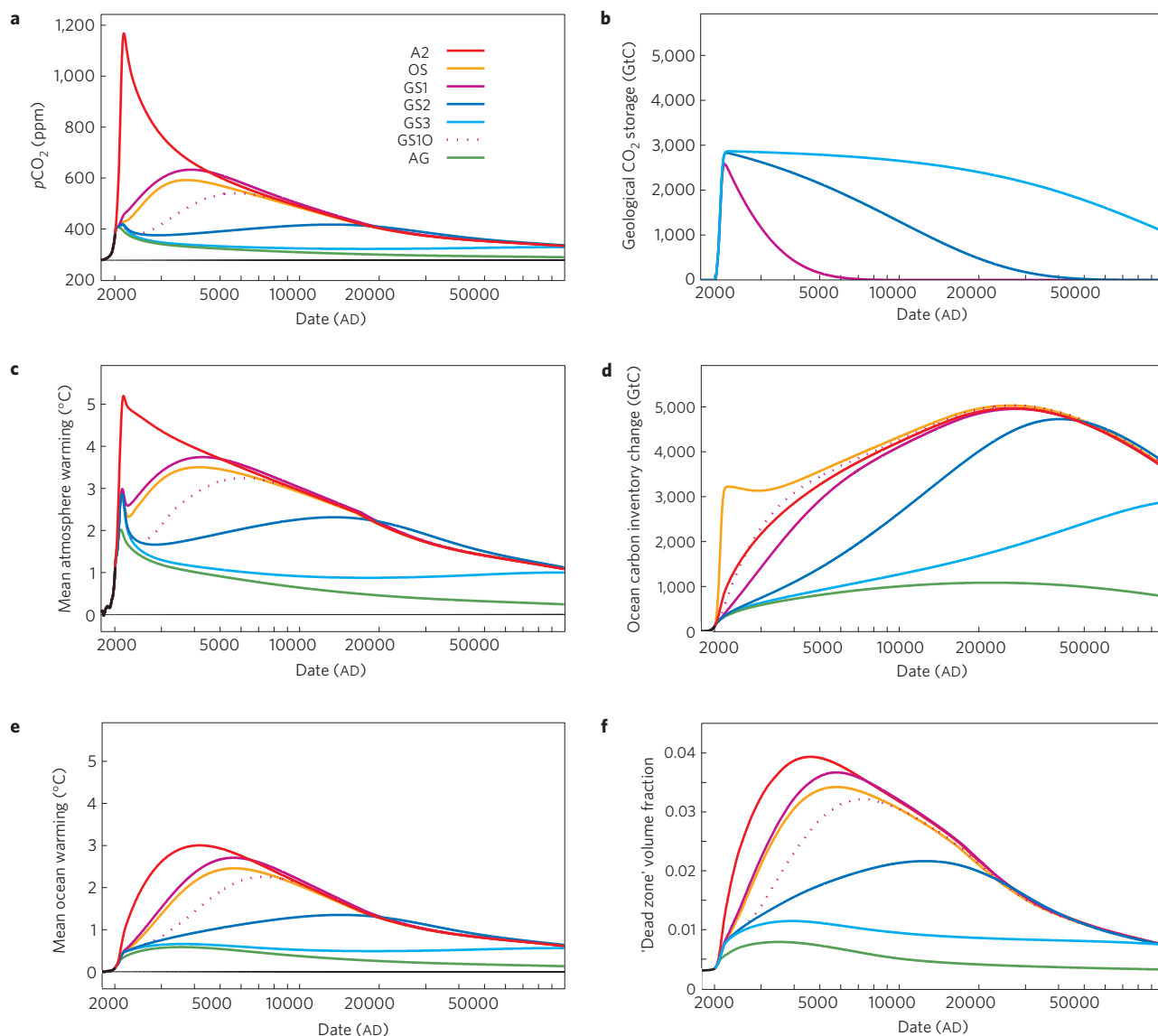


Figure 2 | Long-term Earth system projections for different CO₂ emission and storage scenarios. Note the logarithmic timescale. The solid red and green lines are for projections without CO₂ sequestration (A2 and AG), forced by A2 and AG emissions, respectively; all other projections include CO₂ sequestration and are forced by A2 emissions (Fig. 1). The yellow line is for an ocean storage projection (OS). The violet, dark blue and light blue lines are for onshore geological storage projections (GS1, GS2 and GS3) with leakage rates of 1% per 10, 100 and 1,000 years, respectively. The dotted violet line is for an offshore geological storage projection (GS10) with a leakage rate of 1% per 10 years. **a**, Partial pressure of atmospheric CO₂. **b**, Geologically stored, CO₂ reservoirs for the GS1, GS2 and GS3 projections. **c**, Mean atmosphere temperature change from the pre-industrial model value of 15 °C. **d**, Change in ocean carbon inventory from a pre-industrial model value of 37,108 GtC. **e**, Mean ocean temperature change from the pre-industrial model value of 4.03 °C. **f**, Fraction of global ocean volume with dissolved oxygen concentrations $\leq 10 \mu\text{mol kg}^{-1}$, considered 'dead zones' for higher life forms. Fractions are calculated as in ref. 11.

oxygen solubility as well as greater sea level rise from thermal expansion. Furthermore, ocean heating would probably lead to methane release from methane hydrate melting in ocean sediments and still more long-term global warming not considered here¹³.

A future pathway with continued high fossil-fuel use but with fully developed CCS can be formulated as gross CO₂ emissions from the A2 scenario combined with carbon capture at power plants, direct extraction of CO₂ from the atmosphere¹⁴ and subsequent sequestration such as to reduce net CO₂ emissions to those of the AG scenario (Fig. 1). CCS implementation requires considerable energy use¹ such that less net energy is available for society in this pathway than in the A2 scenario although gross CO₂ emissions are the same for both. All emissions other than CO₂, for example those of methane, nitrous oxide and black carbon, are taken to follow the

A2 scenario. In this pathway, total gross and net CO₂ emissions are 3,721 GtC and 822 GtC such that 2,899 GtC is sequestered over two centuries. Such immense sequestration probably does not exceed the storage capacity in geological formations¹⁵ nor in the ocean and may be technologically feasible over the timescale considered¹⁶. Below I consider long projections for this sequestration pathway applied to five different storage/leakage cases.

An ocean storage projection, OS, assumes CO₂ injection at depths between 2,500 and 3,500 m of the low-mid latitude ocean and total dissolution at the depths injected. Such sequestration mitigates much of the initial warming but this warming still exceeds that of the AG projection by about 1 °C because of greater net radiative forcing from non-CO₂ greenhouse gases (Fig. 2c). Ocean storage leads to a mean pH decrease at 3,000 m depth of

Table 1 | Amount and timing of maximum changes of projection results for ocean mean pH and $p\text{CO}_2$ in the surface layer and at 3,000 m depth.

Projection	Ocean surface layer				3,000 m depth			
	ΔpH	Date (AD)	$\Delta p\text{CO}_2$ (ppm)	Date (AD)	ΔpH	Date (AD)	$\Delta p\text{CO}_2$ (ppm)	Date (AD)
A2	−0.529	2175	876	2175	−0.332	4015	700	4215
OS	−0.269	3545	328	3725	−1.082	2165	6,882	2155
GS1	−0.292	3735	353	3915	−0.306	5465	635	5745
GS1O	−0.219	5405	262	5735	−0.450	2915	1,060	2945
GS2	−0.160	2145	156	2145	−0.171	11315	321	13215
GS3	−0.158	2115	153	2115	−0.088	3385	148	3535
AG	−0.134	2065	126	2065	−0.081	3285	119	3405

more than one unit but surface-ocean pH decreases much less than in the A2 projection. In addition, this injection causes a mean $p\text{CO}_2$ increase at 3,000 m depth of almost 7,000 ppm (Table 1). Both increased acidity and $p\text{CO}_2$ can adversely affect deep-sea life^{17,18}. Atmosphere warming drops slightly after the initial warming peak but rises again to 3.5 °C by AD 4000 as some sequestered CO_2 leaks to the atmosphere over ocean circulation and mixing timescales (Fig. 2c). Subsequently, the OS projection follows closely the A2 projection and suffers similar large ‘dead zone’ volume increase (Fig. 2f) and probable large sea level rise. Ocean CO_2 injection also leads to very low carbonate ion concentration ($<15 \mu\text{mol kg}^{-1}$) at depth and enhanced CaCO_3 dissolution in sediments that acts to suppress oceanic, and thereby atmospheric, CO_2 . However, ocean circulation and mixing limits sediment exposure to such low carbonate ion concentrations to times much shorter than CaCO_3 dissolution timescales and not so much extra CaCO_3 is dissolved. By AD 10000 this has led to an extra ocean carbon inventory of 115 GtC and an extra atmospheric $p\text{CO}_2$ drawdown of only 5 ppm compared with the A2 projection (Fig. 2a,d).

Geological storage sites for CO_2 vary greatly in type and location whereby deep saline formations provide by far the largest potential storage capacity^{1,15}. Great heterogeneity in storage sites and conditions at them makes direct modelling of the total leakage of geologically stored CO_2 very difficult. Thus, I base my projections on the Intergovernmental Panel on Climate Change (IPCC) conclusion¹ that the fraction retained in appropriately selected and managed geological reservoirs is very likely to exceed 99% over 100 years and is likely to exceed 99% over 1,000 years. In addition, as a ‘worst case’ leakage scenario I also consider 99% retention over 10 years. The time evolution of the geologically stored, CO_2 reservoir, $G(t)$, consistent with these assumptions may be expressed as

$$\frac{dG(t)}{dt} = S(t) - aG(t)$$

where $S(t)$ is the sequestration rate (Fig. 1) and $aG(t)$ is the leakage rate, whereby a is an inverse-leakage timescale. I consider three onshore geological storage projections, GS1, GS2 and GS3, with leakage to the atmosphere and a equal to 1×10^{-3} , 1×10^{-4} and $1 \times 10^{-5} \text{ yr}^{-1}$ from the above (Fig. 2b).

As for the ocean storage projection, all onshore geological storage projections show mitigation of much of the initial warming of the A2 projection (Fig. 2). Later on, ‘rapidly leaking’ projection GS1 has higher $p\text{CO}_2$ and even more atmosphere and ocean warming and ocean oxygen depletion than the OS projection. Indeed, after several thousand years, $p\text{CO}_2$ and atmosphere warming of the GS1 projection even exceed those of the A2 projection with no sequestration. This is consistent with and extends earlier results for shorter projections with faster leakage⁴. The extra, long-term warming extends far into the future, fostering more ocean warming and ocean oxygen depletion than in the A2 projection then.

‘Moderately leaking’ projection GS2 shows greatly reduced $p\text{CO}_2$, atmosphere and ocean warming, ocean acidification and ocean ‘dead zone’ volume for more than 10,000 years, compared with the A2 projection (Fig. 2). Subsequently, these properties increase to exceed those of the A2 projection. For ‘weakly leaking’ projection GS3, atmospheric $p\text{CO}_2$ drops below 350 ppm by AD 2890 and remains below this level (Fig. 2a). Likewise, atmospheric warming decreases rapidly to levels slightly above present-day global warming and initial evolutions of ocean warming, acidification and oxygen depletion are similar to the AG projection. Subsequently, the GS3 projection diverges towards the A2 projection, exhibiting slow enhancement of ‘dead zone’ volume during the last two-thirds of the projection. A ‘best case’ projection with no leakage from geological storage would converge to very near the AG projection after several hundred years.

I also consider a projection, GS1O, for storage in deep-ocean sediments, with a as for GS1 but with leakage to the ocean, distributed as in the OS projection. The GS1O projection also shows initial mitigation (Fig. 2). In this ‘rapidly leaking’ projection, atmosphere warming and surface-ocean acidification are significantly decreased and delayed compared with the GS1 projection, although the GS1O projection also approaches the A2 projection later on (Fig. 2; Table 1). Furthermore, as the timescale of this leakage is much greater than that of sequestration, the impact of the GS1O projection on deep-ocean conditions is much less than for the OS projection (Table 1). For these and other reasons such as potential CO_2 immobilization in deep-sea sediments¹⁹, CO_2 storage in offshore sediments may be preferable to onshore geological storage or ocean storage. However, the storage capacity of suitable offshore sediment reservoirs is uncertain²⁰.

CO_2 storage would have to last for tens of thousands of years to avoid strong, delayed global warming and marked Earth system changes such as vast expansion of ocean ‘dead zones’. This is much longer than 4,000 years claimed earlier²⁰ from an apparent misinterpretation of carbon–climate models results²¹. Deep-ocean sequestration cannot meet this storage criterion and brings with it serious ecological side effects and hardly any advantage from enhanced CaCO_3 dissolution. It is not clear whether geological sequestration could meet the above criterion, although storage in offshore sediments could help if suitable storage sites could be found. Even if massive, ‘safe’ sequestration could be implemented very soon, great reductions in net radiative forcing from non- CO_2 greenhouse gases and black carbon, as in the AG scenario, would also be needed to avoid warming greater than 2 °C above pre-industrial levels in this century.

The carbon sequestration pathway presented here posits great human effort in the next two hundred years but subsequent human inaction in the spirit of ‘out of sight, out of mind’. Alternatively, long-term leakage from the ocean or geological reservoirs could be actively countered by re-sequestration to stabilize climate at some desired level. However, there are serious concerns connected with

this. First, it would be difficult to gauge the global leakage rate that would have to be matched by the resequstration rate. Long-term monitoring of atmospheric CO₂ concentrations would probably be the best way to address this but natural carbon-cycle fluctuations would complicate this approach. Second, resequstration would have to be carried out over many thousands of years, a burden for future society not unlike that of long-term management of nuclear waste. By greatly limiting carbon emissions in our time, we could reduce the need for massive CO₂ sequestration and thus reduce unwanted consequences and burdens over many future generations from the leakage of sequestered CO₂.

Methods

The DCESS model⁵ includes atmosphere, ocean, ocean sediment, land biosphere and lithosphere modules. Sea ice and snow cover are diagnosed from the atmospheric temperature distribution but there is no ice-sheet module. Earth system data and complex model results were used to calibrate the model to a pre-industrial (AD 1765), steady-state climate with a long-term climate sensitivity of 3.0 °C for a doubling of atmospheric pCO₂. (An instantaneous pCO₂ change from 278 to 556 ppm from this steady state forces mean atmospheric warming of 1.53, 2.58, 2.87 and 3.00 °C after 10, 100, 1,000 and 10,000 years of model integration.) The model is well suited for long projections because it includes CO₂–climate feedbacks in the land biosphere and ocean, organic and carbonate carbon cycling in the ocean sediment and climate-dependent rock weathering.

All 100,000-year projections start with initial conditions from the pre-industrial, steady state and are forced initially by prescribed anthropogenic emissions of CO₂, CH₄ and N₂O and prescribed radiative forcing from other (minor) greenhouse gases, aerosols and solar-radiation changes⁵. Fossil-fuel CO₂ emissions from AD 1765 to 2004 are from ref. 22. Land-use-change CO₂ emissions for AD 1860–1950 are from ref. 23. Linear interpolation from a zero value at AD 1765 is used before AD 1860. For AD 1950–1980, values from ref. 23 are multiplied by a factor varying linearly from 1 to 0.73 over this period and, for AD 1980–2000, are multiplied by 0.73 to yield mean values of about 1.4 and 1.6 GtC yr⁻¹ for the 1980s and 1990s, respectively, in agreement with the recent IPCC assessment²⁴. Anthropogenic CH₄ emissions from AD 1860 to 1994 are from ref. 25 minus a value of 0.04 Gt(CH₄) yr⁻¹, an estimate for anthropogenic, pre-industrial emissions included in the model land-biosphere emissions⁵. Linear interpolation from a zero value at AD 1765 is used before AD 1860. Values for AD 1995–2000 are assigned the 'corrected' AD 1994 value. We take anthropogenic N₂O emissions to be proportional to those of CH₄ (in units mol s⁻¹) with a proportionality constant of 0.007 from a model fit to N₂O observations. Net radiative forcing from AD 1765 to 2000 resulting from other (minor) greenhouse gases, aerosols and solar-radiation changes is calculated from results in ref. 26 by subtracting radiative forcing resulting from CO₂, CH₄ and N₂O from total radiative forcing. For the total radiative forcing described above, the projections reproduce observed evolutions up to AD 2000 of key climate metrics including atmosphere and ocean warming, atmospheric gas contents and ocean and land biosphere CO₂ uptakes⁵.

The 'business-as-usual' A2 scenario and all carbon sequestration scenarios use fossil-fuel and land-use-change CO₂ emissions for the periods AD 2004–2100 and AD 2000–2100, respectively, from the IPCC SRES A2 scenario²⁷. After AD 2100, total CO₂ emissions are extrapolated using polynomial functions to tail off by the early twenty-third century. It is assumed that by then accessible fossil-fuel resources will have become limited and/or alternative energy sources will have been adopted. Anthropogenic CH₄ emissions for the period AD 2000–2100 are taken from the SRES A2 scenario and after AD 2100 are extrapolated as above. Anthropogenic N₂O emissions from AD 2000 onward are estimated from the CH₄ emissions as described above. Radiative forcings from other (minor) greenhouse gases and aerosols for the period AD 2000–2100 were compiled from the SRES A2 scenario and after AD 2100 are extrapolated as above. With the exception of fossil-fuel emissions, the low-emission, AG scenario uses emissions and radiative forcings after AD 2000 as described above but based on the SRES B1 scenario²⁷. For the AG scenario, fossil-fuel emissions are taken to rise after AD 2004, level off by AD 2010 and subsequently fall to meet global emission reduction targets of 20% by 2020 and 60% by AD 2050 (compared with AD 1990). For simplicity, solar forcing is maintained constant after AD 2000 in all scenarios. This is motivated in part by the fact that orbital forcing will be weaker than normal for the next 100,000 years²⁸.

Matlab programs for running the DCESS model can be downloaded at www.dcess.dk and include forcing functions and plotting routines needed to reproduce the results reported here.

Received 2 December 2009; accepted 24 May 2010;
published online 27 June 2010

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Acknowledgements

I thank P. Brewer, R. Garreaud, S. M. Olsen and J. O. P. Pedersen for comments. Part of this research was carried out at the Department of Geophysics, University of Chile, Santiago.

Additional information

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