

## Impact of deep-ocean carbon sequestration on atmospheric CO<sub>2</sub> and on surface-water chemistry

L. D. Danny Harvey

Department of Geography, University of Toronto, Toronto, Ontario, Canada

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[1] The effectiveness of injection into the ocean of CO<sub>2</sub> produced from the use of fossil fuels is investigated using a coupled climate-carbon cycle model. Four fossil fuel emission scenarios are considered, in which emissions peak at 28 Gt C/yr in 2100 (scenario 1), 9 Gt C/yr in 2050 (scenario 2), or decrease to zero by 2100 or 2075 (scenarios 3 and 4). Sequestration sufficient to reduce the net emissions for scenarios 1–3 to that in scenario 4 is considered. Sequestration in scenario 1 results in a CO<sub>2</sub> concentration of 857 ppmv by 3100, compared to a peak of 1614 ppmv without sequestration, and slows the accumulation of atmospheric CO<sub>2</sub>. The supersaturation of the non-polar mixed layer with respect to calcite decreases from 486% pre-industrial to about 250%, while mixed layer pH decreases by 0.66. Only for scenario 3 is sequestration sufficient to limit the peak impact on atmospheric CO<sub>2</sub> and mixed layer chemistry to the impacts obtained in scenario 4. *INDEX TERMS*: 4806 Oceanography: Biological and Chemical: Carbon cycling; 1615 Global Change: Biogeochemical processes (4805); 1620 Global Change: Climate dynamics (3309); 1635 Global Change: Oceans (4203). *Citation*: Harvey, L. D. D., Impact of deep-ocean carbon sequestration on atmospheric CO<sub>2</sub> and on surface-water chemistry, *Geophys. Res. Lett.*, 30(5), 1237, doi:10.1029/2002GL016224, 2003.

### 1. Introduction

[2] This paper assesses the impact on atmospheric CO<sub>2</sub> and surface water chemistry of direct injection of CO<sub>2</sub> from fossil fuels into the ocean at a depth of 3000 m. There is increasing interest in the capture of CO<sub>2</sub> that would otherwise be emitted to the atmosphere and its injection into either deep geological reservoirs or the deep ocean, a process known as “carbon sequestration”. Carbon dioxide could either be separated from the flue gases of electric power plants after the combustion process, or separated during the gasification of coal or biomass prior to combustion or during the conversion of natural gas to hydrogen fuel.

[3] Recent advances in CO<sub>2</sub> separation techniques, and the costs and energy penalty associated with the separation, concentration, transport, and disposal of CO<sub>2</sub> are discussed in detail in *Riemer et al.* [1999]. *Williams* [1998] provides further details concerning carbon sequestration in conjunction with the production of hydrogen fuel from coal, natural gas, and biomass. Estimates of the amount of carbon that could be safely and permanently sequestered in deep aquifers, depleted oil and gas fields, and coal beds range

from as low as 300 Gt C to as high as 13000 Gt C. Estimates of this sequestration potential beyond a few hundred Gt C are highly speculative.

[4] The oceans have the ability to permanently store several thousand Gt C. Carbon dioxide that is sequestered in the ocean would be liquefied first and injected at a depth of 3000 m or so, either from seabed pipes or from pipes suspended from moving ships. Either option poses significant technical challenges, but these are not believed to be insurmountable [*Nihous*, 1997]. However, the fraction of injected carbon that would remain in the ocean in steady state is no more than about 0.85 (prior to the partial dissolution of carbonate sediments), depending on the cumulative injection. This is the same as the fraction that would eventually be absorbed by the oceans following direct emission into the atmosphere. Furthermore, injection of large amounts of CO<sub>2</sub> would likely have profound effects on marine life by decreasing the pH of seawater and significantly reducing the extent of supersaturation of surface water with respect to calcium carbonate. Partial dissolution of carbonate sediments would neutralize the decrease in pH and allow the oceans to store another 5–10% of the originally injected CO<sub>2</sub>, but only after a lag of several thousand years [*Walker and Kasting*, 1992].

### 2. Model Used Here

[5] The coupled climate-carbon model of *Harvey and Huang* [2001] and *Harvey* [2001] is used here to assess the impacts of oceanic carbon sequestration. This is a one-dimensional upwelling diffusion model of the ocean, except that polar regions with convective mixing are treated separately from non-polar regions. The model has been extensively tested against observed steady state and transient variations in a number of tracers. The oceanic part of the carbon cycle model contains the 7-component carbonate chemistry equations given in *Peng et al.* [1987], and so is suitable for assessing the global average effect of the uptake of CO<sub>2</sub> on the pH and concentration of dissolved carbonate ion (CO<sub>3</sub><sup>2-</sup>) in the surface layer of the ocean. The model is driven by anthropogenic emissions of CO<sub>2</sub>, from which it computes the uptake by the terrestrial biosphere and oceans. The accumulation of CO<sub>2</sub> (and other greenhouse gases) leads to an increase in atmospheric temperature, which alters the subsequent uptake of CO<sub>2</sub> through a variety of climate-carbon cycle feedbacks. For the experiments presented here, the equilibrium surface warming for a CO<sub>2</sub> doubling (the so-called climate sensitivity) is specified to be 2°C.

[6] Elsewhere (L. D. D. Harvey, Declining temporal effectiveness of carbon sequestration: Implications for com-

pliance with the United Nations Framework Convention on Climate Change, submitted to *Climatic Change*, 2002, hereinafter referred to as LDDH), the impulse response of the carbon cycle model to the sudden injection of 1 Gt C into the atmosphere and the deep ocean at depths of 500 and 3000 m is shown to be comparable to that obtained with three-dimensional ocean carbon cycle models [Stocker *et al.*, 1994; Caldeira *et al.*, 2001]. Two hundred years after injection into the atmosphere, 70% of the injected carbon is taken up by the ocean, while after 2000 years (and in steady state), about 87% is taken up by the ocean and 13% remains in the atmosphere. When carbon is injected directly into the ocean, some of the carbon flows out of the ocean and into the atmosphere so that, in steady state, the atmosphere:ocean proportions are the same as when the CO<sub>2</sub> is injected directly into the atmosphere.

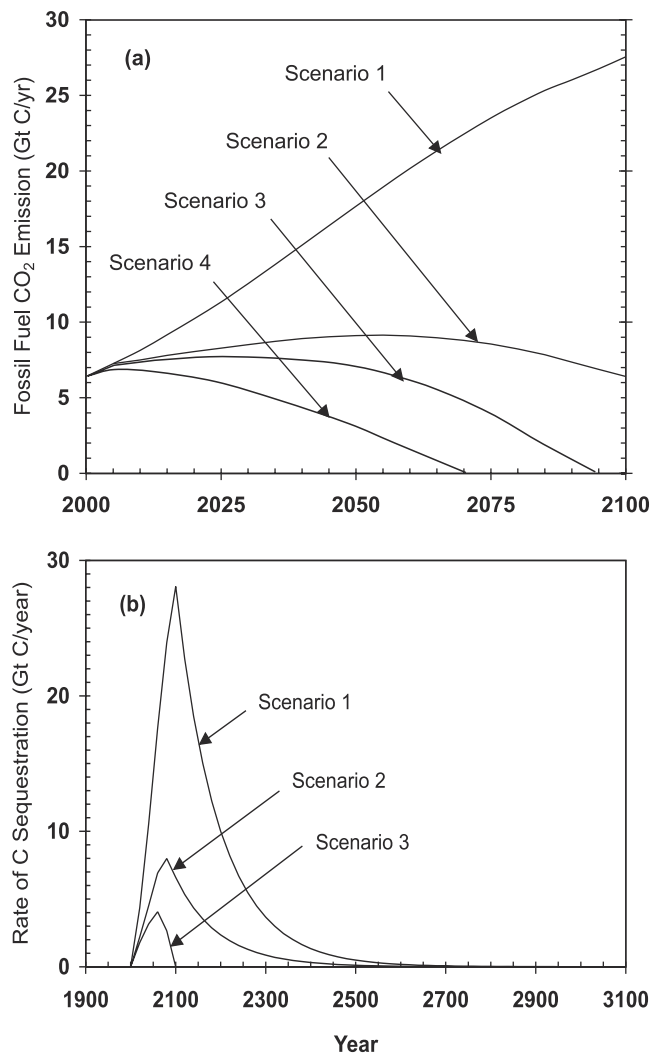
### 3. Emission and Sequestration Scenarios

[7] Four fossil fuel emission scenarios, developed by LDDH, are shown in Figure 1a and used here. Scenario 1 is a business-as-usual scenario in which emissions reach 28 Gt C/yr by 2100, scenario 2 entails modest restraints on fossil fuel emissions, and in scenarios 3 and 4, emissions decrease to zero by 2100 and 2075 respectively. In scenarios 1 and 2, emissions are assumed to decrease by 1% per year after 2100. Land use CO<sub>2</sub> emissions are assumed to reach zero by 2050 in all cases, while cement emissions double between now and 2050, then decrease to a final value of 0.21 Gt C/yr. Three sequestration scenarios are considered, sufficient to reduce the fossil fuel emissions in scenarios 1, 2, and 3 to those that occur in scenario 4. The rates of carbon sequestration for these three cases are shown in Figure 1b. The cumulative sequestration for these three scenarios is 4150 Gt C, 1148 Gt C, and 237 Gt C, respectively.

### 4. Results

[8] Figure 2a shows the variation in atmospheric CO<sub>2</sub> concentration for scenarios 1 and 4. In scenario 4, atmospheric CO<sub>2</sub> peaks at 430 ppmv, while in scenario 1 it peaks at 1614 ppmv. Also shown in Figure 2a is the variation in atmospheric CO<sub>2</sub> when an amount of carbon equal to the difference in fossil fuel emissions between scenarios 1 and 4 is injected into the ocean at a depth of 3000 m. The CO<sub>2</sub> concentration curve begins to diverge from that for scenario 4 by 2200, due to an efflux into the atmosphere of some of the injected CO<sub>2</sub>. By 3100 this efflux has reached 996 Gt C (24% of the cumulative injection). The efflux fraction obtained here is greater than the impulse response steady-state value of 13%, as the latter pertains to a small pulse in the pre-industrial carbon cycle. The efflux is sufficient to produce a CO<sub>2</sub> concentration nearly double the peak concentration in scenario 4 (857 ppmv vs. 430 ppmv). Thus, for a high business-as-usual scenario (but well within the range of what has been considered by the scenario community), massive injection of carbon into the oceans fails to prevent a significant increase in atmospheric CO<sub>2</sub> beyond that already experienced in extreme emission-reduction scenarios.

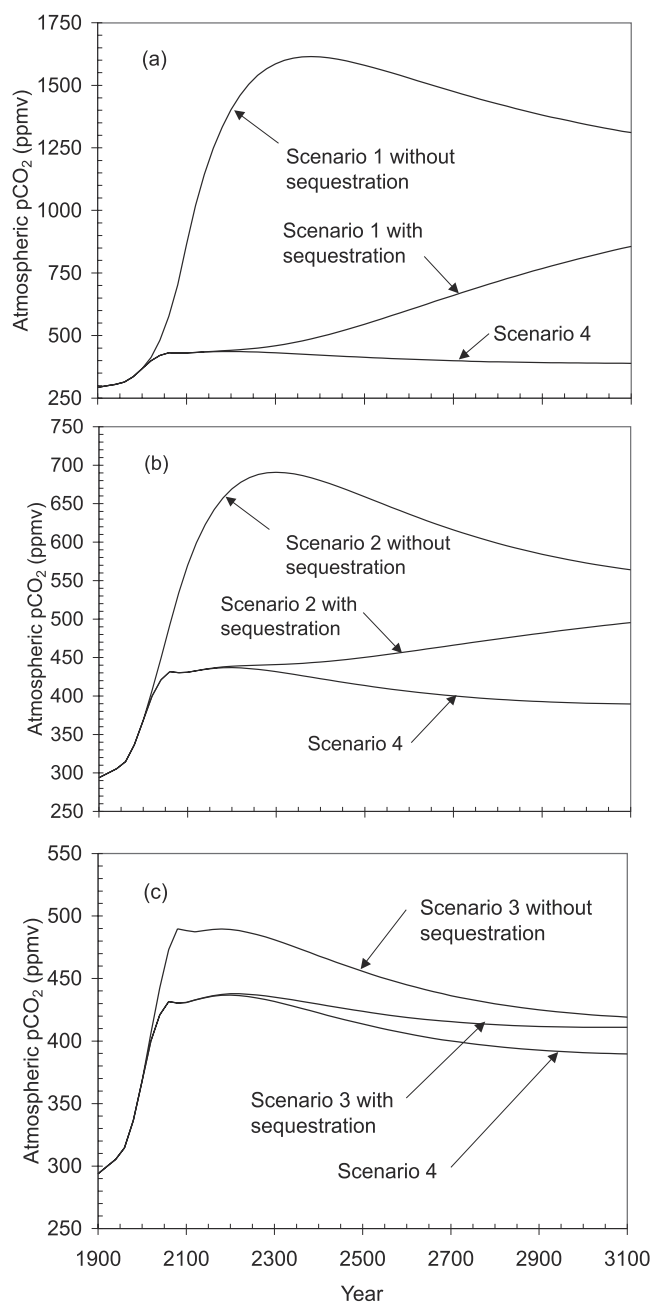
[9] Figures 2b and 2c compare scenarios 2 and 3, respectively, with scenario 4 for cases with and without sequestration of the difference in fossil fuel emissions. In



**Figure 1.** (a) Fossil fuel emission scenarios considered here. (b) Carbon sequestration scenarios.

these scenarios, the efflux fraction by 3100 is smaller than in scenario 1 (about 20% and 19%, respectively). However, sequestration ends sooner in these scenarios than in scenario 1, so the increase in CO<sub>2</sub> concentration by 3100 for the cases with sequestration is a larger fraction of the increase without sequestration, than for scenario 1. The long-term CO<sub>2</sub> concentration for cases with and without sequestration are the same; the impact of sequestration is to avoid the peak concentration prior to the longterm decline in atmospheric CO<sub>2</sub> seen in the cases without sequestration. Only in scenario 3 (fossil fuel use eliminated by 2100) is sequestration in the ocean sufficient to maintain an atmospheric CO<sub>2</sub> concentration at or below the peak concentration (430 ppmv) obtained in scenario 4. In LDDH it is argued that compliance with the objective of the United Nations Framework Convention in Climate Change requires limiting (or returning) the atmospheric CO<sub>2</sub> concentration to a value in the range 350–450 ppmv, and the effectiveness of land and/or ocean carbon sequestration in achieving this, relative to scenarios 3 and 4, is analyzed.

[10] Figure 3 shows the impact on the degree of supersaturation of the non-polar mixed layer with respect to

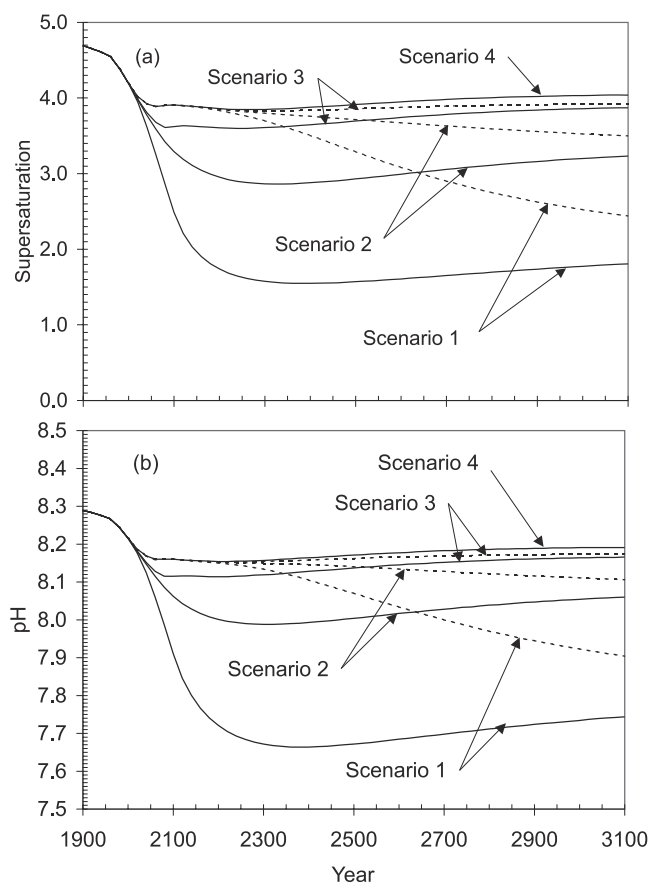


**Figure 2.** Atmospheric CO<sub>2</sub> concentration for emission scenarios 1 and 4 (a), 2 and 4 (b) and 3 and 4 (c).

calcite, and of pH. The supersaturation falls from a pre-industrial value of 486% (i.e.: 4.86 times saturation) to 155%, 286%, 360%, and 381% for scenarios 1, 2, 3, and 4, respectively. Even the smallest of these decreases is likely to adversely affect coral reef and marine calcareous microorganisms by reducing calcification rates [Kleypas, 1999; Wolf-Gladrow et al., 1999; Langdon et al., 2000; Riebesell et al., 2000]. The impact on marine ecology of the more extreme reductions would likely be severe. The corresponding reductions in pH are by 0.66, 0.30, 0.19, and 0.14 units, respectively. The larger of these pH reductions would likely adversely affect both calcareous and non-calcareous marine organisms, independently of the decrease in calcite supersaturation. As with the impact on atmospheric CO<sub>2</sub>, the

response of supersaturation and pH with sequestration converges toward the response without sequestration near the end of the simulation. Beyond 3100, dissolution of CaCO<sub>3</sub> sediments would begin to have an important (and growing) effect in reducing the change in mixed layer supersaturation and pH. Thus, the responses seen in Figure 3 by 3100 for the cases with carbon sequestration are comparable to the peak response that can be expected, given dissolution of CaCO<sub>3</sub> sediments. However, these responses are a significant fraction of the peak responses seen for the cases without carbon sequestration.

[11] In assessing the impact of deep ocean carbon sequestration here, anthropogenic CO<sub>2</sub> is injected into the globally-averaged non-polar domain, which implicitly assumes that carbon is injected uniformly throughout this domain in the real world. In reality, CO<sub>2</sub> would be injected preferentially in specific oceanic regions. For injection depths of 800 m and 1500 m, Caldeira et al. [2001] have shown using a 3-D ocean model that this can lead to greater or slower outgassing compared to that obtained with a 1-D model, depending on the injection site. For injection at 3000 m, the 1-D model retains 5–20% more CO<sub>2</sub> after 500 years than the 3-D model, depending on the injection site, but the differences between the 1-D and 3-D model results are smaller than the differences arising from different injection depths and presumably for widely different injection amounts. Thus, the present results are a useful illustration



**Figure 3.** Impact on non-polar mixed layer supersaturation and pH of scenarios without sequestration (solid lines) and with sequestration (dashed lines).

of the magnitude of the atmospheric CO<sub>2</sub> buildup and of large-scale impacts on oceanic surface layer chemistry for vastly different scenarios of CO<sub>2</sub> sequestration.

[12] It should be noted that the local effects on the marine biota, next to the deep-ocean CO<sub>2</sub> injection sites, are also of concern [Adams *et al.*, 1997; Auerbach *et al.*, 1997; Tamburri *et al.*, 2000; Drange *et al.*, 2001]. Minimization of these effects requires injecting CO<sub>2</sub> from a large number of dispersed points rather than from a small number of large injection sites, a task that is rendered more difficult for the extreme carbon sequestration scenarios. Outgassing of CO<sub>2</sub> and effects on pH of oceanic carbon disposal could be eliminated if captured CO<sub>2</sub> were reacted with seawater in a reactor vessel that contained crushed carbonate minerals, then released to the ocean [Caldeira and Rau, 2000]. However, this requires a substantial infrastructure for supplying carbonate minerals, with its own environmental impacts, as well as to pump seawater from the ocean to the CO<sub>2</sub> recovery site and back.

## 5. Concluding Comments

[13] The results presented here indicate that sequestration of carbon in the deep ocean does not avoid significant climatic and marine impacts, particularly if this is the CO<sub>2</sub> management strategy relied upon in place of a business-as-usual emission scenario. Given doubts about the carbon sequestration potential in geological formations, oceanic carbon sequestration can safely be assumed to play a useful role, with minimal adverse impacts, only as a supplement to strong reductions in fossil fuel use. Carbon sequestration, both in geological formations on land and in the oceans, is analyzed in this context elsewhere (LDDH).

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L. D. D. Harvey, Department of Geography, University of Toronto, 100 St. George Street, Toronto, Ontario, Canada, M5S 3G3. (harvey@geog.utoronto.ca)