



Soil-zone adsorption of atmospheric CO₂ as a terrestrial carbon sink

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Abstract

Identifying and quantifying sources and sinks of CO₂ is integral to developing global carbon budgets and effectively modeling climate change. Adsorption of CO₂ onto mineral and soil surfaces has generally been regarded as an insignificant sink, though few studies have investigated adsorption on natural materials at temperatures and CO₂ concentrations relevant to atmospheric or soil zone conditions. In this study, annual adsorption at the scale of North America was modeled for the upper 3 m of the Earth's surface (the root zone) based on our own and published adsorption data, and results compared with reported estimates for the North American terrestrial carbon sink during 2000–2005. Our results suggest that adsorption can account for 1–3% of the average annual sink during these years. At smaller regional scales where more adsorptive deposits are present, such as volcanic ash or high-organic soils, the sink may be significantly larger.

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1. INTRODUCTION

Efforts to model global climate change rely heavily on our understanding of the global carbon budget. The greatest uncertainties in detailed carbon budgets have been associated with identifying and quantifying the varied terrestrial sources and sinks (Houghton, 2007). Numerous potential sinks have been investigated including forest biomass (e.g. Barford et al., 2001; Luyssaert et al., 2008; McMahon et al., 2010), weathering reactions (Brady, 1991; Andrews and Schlesinger, 2001; Zaihua et al., 2011), soil organic material (Gaudinski et al., 2000; Harrison, 2004; Brown et al., 2010), erosion and burial of carbon (Stallard, 1998; Einsele et al., 2001; Downing et al., 2008), soil pore water (Andrews and Schlesinger, 2001), and even bacteria (Brais-

sant et al., 2002). The total annual uptake of CO₂ for any particular continent or region varies with time and environmental conditions (natural and anthropogenic).

Climate models typically do not consider adsorption (removal from the gas phase by attachment to solid surfaces) as a sink, perhaps because of an impression that adsorption has been well studied and found to be insignificant for this context. There have indeed been a large number of investigations on CO₂ adsorption, but very few have been conducted on common natural minerals at temperatures, moisture contents, and CO₂ concentrations relevant to near Earth-surface conditions. Examples of previous work on CO₂ adsorption include simulated conditions on Mars (e.g. Fanale and Cannon, 1979; Zent and Quinn, 1995), surface area measurements at cryogenic temperatures (Dandy, 1969; Almore, 1974), CO₂ capture using natural and modified clays and zeolites (Volzone, 2007; Bonenfant et al., 2008), influence on mineral surface chemistry (Zeltner and Anderson, 1988), and mechanisms of adsorption at specific surface sites (Fripiat et al., 1974; Rochester and Topham, 1979). None of these studies can be reasonably extrapolated or applied to soil-zone environments.

All gases in the subsurface experience some degree of adsorption, with the magnitude subject to the gas concen-

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tration, affinity for solid phases, presence of competing gases or ions, and temperature. For adsorption of CO₂ to serve as a significant carbon sink, two criteria must be met. First, increases in atmospheric concentration of CO₂ must be followed by a concomitant increase in the concentration in the soil zone to drive additional adsorption. Second, the mass of additional CO₂ adsorbed each year must represent a non-trivial fraction of the overall annual terrestrial sink.

The concentration of CO₂ in the soil zone is much higher than in the atmosphere due to root respiration and decomposition of organic material, with large differences between locations caused by variations in climate and ecosystems. It cannot be automatically assumed that increases in atmospheric concentrations will result in increased soil CO₂ concentrations. However, several studies under a variety of climates and vegetative covers have documented that increases in atmospheric CO₂ can produce increases in subsurface CO₂ production (Nakayama et al., 1993; Johnson et al., 1994; Luo et al., 1996; Griffin et al., 1997; Ball and Drake, 1998; Ball et al., 2000; Andrews and Schlesinger, 2001; Jackson et al., 2009). Although each cited study documented increases, the magnitude varied considerably, making it impossible to apply a fixed increase in subsurface concentration or production of CO₂ following a particular increase in atmospheric CO₂.

2. ADSORPTION MEASUREMENTS ON SOILS AND VOLCANIC SUBSTRATES

The affinity of different natural substrates for CO₂ varies widely, making it difficult to quantify adsorption over a continental scale. Estimates for this study are based on results from our own investigation of volcanic substrates (11% of the North American surface is volcanic; Blatt and Jones, 1975) and on results from Ravikovitch et al. (2005) for a suite of soils representing a range of clay and organic contents.

2.1. Soil substrates

Ravikovitch et al. (2005) measured surface area, clay content, and total organic content for 12 soils. Surface area ranged from 3.0 to 30.0 m²/g, clay content ranged from 10.7% to 41.5% by mass, and organic matter ranged from 2.2% to 27.4% by mass (tabulated values in original paper). The experimental parameters of the adsorption study were similar to soil-zone conditions, though not specifically designed to simulate the natural environment. Isotherms were measured on dry soils at a temperature of 0 °C for atmospheric CO₂ concentrations up to 29,000 ppm; graphical adsorption isotherms were reported for six of the soils. We generated Freundlich adsorption equations for each of the six reported isotherms for use in our model (Fig. 1, Table 1). In general, higher adsorption was observed for soils with higher organic content.

2.2. Volcanic substrates

Our own adsorption measurements were performed on a suite of volcanic substrates under dry and wetted conditions

at 20 °C, more closely simulating natural conditions, for CO₂ concentrations up to 10,000 ppm (Fig. 1). Substrates included volcanic ash, basalt, moderately weathered tuff, and highly-weathered zeolitic tuff (the last two are hereafter referred to as *tuff* and *zeolitic tuff*, respectively). Organic content of all volcanic substrates was <0.1% based on weight loss on combustion.

2.2.1. Sources and mineralogy

Unweathered ash was obtained from the August 18, 1992 eruption of Mount Spurr, Alaska. Swanson et al. (1995) characterized the ash as andesitic, with fragments of plagioclase, pyroxene, hornblende, and iron-titanium oxide in a microlite-rich matrix of brown glass.

The basalt sample was collected from an outcrop in the Snake River Plain near Idaho Falls, Idaho. Twenty Snake River basalt samples analyzed by Stout and Nicholls (1977) were rich in plagioclase, with lesser amounts of magnetite, ilmenite, olivine, apatite, and iron-titanium oxide.

The tuff sample was collected from a core drilled in the upper “white unit” of the Apache Leap Tuff near Superior, Arizona. Peterson (1961) described the sampled zone as being rich in plagioclase, with lesser amounts of biotite, hornblende, magnetite, cristobalite, orthoclase, and quartz.

The zeolitic tuff sample was obtained from the USW SD-9 drill hole at Yucca Mountain, Nevada, at a depth located in unit 1 of the Calico Hills Formation (Engstrom and Rautman, 1996). Bish et al. (1982) described the unit as highly zeolitized (clinoptilolite and mordenite) with lesser amounts of quartz, cristobalite, sanidine, plagioclase, and biotite.

2.2.2. Surface area measurement

Surface area was determined by the multi-point Brunauer–Emmett–Teller (BET) method using a Quantachrome Nova 1200 Surface Area Analyzer. Adsorption isotherms were measured on crushed rock for three of the four volcanic substrates (no crushing required for ash). Crushing creates additional surface area with the potential for significantly increasing adsorption. The effect of crushing on surface area was determined by measuring the surface area of centimeter-sized fragments, and again after crushing to <0.1 mm for the zeolitic tuff and the basalt. The moderately weathered tuff was received already crushed. Surface area for the basalt increased from 0.15 to 1.07 m²/g. The initial surface area of the zeolitic tuff was 16 m²/g, and did not measurably increase after crushing, likely due to the high intragranular porosity of zeolites.

Isotherms for the basalt and tuff in Fig. 1 are corrected for the increase in surface area created by crushing. The observed increase in surface area for the basalt was approximately one order of magnitude, so the mass of adsorbed CO₂ in each measured isotherm was divided by 10 before plotting and developing Freundlich models to fit the data. Samples of uncrushed tuff were not available, so the increase in surface area was assumed to be similar to the basalt and also corrected by a factor of 10 to represent *in situ* adsorption.

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