



The hydromagnesite playas of Atlin, British Columbia, Canada: A biogeochemical model for CO₂ sequestration

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ABSTRACT

Anthropogenic greenhouse gas emissions may be offset by sequestering carbon dioxide (CO₂) through the carbonation of magnesium silicate minerals to form magnesium carbonate minerals. The hydromagnesite [Mg₅(CO₃)₄(OH)₂·4H₂O] playas of Atlin, British Columbia, Canada provide a natural model to examine mineral carbonation on a watershed scale. At near surface conditions, CO₂ is biogeochemically sequestered by microorganisms that are involved in weathering of bedrock and precipitation of carbonate minerals. The purpose of this study was to characterize the weathering regime in a groundwater recharge zone and the depositional environments in the playas in the context of a biogeochemical model for CO₂ sequestration with emphasis on microbial processes that accelerate mineral carbonation.

Regions with ultramafic bedrock, such as Atlin, represent the best potential sources of feedstocks for mineral carbonation. Elemental compositions of a soil profile show significant depletion of MgO and enrichment of SiO₂ in comparison to underlying ultramafic parent material. Polished serpentinite cubes were placed in the organic horizon of a coniferous forest soil in a groundwater recharge zone for three years. Upon retrieval, the cube surfaces, as seen using scanning electron microscopy, had been colonized by bacteria that were associated with surface pitting. Degradation of organic matter in the soil produced chelating agents and acids that contributed to the chemical weathering of the serpentinite and would be expected to have a similar effect on the magnesium-rich bedrock at Atlin. Stable carbon isotopes of groundwater from a well, situated near a wetland in the southeastern playa, indicate that ~12% of the dissolved inorganic carbon has a modern origin from soil CO₂. The mineralogy and isotope geochemistry of the hydromagnesite playas suggest that there are three distinct depositional environments: (1) the wetland, characterized by biologically-aided precipitation of carbonate minerals from waters concentrated by evaporation, (2) isolated wetland sections that lead to the formation of consolidated aragonite sediments, and (3) the emerged grassland environment where evaporation produces mounds of hydromagnesite. Examination of sediments within the southeastern playa-wetland suggests that cyanobacteria, sulphate reducing bacteria, and diatoms aid in producing favourable geochemical conditions for precipitation of carbonate minerals.

The Atlin site, as a biogeochemical model, has implications for creating carbon sinks that utilize passive microbial, geochemical and physical processes that aid in mineral carbonation of magnesium silicates. These processes could be exploited for the purposes of CO₂ sequestration by creating conditions similar to those of the Atlin site in environments, artificial or natural, where the precipitation of magnesium carbonates would be suitable. Given the vast quantities of Mg-rich bedrock that exist throughout the world, this study has significant implications for reducing atmospheric CO₂ concentrations and combating global climate change.

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1. Introduction

Atmospheric concentrations of greenhouse gases (e.g., carbon dioxide, methane, and nitrous oxide) have increased significantly as a

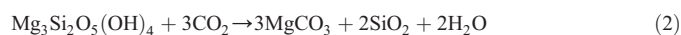
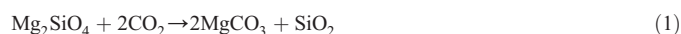
result of human activities since the pre-industrial era (AD 1000–1750). Most notably, carbon dioxide (CO₂) has increased from a pre-industrial level of approximately 275–285 ppm to 379 ppm in 2005, which has been caused mainly by fossil fuel consumption and to a lesser extent land use changes (Solomon et al., 2007). The increase in globally averaged temperatures since the mid-20th century can very likely be attributed to the increase in anthropogenic greenhouse gases (Metz et al., 2005). One means of reducing net greenhouse gas emissions is through the capture and storage of CO₂. Several methods

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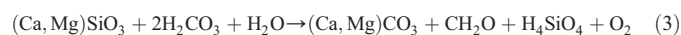
have been proposed including mineral carbonation, oceanic storage, underground injection for enhanced fossil fuel recovery, and injection into saline aquifers (Giammar et al., 2005; Metz et al., 2005; Xu et al., 2005).

Mineral carbonation has traditionally been defined as a chemical process that involves the reaction of CO₂ with silicate minerals to form carbonate minerals (Lackner et al., 1995; Gerdemann et al., 2007). However, this reaction is a natural process of weathering that has occurred over geological time and currently sequesters an estimated 100 million tons of carbon each year (Siefritz, 1990). The abundance of silicate minerals and the formation of a stable form of carbon, as carbonate minerals, are advantageous in that carbon storage is easily verifiable and stable on a geologic timescale. Ultramafic rocks typically contain high proportions of magnesium oxide (up to 50% by weight) in comparison to the CaO in calcium silicate rocks. For this reason, ophiolite belts that contain high abundances of magnesia as serpentine and olivine represent the best potential feedstock for mineral carbonation (Metz et al., 2005). The chemical reactions for the direct carbonation of serpentine and olivine are given in Eqs. (1) and (2) (Lackner et al., 1995; Metz et al., 2005).



In the Atlin, British Columbia region, Hansen et al. (2005) documented the controls on the formation of listwanite, a carbonate altered serpentinite, in the ultramafic rocks of Monarch Mountain. The hydrothermal transformation of these rocks is similar to industrial processes that are being considered for CO₂ sequestration using mineral carbonation. Lackner et al. (1995) used chemical treatment with high temperature and pressure to accelerate ex situ mineral carbonation, which they proposed could be implemented on an industrial scale. However, the current state of this technology requires a large input of energy and is more expensive than most other applications of geological storage (Metz et al., 2005). For these reasons, CO₂ sequestration by this means may not be feasible on a large scale. Alternatively, Kelemen and Matter (2008) have proposed in situ carbonation of peridotite and have shown that 10⁴ to 10⁵ tons per year of CO₂ is sequestered by natural carbonation of the Samail ophiolite, Sultanate of Oman.

In contrast to chemical treatment, biological processes involved in natural mineral carbonation have been largely unexplored. Ferris et al. (1994) suggested that weathering of silicate minerals in bedrock is biogeochemically coupled to the precipitation of carbonate minerals by microorganisms. The soil biota enhance weathering rates by increasing the partial pressure of CO₂, generating chelating agents, and both inorganic and organic acids (Schwartzman and Volk, 1989; Oh and Richter, 2004). Microbial processes that mediate or promote carbonate precipitation include photosynthesis, ammonification, denitrification, sulphate reduction, anaerobic sulphide oxidation, methanogenesis, and manganese and iron oxide reduction (Ferris et al., 1994; Riding, 2000; Roberts et al., 2004). With particular relevance to this study, biologically-aided precipitation of dypingite [Mg₅(CO₃)₄(OH)₂·5H₂O] was experimentally demonstrated by Power et al. (2007) using cyanobacteria cultured from the Atlin site. The combined processes of bedrock weathering and carbonate precipitation by oxygenic phototrophic microorganisms are described by Eq. (3) (Ferris et al., 1994).



The hydromagnesite playas near Atlin provide a natural model for developing a passive process for CO₂ sequestration that involves (1)

the weathering of ultramafic rock and (2) precipitation of magnesium carbonate minerals via a pathway that is mediated by microorganisms. This model differs significantly from the many industrial methods that have been proposed for CO₂ sequestration. Other environments where this process may be occurring naturally include the playas at Salda Gölü in southwestern Turkey (Braithwaite and Zedef, 1994; Braithwaite and Zedef, 1996; Russell et al., 1999) and the hydromagnesite–magnesite playas of the Cariboo Plateau in British Columbia (Renaut, 1990, 1993).

The aim of this study is to characterize the hydromagnesite playas near Atlin and their surrounding environment in the context of a natural model for CO₂ sequestration with emphasis on microbial processes that accelerate mineral carbonation. This is accomplished by examining soil–microbial–bedrock interactions in the weathering regime, tracking the movement of carbon, and determining the genesis of the playas.

2. Background and general description of study area

Atlin is located in northwestern British Columbia, Canada (59°35' N, 133°42' W) at an elevation of 674 m. The area is situated in the Teslin Plateau ecosection, which is characterized by rolling hills and high plateaus with several large lakes (e.g., Atlin Lake) in wide valleys. This ecosection lies in a distinct rain shadow and has a dry sub-arctic climate. The yearly average temperature in Atlin is 0.5 °C with January (−15.4 °C) and July (13.1 °C) having the coldest and warmest average temperatures of the year, respectively. Precipitation (348 mm/yr) includes an average of 193 mm of rainfall and 155 mm of snowfall per year (Environment Canada). The Teslin Plateau ecosection is further subdivided into three biogeoclimatic zones. These include an alpine tundra zone (Boreal Altae Fescue Alpine) that is present on middle to upper slopes of most mountains, a spruce–willow–birch zone in high valleys and middle slopes, and a boreal white and black spruce zone that occurs throughout valley bottoms (British Columbia Ministry of Forests and Range). In this study, the role of bioweathering of ultramafic rock is examined on the slopes of Monarch Mountain near the town of Atlin.

The bedrock in this area is generally ultramafic and represents a tectonically emplaced upper mantle section of oceanic lithosphere consisting of an ophiolitic assemblage that formed during the Early to Middle Jurassic. The rocks are mainly serpentinitized harzburgite with variable serpentinitized, carbonitized, and deformed harzburgite, and with minor dunite lenses and pyroxenite veins (Hansen et al., 2005). The weathering of ultramafic bedrock in the Atlin area is reflected in the geochemistry of various waters that have been sampled and analyzed in this study.

In topographic lows near the town of Atlin, glacio–fluvial materials underlie two groups of playas that have a total area of approximately 11 ha (Young, 1916). In this study, these two groups are referred to as the northern and southern playas. The southern group consists of two playas, referred to as the southwestern and southeastern playas. Fig. 1 shows the locations and general shapes of each playa; aerial photographs of the northern and southeastern playas are shown in Fig. 2. This study is mostly concerned with the southeastern playa where present-day groundwater discharge forms a small wetland. As seen in Fig. 3, this playa area is subdivided into four terrains: wetland, grassland, localized (metre-scale) and amalgamated (decametre to hectometre-scale) hydromagnesite mounds.

3. Methodology

3.1. Monarch Mountain soil analyses

To examine the onset of (bio)weathering of magnesium silicates, a sample of serpentinite, collected from an outcrop in the southeastern playa, was cut into cubes (~1 cm³) and polished on one

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