Carbon Dioxide Sequestration
A Solution to a Global Problem

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H uman and industrial development over the past hundred years has led to a huge increase in fossil fuel consumption and CO₂ emissions, causing a dramatic increase in atmospheric CO₂ concentration. This increased CO₂ is believed to be responsible for a significant rise in global temperature over the past several decades. Global-scale climate modeling suggests that the temperature increase will continue, at least over the next few hundred years, leading to glacial melting and rising sea levels. Increased atmospheric CO₂ also leads to ocean acidification, which will have drastic consequences for marine ecosystems. In an attempt to solve these problems, many have proposed the large-scale sequestration of CO₂ from our atmosphere. This introductory article presents a summary of some of the evidence linking increasing atmospheric CO₂ concentration to global warming and ocean acidification and our efforts to stem this rise though CO₂ sequestration.

**KEYWORDS:** global carbon cycle, CO₂ sequestration, global warming, ocean acidification

**CO₂ AND GLOBAL CLIMATE CHANGE**

Few subjects have been more polarizing over the past decade than global warming. A large body of evidence demonstrates that global temperatures are rising. Eleven of the past 12 years rank among the 12 warmest since the 1850s, when temperature began to be regularly recorded (IPCC 2005). This temperature increase has linked to more-intense precipitation events, including hurricanes (Groisman et al. 2005), a decrease in ocean thermohaline circulation (where higher-density, cold, saline water drags ocean currents down) (Broecker 1997), and rising sea levels in response to glacial melting (e.g. Manabe and Stouffer 1994; Rignot 1998). Rising sea level can lead to the flooding of vast stretches of the coast. It has been estimated that an increase in sea level of just 4 meters would displace over 300 million people and flood 1.7 × 10⁶ km² of land (equivalent to the combined land area of France, Spain, Germany, and Italy) (Rowley et al. 2007). A temperature increase can also have significant effects on global ecosystems (Cole and Monger 1994), including plant distribution (Mckenney et al. 2007), and has already led to the extinction of numerous species, particularly in polar and mountain-top environments (Parmesan 2006). Other investigations suggest that an increase in global temperature can significantly increase the number and extent of diseases that can infect humans (Patz et al. 1996).

Many attribute this recent global warming to human influence on atmospheric composition (e.g. Crowley 2000; Karl and Trenberth 2003). Global circulation models suggest that much of the observed global temperature increase stems from an increase in atmospheric CO₂ (Manabe and Stouffer 1994; Johns et al. 2003). These models are apparently confirmed by correlations between historic CO₂ concentration and temperature. An example of one such correlation is shown in Figure 1. A strong connection exists between temperature and atmospheric CO₂ content, as shown by data covering the past 400,000 years from Antarctic ice cores. Perhaps most disconcerting is that there may be a positive feedback between increasing atmospheric CO₂ concentration and climate. This positive feedback results from a decreasing ability of the terrestrial biosphere to act as a carbon sink as temperature increases (Cox et al. 2000). As a result of the combined effects of human CO₂ emissions and this positive feedback, global climate models predict average temperature increases of 2 to 5°C by 2100 (Johns et al. 2003). Reducing the impact of CO₂ emissions on the atmosphere and global climate change is thus considered one of the main challenges of this century (e.g. Gunter et al. 1996; Lackner 2003; Pacala and Socolow 2004; Oelkers and Schott 2005; Broecker 2005; Schrag 2007).

The link between increased atmospheric CO₂ content and global warming is not without its controversies. Some evidence suggests that increased atmospheric CO₂ content in the past followed, rather than led, global warming events. In a high-resolution analysis of Antarctic ice-core data, Stott et al. (2007) observed that the beginning of the rise in CO₂ content lagged the increase in global temperature by 700 to 1000 years. The possibility that increased atmospheric CO₂ follows global warming is consistent with the retrograde solubility of CO₂ in seawater. As temperature increases, the solubility of CO₂ in the oceans decreases. Thus, with increasing temperature, CO₂ would evolve from the oceans, increasing the proportion of CO₂ in the atmosphere. There are indications that global warming and polar-ice melting could provoke a new ice age. Global cooling could be triggered...
by the decreasing salinity of the oceans as the glaciers melt; decreased ocean salinity could dramatically decrease thermohaline ocean circulation, leading to slowing of the currents that carry warm Gulf of Mexico water north of Britain and Norway, thus cooling the North Atlantic (Rahmstorf and Ganopolski 1999; McManus et al. 2004).

It has also been argued that celestial factors have a significant effect on climate. Marsh and Svensmark (2003) presented evidence suggesting that variations in solar activity have had an important influence on the Earth’s climate in the past. Veizer (2005) reviewed models advocating cosmic-ray flux as the principal driver of global temperature variation. Hays et al. (1976) proposed that variations in the Earth’s orbit could affect global temperature. Model results of Stott et al. (2003), however, suggest that although solar forcing likely played a large role in controlling global mean temperatures during the first half of the twentieth century, greenhouse gas increases were responsible for most of the warming observed during the last 60 years.

THE/global/carbon/cycle

Carbon dioxide is a trace gas in the Earth’s atmosphere. Its current overall concentration is ~385 parts per million (ppm) by volume or 582 ppm by mass. Pre-industrial levels are estimated at ~280 ppm by volume. The variation in CO₂ concentration measured at the Mauna Loa, Hawai‘i, observatory is shown in Figure 2. This concentration has increased from roughly 325 ppm in 1970 to 380 ppm at the beginning of this century. The smaller seasonal variation stems from biological activity: CO₂ is consumed by biota during the summer and released in the winter. In urban areas, CO₂ is generally higher, and indoors it can reach 10 times the background outdoor concentration. The mass of the Earth’s atmosphere is 5.14 × 10¹⁸ kg (Trenberth et al. 1988), so the total mass of carbon dioxide can be estimated to be ~3.0 × 10¹⁵ kg, which is 3000 gigatons (Gt) of CO₂ or 800 Gt of carbon (1 Gt = 10⁹ metric tons).

The atmosphere, however, is one of the smallest global CO₂ reservoirs. The world’s oceans contain ~39,000 Gt of carbon, while soils, vegetation and detritus contain ~2000 Gt C and carbonate rocks (limestone, marble, chalk) ~65,000,000 Gt C. So, in total, the atmosphere only contains roughly 0.001% of the carbon present in the atmosphere–ocean–upper crust system. Moreover, the mass of carbon in the lower crust and mantle far exceeds that found in these near-surface reservoirs (Holland 1978). There are huge exchanges among these reservoirs (Fig. 3). Each year, the atmosphere exchanges an estimated 90 Gt C with the surface ocean and 110 Gt C with vegetation (Houghton 2007). These numbers imply that the residence time of CO₂ in the atmosphere is no more than ~4 years. CO₂ exchange currently sequesters roughly half the annual anthropogenic global CO₂ emissions into the oceans and soils. This large-scale natural sequestration provides confidence that it may be feasible to remove CO₂ from the atmosphere and sequester it in other reservoirs in quantities sufficient to moderate the effects of anthropogenic CO₂ emission (Lal 2008).

CO₂ and Ocean Acidification

CO₂ exchange between the atmosphere and the ocean is so rapid that the increase in the atmospheric concentration of CO₂ has already had substantial effects on marine chemistry (Caldeira and Wickett 2003). The pH of the ocean’s surface water has already decreased by 0.1 units compared to pre-industrial values (Haugan and Drange 1996). Model calculations indicate that continued anthropogenic CO₂ emissions could lead to a pH decrease in the oceans by as much as 0.3 to 0.4 units by the end of this century (Orr et al. 2005). This pH decrease would destabilize calcite and aragonite, the minerals that form the shells of many marine organisms (Andersson et al. 2006). As a consequence, it has been suggested that calcifying organisms, including corals,
coralline algae, molluscs and foraminifera, would have difficulties building their skeletons. This could have drastic implications for marine ecosystems, including disaster for some planktonic species, which would affect the entire food chain, and vast expanses of coral reefs (Andersson et al. 2006).

THE HUMAN IMPACT ON THE GLOBAL CARBON CYCLE

Many people are not aware of just how much CO$_2$ each of us produces. Many car drivers fill their gas tank at least twice a month. Each fill-up uses about 50 litres of gasoline. The density of gasoline is roughly 730 grams per litre, so the mass of each fill-up is about 36.5 kilograms. Gasoline is a mixture of hydrocarbons, including alkanes (straight chain molecules) such as heptane (C$_7$H$_{16}$) and aromatic (ring) compounds such as benzene (C$_6$H$_6$); gasoline can be represented by the general formula CH$_2$. When burned, it produces energy by the reaction:

$$\text{CH}_2 + \frac{3}{2}\text{O}_2 = \text{CO}_2 + \text{H}_2\text{O}.$$ 

The molecular weight of CH$_2$ is 14 g/mole, and that of CO$_2$ is 44 g/mole, so 3.1 kg CO$_2$ is produced for every kilogram of gasoline burned. It follows that each 50 litre tank of gasoline produces about 113 kg CO$_2$, which, for 24 annual fill-ups, translates to roughly 2.7 metric tons of CO$_2$ per year! If this CO$_2$ were stored mineralogically by creation of calcite (CaCO$_3$), in accord with the reaction:

$$\text{Ca}^{2+} + \text{CO}_2 + \text{H}_2\text{O} = \text{CaCO}_3 + 2\text{H}^+,$$

the mass of waste increases even more; the molecular weight of calcite is 100, so each kilogram of CO$_2$ would create 2.3 kg of calcite. To fix the CO$_2$ produced annually by this car in mineral form, where it would be geologically stable, one would need to produce 6.2 metric tons of calcite, a volume of 2.3 m$^3$.

By this simple example, it is easy to imagine how the world’s 6.6 billion people can contribute so much CO$_2$ annually to the Earth’s atmosphere. Manufacturing, transport of goods, and production of cement all add to the total. Human industrial CO$_2$ emissions, primarily from the use of coal, oil and natural gas, and from the production of cement, currently contribute about 8 Gt C (29 Gt CO$_2$) per year. The evolution of global CO$_2$ emissions over the past 250 years is shown in Figure 4. Total human addition to the atmosphere since 1751 is estimated to be 315 Gt C (Marland et al. 2007). Of the current CO$_2$ emissions, 18% originate from burning (natural gas, 42% from oil, 36% from coal, and 4% from making cement. In addition, the human population produces an estimated 0.6 gigatons of CO$_2$ per year just by exhaling.

Biofuels have been proposed as a solution, because the CO$_2$ produced by burning came originally from the atmosphere and was fixed by plants via photosynthesis. However, the production and burning of biofuels (1) requires substantial energy for farming, transport and processing, thus producing substantial CO$_2$; (2) requires the use of land, water and fertilizer that could otherwise be used to produce food; and (3) generates aldehydes and other compounds that are dangerous to humans, animals and the Earth’s ozone layer.

The global average per capita CO$_2$ emissions over the past 50 years are presented in Figure 5. Emissions roughly doubled over two decades, from 0.65 tons in 1950 to 1.2 tons in 1970, and have remained relatively stable since. Per capita emissions vary greatly from country to country. The top 10 emitters for 2004, on a per capita basis, are listed in Table 1. Oil-producing countries dominate. The United States and Canada are high on the list, with an average of >5 t C per person per year. Most European countries emit between

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**Table 1**

<table>
<thead>
<tr>
<th>Rank</th>
<th>Country</th>
<th>Per capita carbon emissions (ton C/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Qatar</td>
<td>21.63</td>
</tr>
<tr>
<td>2</td>
<td>Kuwait</td>
<td>10.13</td>
</tr>
<tr>
<td>3</td>
<td>United Arab Emirates</td>
<td>9.32</td>
</tr>
<tr>
<td>4</td>
<td>Aruba</td>
<td>8.25</td>
</tr>
<tr>
<td>5</td>
<td>Luxembourg</td>
<td>6.81</td>
</tr>
<tr>
<td>6</td>
<td>Trinidad and Tobago</td>
<td>6.80</td>
</tr>
<tr>
<td>7</td>
<td>Brunei</td>
<td>6.56</td>
</tr>
<tr>
<td>8</td>
<td>Bahrain</td>
<td>6.53</td>
</tr>
<tr>
<td>9</td>
<td>United States</td>
<td>5.61</td>
</tr>
<tr>
<td>10</td>
<td>Canada</td>
<td>5.46</td>
</tr>
</tbody>
</table>
1.5 and 3 t C per person per year while less-industrialized Asian and African countries average <0.03 t C per person per year. The top overall CO$_2$ emitters are listed in Table 2. Industrialized countries currently lead, and those in a phase of active growth, such as China and India, will increase emissions dramatically as their economies grow.

**CO$_2$ SEQUESTRATION EFFORTS**

CO$_2$ storage and mineralization or immobilization, whether natural or engineered, depend on a complex set of chemical processes to assure capture, transport and final deposition (IPCC 2005). Capture is complicated by the diverse nature of CO$_2$ sources (Rubin 2008 this issue). Roughly 60% of emissions originate from large stationary facilities such as power plants, cement production and oil or gas refineries. Such emissions are commonly a mixture of gases (CO$_x$, NO$_x$, SO$_x$, where x represents an integer), often requiring some sort of separation prior to CO$_2$ storage (Rubin 2008). In the ideal case, capture results in a concentrated CO$_2$ stream that can readily be collected and transported. The rest of the CO$_2$ originates from moving sources such as motor vehicles and airplanes. Capturing CO$_2$ emissions from these sources may require direct removal of CO$_2$ from the atmosphere.

Three major types of carbon storage have been proposed: geological storage, ocean storage, and mineral carbonation. In this issue, each of them is explored in detail in separate articles (Benson and Cole 2008; Adams and Caldeira 2008; Oelkers et al. 2008). In addition, there may be some potential for increasing the amount of CO$_2$ that is stored as biomass in forests and soils.

**Geological Storage**

Geological storage relies on the injection of CO$_2$ into porous rock formations (Holloway 2001; Friedmann 2007; Benson and Cole 2008). CO$_2$ storage reservoirs include sedimentary basins, depleted oil reservoirs and non-economic coal beds. An impermeable cap rock is essential because CO$_2$ density is generally less than that of water, so buoyancy tends to drive CO$_2$ upwards, back to the surface. Several industrial-scale geologic CO$_2$ storage programs are already underway, including the Norwegian Sleipner project in the North Sea (Korbøl and Kaddour 1995) and the Weyburn project in Canada (Emberley et al. 2004); at these sites, a million tons or more of CO$_2$ is injected into the subsurface each year. CO$_2$ can also be injected into oil field reservoirs in an attempt to enhance petroleum recovery (known as EOR). Despite the apparently large annual injected volume, these projects, and other similar efforts, currently store less than one-tenthousandth, 0.01%, of the global annual anthropogenic CO$_2$ production.

**Ocean Storage**

Ocean storage means the injection of captured CO$_2$ into the ocean, usually at depths greater than 1000 metres, where it would be isolated from the atmosphere (Adams and Caldeira 2008). CO$_2$ would subsequently dissolve into the ocean and become part of the global carbon cycle. This storage method has yet to be attempted at a pilot scale. Ocean storage capacity may be enhanced by the formation of CO$_2$ hydrates or by the creation of liquid CO$_2$ lakes on the ocean floor.

**Mineral Carbonation**

Mineral carbonation aims to create stable carbonate minerals such as magnesite (MgCO$_3$) and calcite (CaCO$_3$) by reacting CO$_2$ with silicate minerals containing magnesium and calcium (Oelkers et al. 2008). Such minerals are stable over geologic timescales, so sequestration by this method would minimise risk of later leakage back to the atmosphere. Mineral carbonation mimics natural weathering, but an industrial-scale operation may require the mining and grinding of suitable Mg- and Ca-bearing silicate minerals and the disposal of vast quantities of end-product carbonate minerals. On the positive side, the resulting material could be used as a building material, as an additive to concrete or paper, or as a soil amendment to improve texture, pH and fertility of low-productivity soils. This enhances overall carbon sequestration by increasing below- and above-ground biomass and soil organic-matter content.
Biomass and Soil

Partial alternatives to these industrial solutions may be to store CO$_2$ in forests and soils. The biomass of forests is both a sink and a source for atmospheric CO$_2$. Vegetation absorbs carbon through photosynthesis. Although some is emitted again during respiration, there is net CO$_2$ storage. The stored carbon eventually returns to the atmosphere when the biomass decays or is burned. Forests can be managed to increase their stored carbon, thus reducing atmospheric CO$_2$ concentrations. The use of forests for CO$_2$ storage will require (1) that forests be managed to grow continuously and (2) that the carbon harvested from forests not be returned to the atmosphere. Management practices to maintain, restore, and increase carbon storage in forest soil include: the use of fertilizer; increased density of agriculture and decreased slash-and-burn practices; the preservation of wetlands, peatlands, and old-growth forest; and the forestation of degraded and nondegraded sites, marginal agricultural lands, and lands subject to severe erosion (Johnson 1992). One solution for storing carbon fixed by, then harvested from, forests is its addition to soils in the form of biochar, which also has the potential to greatly enhance soil fertility (see box). The potential for forests to sequester CO$_2$, however, is limited. Nilsson and Schopfhauser (1995) estimated that only 345 million hectares are available worldwide for afforestation (planting trees on land that has been without forest cover for more than 50 years); an afforestation program of this scale would fix only 1.5 Gt C/year, which is less than 20% of the current anthropogenic carbon input to the atmosphere. Moreover, even attaining this limited land area for afforestation is challenging due to pressure to use land for other purposes, in particular agriculture and development.

PERSPECTIVES

The public desire to address global warming, thus to sequester large quantities of CO$_2$ to stem the increase in atmospheric CO$_2$ content, poses many challenges and opportunities for the geological community. Our geochemistry, mineralogy and petrology community has unique expertise that is essential for designing successful, long-term strategies for storing CO$_2$. This expertise is essential for selecting suitable carbon-storage sites, designing the injection facilities, developing monitoring techniques, predicting the fate of CO$_2$ once injected into the subsurface, and assessing the reactivity of the host material with the CO$_2$ introduced, under a spectrum of diverse environments. This Elements issue is an attempt to further motivate our community to address these challenges.

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