

Opportunities for Geologic Carbon Sequestration in Washington State

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Preface and Acknowledgment

This report is the product of reading, conversation, writing and revision by a group of undergraduate student authors during 10 weeks of March, April and May 2020. Our intent is to review the basic processes and state of current scientific understanding of geologic carbon sequestration relevant to Washington State.

We would like to thank Dr. Thomas L. Doe (Golder Associates) for reading the final report and asking us challenging questions.

I. Introduction

Anthropogenic emissions of greenhouse gasses like CO₂ are raising global temperatures at an unprecedented rate. According to the most recent United Nations Intergovernmental Panel on Climate Change report, global emissions have caused ~1°C global warming above pre-industrial levels, with impacts such as rising sea level and changing weather patterns (IPCC 2018). The IPCC states that the current rate of emissions will lead to global warming of 1.5°C sometime between 2030 to 2052, and that temperature will continue to rise above that if emissions are not halted. They state that impacts of global warming, such as drought or extreme precipitation, will increase with a 1.5°C temperature increase. However, these effects will be less than the impacts of global temperature increase of beyond 2°C. The IPCC also projects that global sea level rise will be 0.1 m lower at 1.5°C warming than at 2°C warming, exposing 10 million fewer people to risks related to sea level rise. Because of these factors, it is critical that global warming be limited to a maximum of 1.5°C.

The IPCC (2018) suggests multiple pathways to reduce emissions. The pathways include a myriad of strategies, such as land use change and an almost exclusive reliance on renewable energy. All suggested pathways that limit global warming to 1.5°C include some type of carbon dioxide removal (CDR) or carbon capture and storage (CCS) strategy to offset residual emissions and reach net-negative emissions. Many CDR strategies involve land use change to capture CO₂ in plants and store the carbon as biomass. Many CCS strategies, including geologic carbon sequestration (GCS), capture CO₂ at the source and sequester it in a reservoir. While CDR strategies remove CO₂ from the atmosphere, resulting in net-negative emissions, CCS strategies prevent CO₂ from entering the atmosphere.

Washington State has set ambitious carbon reduction goals. In 2007, the state set a goal of reducing emission levels (around 100 million metric tons (Mt)) to 1990 levels (88.4 Mt) by 2020 (Washington State Greenhouse Gas Emissions Inventory, 2018), a target that was 90% achieved (Washington State Department of Ecology, 2018). In March 2020, Gov. Jay Inslee signed HB 2311, pledging that Washington will reach net-zero greenhouse gas emissions by 2050. In addition, the bill promotes the removal of excess carbon from the atmosphere through

the prioritization of voluntary and incentive-based sequestration activities in order to reach this goal. The State predicts that at least 5% of the carbon emissions to be removed by 2050 will require carbon sequestration.

In this report, we focus on methods of carbon dioxide sequestration that follow geologic processes. These methods provide us with long-term or permanent removal of CO₂ from the atmosphere. Geological carbon sequestration efforts encompass several different methods (National Academies of Sciences, Engineering, and Medicine., 2019):

- *Sequestration in saline aquifers* requires injection of CO₂ as supercritical fluid into sedimentary basins. CO₂ is trapped stratigraphically, by capillary action, dissolution into groundwater, and incorporation of CO₂ into minerals. In Washington, there are several deep sedimentary basins proximal to major emission sources. We address sequestration in sedimentary basins in section II.
- *In situ carbon remineralization* provides a permanent trap of CO₂ in the form of carbonate minerals, by injecting CO₂ into the subsurface, where it reacts with the host rock to produce new minerals. In Washington, widespread voluminous basalts of the Columbia River Basalt Group, basalts of the Juan de Fuca plate off Washington's coast, and sizable ultramafic formations, such as the Twin Sisters dunite, offer opportunity to sequester large volumes of CO₂. We address in situ remineralization in basalt and ultramafic rocks in sections III and IV, respectively.
- *Ex situ carbon mineralization* occurs when solid or liquid materials, such as mine tailings, are reacted with CO₂-rich fluid or gas to precipitate a solid carbonate mineral. These wastes could potentially be used to produce industrial building supplies. Because appropriate mine-tailings are not abundant in Washington, we do not address this possibility further.
- *Surficial enhanced weathering* occurs when atmospheric CO₂ reacts with minerals at Earth's surface, converting CO₂ into dissolved CO₃²⁻ as a weathering product, which is ultimately sequestered by precipitation of carbonates in the ocean. The weathering process is accelerated by spreading crushed mafic and ultramafic rock over agricultural

lands. In section V, we look at the potential for carbon dioxide removal in Washington by surficial enhanced weathering of regional mafic and ultramafic rocks by distribution over cropland.

In addition to these methods of geologic carbon sequestration, we discuss the opportunities for enhancing the aforementioned methods by incorporating seawater and other advancements in section VI.

Considering the advantages of geologic carbon sequestration, the geologic resources available for use in Washington State, and the state government's commitment to becoming carbon-neutral, a thorough understanding of the GCS options available to the state is needed. Our objective in this report is to review several possibilities for implementing geologic carbon sequestration schemes in Washington, weighing the state of the science, the viability of the method, the applicability to Washington State, and potential costs. We then make recommendations for moving forward.

II. Sequestration in Saline Aquifers

Leo F. MacLeod

Introduction

Deep saline aquifer carbon sequestration is a method of carbon storage that involves the injection of supercritical CO₂ into a basin of permeable rock, such as sandstone, that is saturated with brine to the point of being non-potable. The technology is well developed, with many projects around the globe finding it successful and stable. In the United States, there are several facilities implementing this strategy.

Washington has several major sandstone deposits that have the potential for carbon sequestration, with three already evaluated by the USGS. However, there are important drawbacks to consider. In this section, I review how saline aquifer sequestration works, summarize a successful demonstration from Japan, and discuss the opportunities, concerns, and potential benefits of saline aquifer carbon sequestration.

Process of Saline Aquifer Sequestration

The pressure, temperature, and purity of the CO₂ injected is crucial to the process of sequestration, because the greater the density of the carbon, the greater the amount that can be sequestered and the more secure the storage (Bachu 2000; 2002; 2003). As a result, the CO₂ must be in a supercritical state (sCO₂), where it will spread like a gas but with the density of a liquid. This means that the first step to carbon sequestration is the separation and capture of CO₂ from other exhaust gasses at an anthropogenic source like a power plant and then compressing it to supercritical conditions for injection (Maroto-Valer, 2010). Once the CO₂ is separated and compressed, it is transported through a pipeline to the injection well, where it is pumped below the aquifer caprock (e.g. Bachu, 2015).

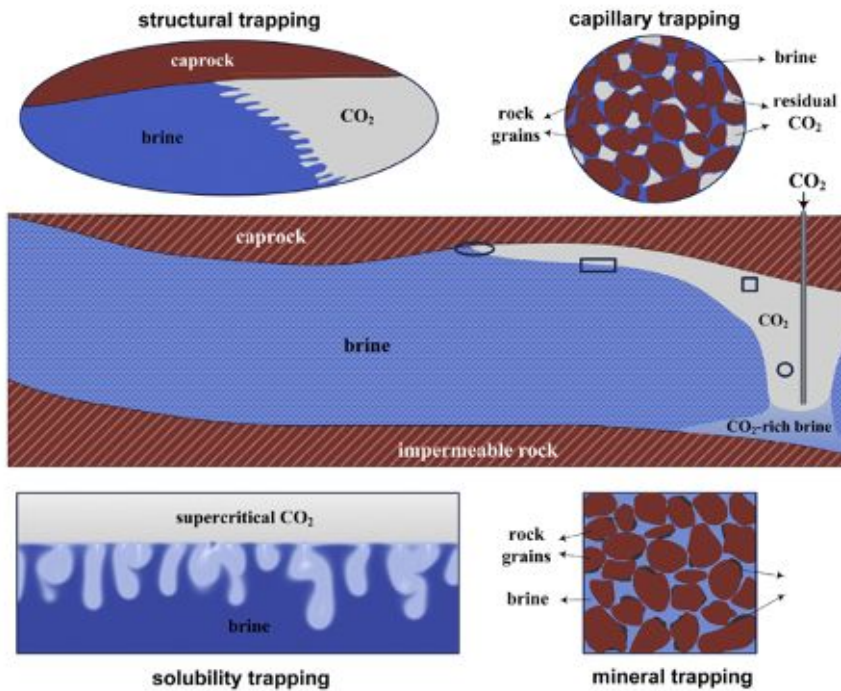
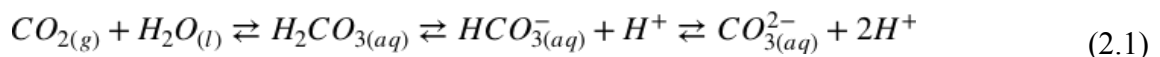


Figure 2.1. Illustration of the four key processes involved in saline aquifer carbon sequestration. Reproduced from Emami-Meybodi et al., 2015.

Carbon is trapped in saline aquifers by four different mechanisms, each increasingly more secure (Fig. 2.1): structural trapping, capillary trapping, solubility trapping, and mineral trapping. These can be envisioned as four stages of sequestration (Fig. 2.2). At the time of injection, the pure sCO₂ is less dense than the saline water. As a result, the sCO₂ will rise through the aquifer until it reaches an impermeable caprock, where it will be stopped and begin to spread laterally throughout the aquifer (Lindeberg and Wessel-berg, 1997). This process of physically confining the carbon beneath a low permeability caprock is called structural and stratigraphic trapping and is initially how a majority of the CO₂ is stored (IPCC, 2005).

The second way the carbon is stored is through residual trapping of smaller volumes of CO₂ within the porous rock space, called capillary trapping (Trevisan et al., 2016). As the CO₂ plume moves away from the injection site, the trailing carbon becomes immobilized within the rock and spreads over a larger area (Macminn et al., 2010; 2011).

The third process is solubility trapping. This is the dissolution of CO₂ in the aquifer water described in Equation 2.1 (IPCC, 2005):



Here, carbon dioxide gas combines with water to produce carbonic acid that steadily breaks down to form free hydrogen ions. This process results in a solution that is denser than the surrounding water and $s\text{CO}_2$, causing it to sink to the bottom of the aquifer. This process also results in convective mixing, causing more of the $s\text{CO}_2$ to become exposed to the saline water over time and create a sustainable circulation of carbon-saturated water to the bottom of the aquifer (MacMinn et al., 2011). A beneficial by-product of the reaction, described in Eq. 2.1, is the lowering of the water pH, which then reacts with the surrounding rocks to enhance host rock dissolution for mineral trapping (Jun et al., 2013).

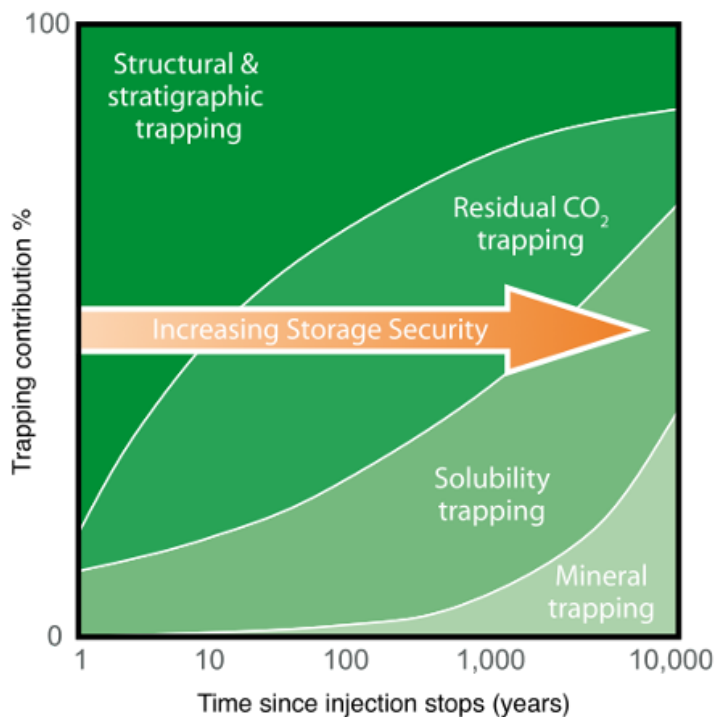
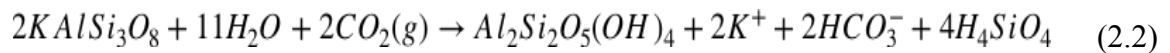


Figure 2.2. Graph showing the change in the type of storage in saline aquifers over time. Reproduced from IPCC, 2005.

Mineral trapping is the fourth form of storage in a saline aquifer. This is the process by which the CO_2 is converted into carbonate minerals through reactions with the aquifer host rocks (Rosenbauer and Thomas, 2010). The rate at which mineral trapping occurs is highly dependent on the rock composition. Carbonates will react more readily with the acidic CO_2 saturated water, while feldspars and clays will have a longer reaction time (Jun et al., 2013). The volume of rock produced is expected to increase over time (Fig. 2.2). As the host rocks are dissolved and new

carbon-containing minerals are precipitated, the water comes into contact with more of the parent rock.

The sandstones that host saline aquifers are most commonly composed of the minerals quartz and feldspar. One example of mineral change is the conversion of potassium feldspar to kaolinite, shown in Equation 2.2 (Harvey et al., 2013):



Here, potassium feldspar is reacted with the acidic water, resulting in the corrosion of the feldspar host rock. This reaction results in the trapping of the carbon in the precipitate (Harvey et al., 2013; Zhao et al., 2014). The 2005 IPCC reported mineralization as a slow process, however, there is increasing research that these reactions happen faster than previously expected (Metz et al., 2005 and Takaya et al., 2012). There is also evidence to suggest that the rate of mineralization in a feldspar dominated aquifer can be increased through the introduction of certain microbes to the system (Zhao et al., 2014). Both of these factors contribute to the evolving safety and long-term effectiveness of saline aquifer storage.

Case Study: Tomakomai Carbon Capture and Storage Demonstration Project, Japan

There are many projects being undertaken around the world to sequester carbon in saline aquifers (summarized in Table 2.1). All of these projects successfully injected or are currently injecting CO₂ into a saline aquifer. The technology involved in saline aquifer sequestration is well developed, having started in 1996 at the Sleipner gas field in the North Sea (Gale and Freund, 2001), and applied in many subsequent trials around the world (Rosenbauer and Thomas, 2010). Thus, saline aquifer storage is considered a practical process for carbon sequestration (Hitchon et al., 1999).

The Tomakomai (Japan) CCS Demonstration Project is an example of a recent and highly successful project that has offered insight into many of the residual questions about long term carbon storage. Two sites were identified 3-4 km off the coast of Tomakomai for the project, at depths of 2400-3000 m and 1100-1200 m below the seafloor, respectively (Tanaka et al., 2014). The project started injection in April 2016, after several years of preparation (Japan CCS Co.,

Ltd., 2020). This pre-injection period involved a three-dimensional seismic survey, exploration well drilling, and tests using the collected core samples to evaluate the potential of the site and to ensure that it was safe for injection (Shoji, 2016).

By the time the project suspended injection in November 2019, the project had a cumulative CO₂ injection of over 300,000 tons (Japan CCS Co., Ltd., 2020). The project is now moving on to long-term monitoring of the site. This monitoring is to collect data on the pressure and temperature of the aquifer, to observe any changes in caprock safety, and to create a reliable before-during-after time series of earthquakes. Even though the project has yet to conclude, the findings have been very promising. This has important implications for carbon sequestration in Washington. One of the Tomakomai wells injected sCO₂ into the Moenetsu Formation, a layer of Lower Quaternary sandstone (Tanase et al., 2013). This is similar to the formation near Aberdeen, WA, which appears to have many feet of sandstone ranging from the Tertiary to Quaternary (Golder Associates, 2007). In addition, the locations under consideration in Japan and in Washington both lie along an oceanic subduction zone, with similar tectonic activity.

Another important result of the Tomakomai Project is the public response. Locally, this form of carbon sequestration has received a neutral to cautiously supportive response. While there are some concerns, in general there is a common understanding that this is a necessary action to slow down climate change, a goal that the city and surrounding areas believe very strongly in. This overall neutral view appears to be a result of the communication between the local government, lead scientists, and the general public (Mabon et al., 2017). This has important implications for managing public response to the integration of CCS activities with Washington state clean energy initiatives.

Implementation in Washington State

There are several key requirements for saline aquifer usability. The basin should be at least 800 m deep to achieve the right pressure and temperature to keep the carbon in a supercritical state (Baccu, 2003). In addition, the basin must also be shallower than 3,000 meters or else the porosity of the formation will be reduced due to compaction (Shoji, 2016). Another condition is that the site is easy to access and is on legally accessible land. This is part of

reducing the cost and ensuring an ethical storage of the sCO₂. It is also highly recommended that the basin be located in a region with low seismic activity (Mercedes et al., 2010).

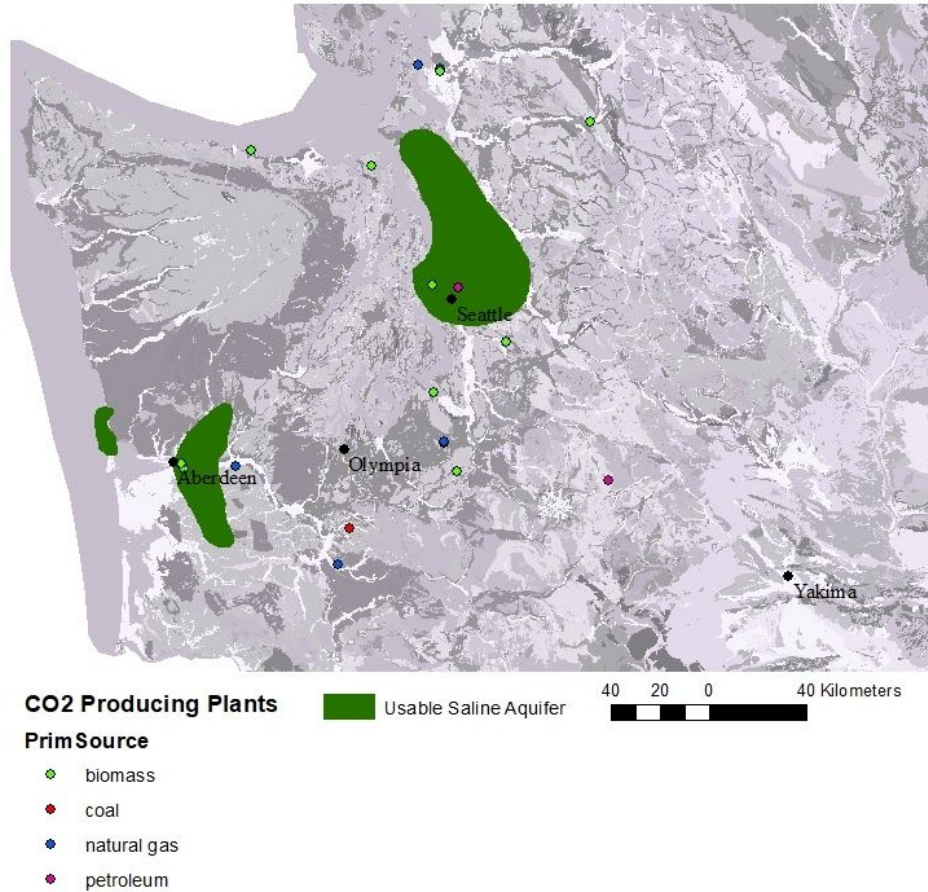


Figure 2.3. Distribution of saline aquifers evaluated for carbon sequestration (Covault et al. 2013) overlain on the geologic map of Washington. CO₂-producing power plants are shown with colored dots. Geologic data from WADNR (2020).

A 2013 review by the USGS gave a preliminary estimate of “technically accessible” CO₂ storage in sedimentary basins of western Oregon and Washington, with sequestration potential estimated to be ~14,000 Mt CO₂. In western Washington there are three key locations that have been evaluated. Two sites fall in the Greys Harbor Basin, one near Aberdeen and one off the coast, and one site falls in the Puget Lowlands, from around Seattle to Port Townsend (Covault et al., 2013) (Fig. 2.3). Each of these locations have over 4,000 meters of Tertiary arkosic sandstone,

with a caprock of coal containing mudstone (Covault et al., 2013). Mudstone has shown to have a low permeability that can support the trapping of carbon (Armitage et al., 2016). The Aberdeen site and the Seattle site both lie under power plants and important pipeline infrastructure. This is crucial when considering the economic viability of the sites. Transportation from source to sink is the most costly part of sequestration, and pipelines are the only cost effective form of onshore carbon transportation (Eccles et al., 2012; Svensson et al., 2004). This means Washington is well situated to utilize these resources.

In eastern Washington there are several large sandstone formations that could be potentially useful sites. Both fall along the boundary with Canada, one in Okanogan County and one in Pend Oreille County. However, these regions have not been evaluated, and are far from power plants. More information is required to assess the viability of the sites.

Conclusion

Saline aquifer carbon sequestration is one of the most mature forms of carbon storage and could be implemented right away. The technology is already developed and being used, though mostly by the energy and petrochemical industries. In addition, this form of storage does not compete with agriculture, fishing, and other important commercial and economic land uses and can be done safely within national borders (Bachu, 2003). Saline aquifer sequestration has the potential to be applied in Washington state. However, before this can be done, we need to consider the stability of the formations under consideration, the incentives that can be provided to carbon producing industries, and the approach for educating the public and addressing concerns.

III. In Situ Remineralization in Mafic Rocks

Jonathan D. Rowe

Introduction

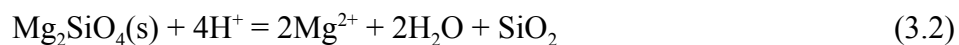
While pumping CO₂ into deep sedimentary basins largely relies on the host rock's physical trapping capabilities (at least initially), an alternative method of sequestering carbon is the in situ remineralization of basaltic rocks, which takes advantage of the geochemical trapping mechanism (Matter et al., 2007). The potential for chemical reactions between CO₂ and the host basalt is the main advantage this carbon storage method offers, since once the carbon is mineralized into stable, not-toxic carbonates, the risk of carbon leakage into aquifers or the atmosphere is very low (Goldberg et al., 2010; McGrail et al., 2017a). Numerous laboratory experiments have investigated this method of carbon sequestration, and two major field experiments have been conducted: the Wallula Basalt Pilot Project in eastern Washington State and the CarbFix project in Iceland. This section discusses the results of these studies and their potential application to Washington State, home to much of the Columbia River Basalt Group (CRBG).

The Process

During in situ basalt remineralization, either supercritical (liquid) CO₂ (sCO₂) or CO₂-charged water is injected into the target basalt. The dominant silicate minerals in the basaltic host rock dissolve in the presence of the CO₂ via two complementary processes (Snæbjörnsdóttir et al., 2020). First, the injected CO₂ reacts with divalent metal cations and water already present in the groundwater of the reservoir, as shown in equation 3.1 (Matter et al., 2007):



In this reaction, the water and divalent metal cations combine with the injected CO₂ to produce carbonate minerals and release hydrogen ions, lowering the pH of the solution. The second process involves silicate minerals dissolving and releasing more cations, as shown in equations 3.2 and 3.3 (Matter et al., 2007).



These reactions illustrate the dissolution of the magnesium and calcium silicates prominent in basalts, such as olivine and Ca-plagioclase. The dissolution produces silica, aluminum silicates like kaolinite, and metal cations, which drive the precipitation of carbonates, as in equation 3.1. The dissolution represented by equations 3.2 and 3.3 is the limiting factor for the speed of this geochemical process (Matter et al., 2007). Matter et al. (2007) conducted a single-well push-pull test in mafic rocks in New York State, injecting CO₂-charged water 230 meters deep into a coarse-grained basalt. While dissolution rates are fastest when CO₂ is below 800 meters, where it can remain in a supercritical state, they concluded that even at shallower depths conditions were sufficient for permanent storage of CO₂ as carbonate minerals (Matter et al., 2007).

The Columbia River Basalt Group (CRBG) is a young continental flood basalt that mostly erupted 17-14.5 Ma in numerous successive flows and covers much of central and southeast Washington. The CRBG is composed of tholeiitic (iron-rich) basalt, and the predominant minerals are plagioclase feldspar, augite (pyroxene), and a non-crystalline glassy groundmass (Schaefer and McGrail, 2009). Schaefer and McGrail (2009) investigated the dissolution behavior of the CRBG rocks over 90-132 days, under a variety of temperature and pH conditions. They found cation concentrations remained steady after 60 days and concluded the CRBG offers a good opportunity for in situ carbon sequestration (Schaefer and McGrail, 2009).

Wells et al. (2017) subjected two polished samples of CRBG rock to CO₂-charged water under conditions of 150°C and 100 bars. Surface roughness parameters for the polished samples increased significantly over 1-2 months, and the surface area of the samples increased by around 20%. These observations are promising for the dissolution potential of the rock, which depends on the access of acidic water to fresh mineral surfaces (Wells et al., 2017). Xiong et al. (2018) subjected whole-core samples of the CRBG to CO₂-charged water at temperatures of 100°C and pressures of 100 bar for 40 weeks. Carbonate minerals were detected within 20 weeks, and they calculated a carbonate mineral trapping rate of 1.24 kg of CO₂ / m³ basalt per year. They calculated a porosity of 3.65% and a potential for ~47 kg of CO₂ to be stored in one cubic meter

of basalt. By their calculations, 1000 tons of CO₂ would take ~40 years to mineralize completely in basalt, compared to an estimated 100,000 years in a sandstone reservoir (Xiong et al., 2018).

One concern regarding this method is the potential decrease in permeability of the basalt due to the precipitation of carbonate minerals (Xiong et al., 2018). Open pathways allow the injected fluid to disperse and interact with the primary minerals. If the pathways become clogged by precipitating carbonates, fluid migration may be impeded (Snæbjörnsdóttir et al., 2020). The CarbFix project, discussed below, addresses this concern directly.

Case study: Wallula Basalt Pilot Project, Washington

The Wallula Basalt Pilot Project, which was part of the Department of Energy's Regional Carbon Sequestration Partnerships Initiative, is a well-characterized case study that demonstrates the potential of this carbon storage method. CRBG rocks consist of two main lithofacies: porous and brecciated flow tops and massive flow interiors (Zakharova et al., 2012). The vesicular and brecciated flow boundaries were created from the flow's bottom boundary cooling quickly due to contact with the surface, and the top boundary cooling quickly due to contact with the atmosphere (Goldberg et al., 2010). The quality of a reservoir is determined by the reservoir's storage capacity and hydraulic conductivity, which are determined by the porosity and permeability, respectively (Zakharova et al., 2012). Flow tops, therefore, are the potential reservoirs, with the flow interiors serving as caprock.

In the summer of 2013, 1000 tons of sCO₂ were injected into the CRBG near Wallula, Washington, at a depth of 828-887 m. Forty tons of sCO₂ were injected per day over a three-week period to avoid disrupting the hydrostatic conditions of the reservoir. Dissolved

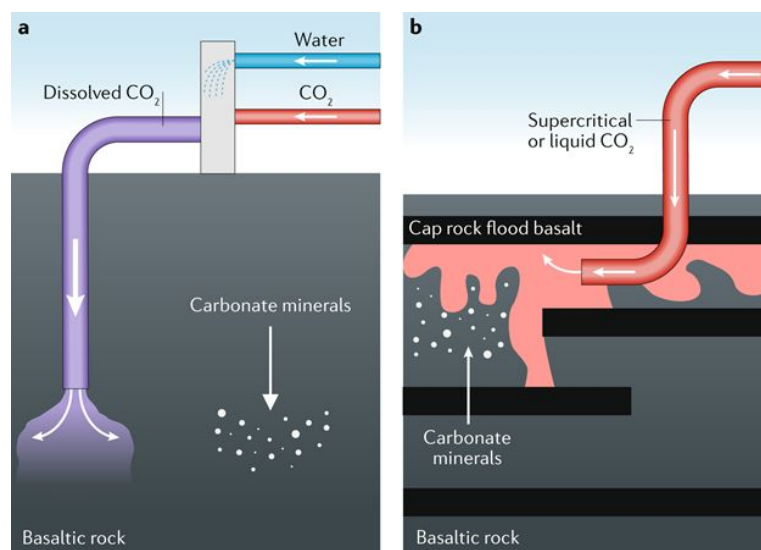


Figure 3.1: Comparison of injection methods. The CarbFix project is using method (A), injecting CO₂ dissolved in water. The Wallula project used method (B), injecting liquid CO₂. Source: Snæbjörnsdóttir (2020).

solids increased 40-fold compared to pre-injection measurements, and the concentration of cations crucial to carbonate formation increased by several orders of magnitude. Extracted sidewall cores revealed globular deposits of ankerite (iron carbonate) in the basalt's vugs and voids, with isotopic analyses confirming that the carbonate was precipitated from the injected CO₂ (McGrail et al., 2017a). The result of the Wallula project was a successful demonstration of the feasibility of sequestering carbon in the CRBG.

Case study: CarbFIX Project, Iceland

The other major demonstration of in situ remineralization of basalt is the CarbFix project in Iceland. CarbFix is an ongoing carbon sequestration project that sequesters approximately 12,000 tons of CO₂ per year and a comparable mass of H₂S (Kelemen et al., 2019). In contrast to the Wallula project, the CarbFix project dissolves the injected CO₂ in water during injection (see Fig. 3.1). Another significant difference is the temperature of the host rock formation: 36°C in Wallula and greater than 250°C at the CarbFix site (Clark et al., 2020). In Phase I of the CarbFix project, techniques were developed to inject the water and CO₂ separately while still enabling the CO₂ to dissolve completely in the water at depth, eliminating the need for a caprock and expanding the portfolio of potential reservoirs (Kelemen et al., 2019). Phase II of the project doubled the amount of gas dissolved in the injected water and saw significant increases in carbon and sulfur mineralization percentages due to increased acidity. Clark et al. (2020) found that the permeability of the host rock has not been significantly affected over 3.5 years of injections and that the mineralization rate is orders of magnitude faster than the estimated mineralization rate for deep saline aquifers. The carbon storage capacity for an 80 km² reservoir in Iceland is estimated at 2400 MtCO₂, assuming a 100-meter thick target formation (Clark et al., 2020). The long-term success of the CarbFix project demonstrates the potential for sequestering CO₂ in basalts, and the CarbFix project remains a good analogue for how a second Wallula project might look.

Relevant Geologic Resources in Washington State

As indicated by the Wallula project and the extensive laboratory studies done on CRBG samples, Washington state is home to a unique geologic resource when it comes to carbon

sequestration (see Fig. 3.2). A significant portion of the 220,000 km³ Columbia River Basalt Group lies within Washington's borders (Schaefer and McGrail, 2009). McGrail et al. (2006) estimated the CRBG to be capable of storing 100 Gt CO₂, assuming ten interflow target reservoirs with an average thickness of 10 meters each. With the United States emitting 5-6 Gt CO₂ annually and Washington state emitting ~100 Mt CO₂ annually (Washington Department of Ecology, 2020), the CRBG could store enough CO₂ to offset regional emissions for the foreseeable future. Regarding reservoir stability, the CRBG is in an area with low observed seismicity, with the minor seismic events that do occur having no observed effect on the aquifers of the region (McGrail et al., 2006).

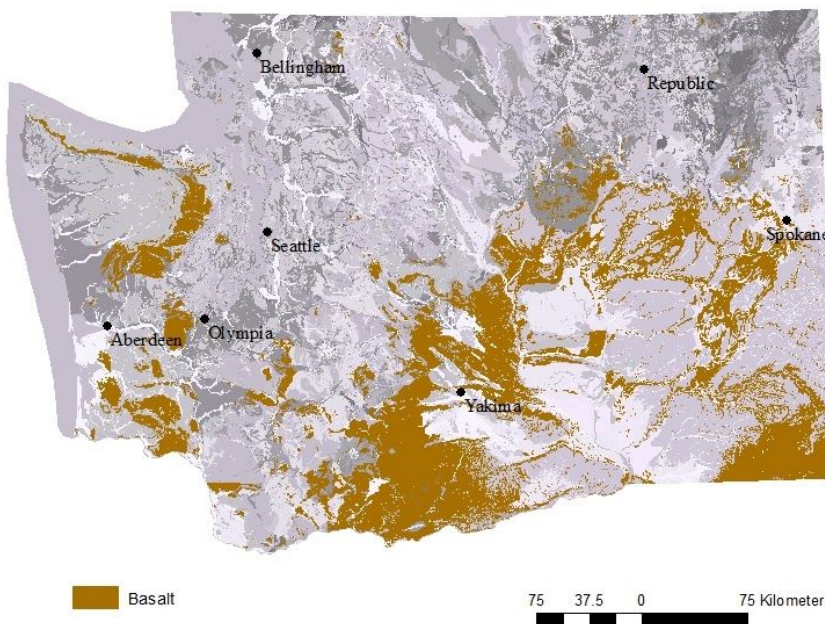


Figure 3.2. Distribution of basalt, overlain on the geologic map of Washington (WA DNR 2020). The Columbia River Basalt Group underlies the southeast quadrant of the state.

Opportunities in Juan de Fuca Basalts

Aside from the CRBG, the oceanic basalt on the Juan de Fuca plate off Washington's coast has also been investigated as a potential host rock for carbon sequestration (see Fig. 3.3) (Goldberg et al., 2018a). Goldberg et al. (2008) proposed that injecting CO₂ into off-shore basalts has distinct advantages over using on-shore basalts: larger capacity, long residence times for the fluids in the basalts, and several trapping mechanisms not available on land. The blanket of impermeable sediment on top of the oceanic basalts will prevent upward migration of the CO₂. Also, CO₂ hydrate, which is denser than seawater, will form at temperatures lower than 2°C. And

at depths of 2,700 m below water, injected CO₂ will be denser than seawater. Goldberg et al. (2008) estimated hundreds of gigatons of CO₂ could be stored in the Juan de Fuca basalts. Goldberg et al. (2018b) found the lateral extent of the proposed reservoir to be 50-100 km and the permeability of the upper 600 m of basalt to be ideal for CO₂ sequestration.

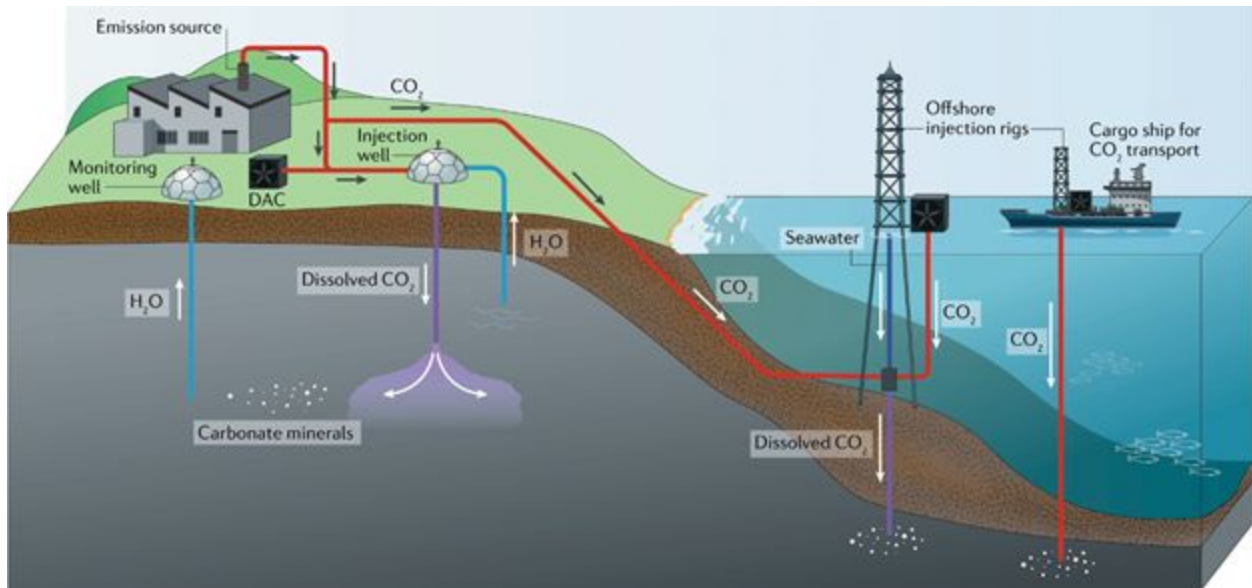


Figure 3.3: Hypothetical carbon mineralization scheme involving both on-shore and off-shore injection of CO₂ into basalts. Washington state is uniquely positioned to implement this type of scheme. Source: Snæbjörnsdóttir et al. (2020).

Conclusion

In situ remineralization of basalts, either on-shore or off-shore, is a physically feasible, proven method of sequestering and storing carbon in the long term. Extensive laboratory experiments, the Wallula project, the CarbFix project, and the CarbonSAFE program have shown the feasibility of this method and its applicability to a wide range of settings on Earth. Washington state has a unique opportunity when it comes to in situ basalt remineralization due to the presence of much of the Columbia River Basalts within its borders and the Juan de Fuca basalts nearby. By taking advantage of these resources, regional CO₂ emissions could be offset for the foreseeable future.

IV. In Situ Remineralization in Ultramafic Rock

Christopher R. Walker

Introduction

In situ carbon remineralization by CO₂ injection into ultramafic host rocks is another promising geologic sequestration method. Ultramafic rocks are igneous rocks with low silica content and high magnesium and iron. Ultramafic rocks form in the Earth's mantle and are exposed at Earth's surface in ophiolite sequences. Ophiolites are formed when an oceanic plate is thrust onto the edge of a continental plate at a convergent plate boundary, exposing a portion of the mantle to the Earth's surface. Although relatively rare compared to basalt, ultramafic rocks are relatively enriched in magnesium and therefore have the potential to sequester more CO₂ per volume of rock than basalt. To sequester carbon in the ultramafic rocks, the rock formations would need to be drilled and fractured. With the rock formation fractured, pressurized CO₂ would be injected in the new porous space.

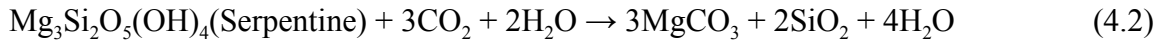
Geochemical Reactions for CO₂ Sequestration in Ultramafic Rocks

Two common ultramafic rocks are peridotite and serpentinite. Peridotite is a coarse-grained igneous rock consisting of olivine and pyroxene. Serpentinite is a metamorphic rock that is produced by the hydrous alteration of ultramafic rocks primarily made from magnesium silicate minerals.

Just as with basalt, carbon is sequestered when silicate minerals in the rocks dissolve in the presence of acidic fluids, and resulting cations combine with C and O to precipitate carbonate minerals. With the pressurized liquid CO₂ in contact with the ultramafic minerals, the divalent cations (Mg²⁺) will be released from the host rock and be in solution where they will slowly produce magnesite, a magnesium carbonate. Ultramafic rocks are unstable near Earth's surface, so the minerals are easily dissolved. The reaction involving the dissolution of peridotite and precipitation of magnesite and quartz is summarized (Paukert et al., 2012):



where forsterite (Mg_2SiO_4) dissolves, and magnesite (MgCO_3) is precipitated. In serpentinite, the reaction is similar (Park and Fan, 2004):



Because ultramafic minerals such as olivine and serpentine contain more divalent cations than mafic minerals such as pyroxene, they have the potential to sequester more CO_2 .

Laboratory Experiments in Ultramafic Rock

Although no large-scale field experiments have evaluated CO_2 injection into ultramafic rock formations, experimental studies and a well-documented natural example provide a foundation for future work.

One challenge for sequestration in any rock type is that low pH is needed to dissolve the host rock, but higher pH is required to precipitate carbonates. Park and Fan (2004) conducted an experimental study using serpentine minerals where they developed a method called the pH swing method. The concept behind the method is to lower the pH to very acidic levels to allow divalent cations to dissociate in solution. Once the cations are dissociated, the pH is increased to 8.6 - 9.5 for magnesium carbonate to precipitate out of solution (Park and Fan, 2004). The pH “swing” is a way to produce more magnesium carbonate to sequester additional CO_2 in mineral form than might be possible in conditions of constant pH.

Andreani et al. (2009) conducted an experimental study on carbon sequestration using dunite samples and monitored the changes in the hydrodynamic properties of the dunite. They were concerned about the possible feedback effects of the carbonation reactions and changes in the permeability of the reservoir (Andreani et al., 2009). They concluded that carbonate precipitation decreases porosity only in zones where diffusion-controlled transport is dominant. High flow rates decrease carbonation efficiency of the reservoir, but low flow rates could potentially reduce permeability and close the injection point (Andreani et al., 2009).

These two studies suggest that management of pH and flow rate could be important to successful sequestration ultramafic rocks.

Natural Carbon Sequestration in the Semail Ophiolite, Oman

The largest ophiolite in the world, the Semail Ophiolite, is located in the Sultanate of Oman. The ophiolite has been well studied for the feasibility of using peridotites for carbon sequestration. Listvenite is a fully carbonated ultramafic rock composed of magnesium carbonate and quartz that is present in the Semail Ophiolite. Falk and Kelemen (2015) use Rb-Sr to date listvenite in the Semail and conclude that it formed during the emplacement of the ophiolite. There are billions of tons of fully carbonated peridotite (listvenite) in the Oman peridotites, and all known listvenite deposits have formed within 500 meters of the basal thrust (Kelemen et al., 2011). The likely source of the CO₂-rich fluids necessary for this reaction to occur is from the metamorphic dehydration of the underlying sediments as the hot peridotite overthrust the underlying sediment (Kelemen et al., 2011). The listvenite deposits demonstrate that carbon sequestration from peridotite carbonation can happen naturally on large scales. This would further suggest that in situ remineralization of peridotite can offer a viable solution for carbon storage (Falk and Kelemen, 2015).

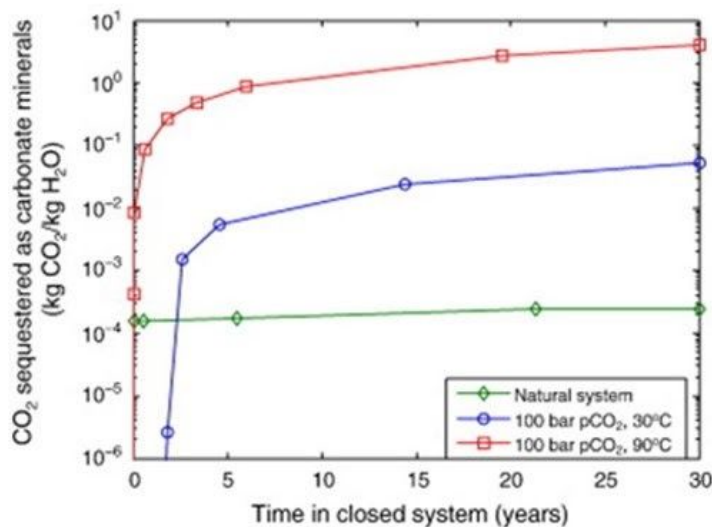


Figure 4.1. Modeled cumulative mass of CO₂ sequestered in three different scenarios: the natural system, CO₂ injection at elevated pressure and 30°C, and CO₂ injected at elevated pressure and 90°C. Reproduced from Paukert et al. (2012).

Based on the observed natural remineralization of the rocks in the Semail Ophiolite, the carbon sequestration potential for the remaining peridotites in Oman is approximately 1 Gt of CO₂ sequestered (Kelemen and Matter, 2008). Paukert et al. (2012) modeled reactions in the Semail Ophiolite to evaluate how to enhance natural carbon sequestration. Fig. 4.1 shows how the amount of CO₂ sequestered in a natural closed system differs from an enhanced system with

increased temperature and pressure (Paukert et al. 2012). Compared to the natural system (Earth surface temperatures and pressures), if CO₂ is injected at 90°C, at a partial pressure of 100 bars, then there will be an increase in the amount of carbon can be sequestered as a carbonate mineral. The reaction path modeling for this system highlights the natural pathway in an open and closed system. In the open system, surface water infiltrates the aquifer. The peridotite reacts with atmospheric levels of CO₂ and O₂ dissolving magnesium, calcium, silicon, and carbon dioxide. This solute-rich water infiltrates to further depths, where it is cut off from the atmosphere but continues to react with the peridotite in a closed system, precipitating serpentine, brucite, magnesite, and dolomite (Paukert et al., 2012). Magnesium, in this case, prefers to go to the secondary minerals chrysotile and brucite (Paukert et al., 2012) rather than for carbonates. This is a possible concern for large-scale sequestration projects.

Sequestration in Ultramafic Rocks in Washington State

Washington state has ultramafic rocks in the Fidalgo and Ingalls Ophiolite complexes as well as the Twin Sisters dunite (Fig. 4.2). The Fidalgo Ophiolite complex is located near the San Juan Islands, and the Ingalls Ophiolite is in the central Cascade Mountains. The Twin Sisters Range is part of the North Cascade Mountains. The Twin Sisters body consists of dunite that was accreted from oceanic terrains (Tikoff et al., 2010). The accumulated ultramafic rock outcrops in Washington State have a total area of ~413 km². By assuming a thickness of 1 km the total volume of rock, based on surficial data, is estimated at 413 km³ (257 mi³). By analogy with the Semail Ophiolite, the ultramafic rock in Washington may have the potential to sequester hundreds of Mt or more of CO₂. The advantage of these ultramafic rocks is their locations. They are near highly populated areas that are producing anthropogenic CO₂ and near natural gas pipelines making transportation from existing infrastructure possible. One major disadvantage to ultramafic rocks is there is very little natural permeability, so the rocks would need to be fractured before injecting CO₂, significantly limiting the volume of rock available for sequestration.

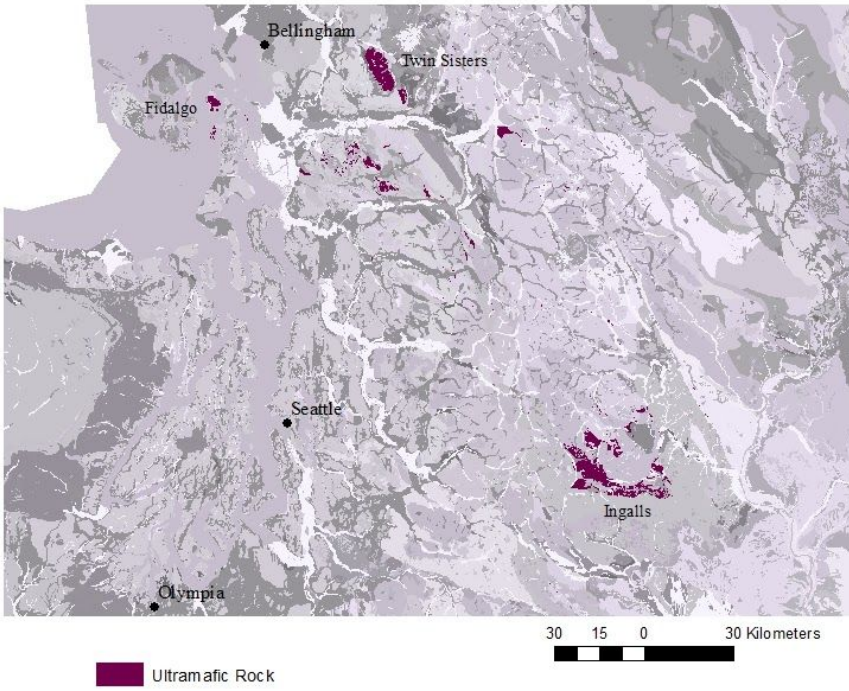


Figure 4.2. Distribution of ultramafic rocks in northwest Washington, overlain on the geologic map. Data from WA DNR (2020).

Conclusion

Ultramafic carbon sequestration is well studied for peridotites due to the natural formations of listvenite deposits, but very little is known about practical usage for serpentinite minerals in in situ remineralization outside of laboratory experiments (Thom et al., 2013). With the volume of ultramafic rocks in Washington State, these methods are worth exploring.

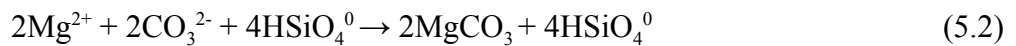
V. Sequestration by Enhanced Weathering

Jacob R. Childers

Introduction

An alternate approach to in situ remineralization is surficial CO₂ removal by enhanced weathering of finely ground silicate minerals sprinkled on cropland. It has been suggested that by grinding mafic and ultramafic rock rocks into a fine powder and sprinkling them in rivers, in the ocean, and on land, the high weathering rate of olivine can be used to passively capture and sequester CO₂. Grinding artificially speeds up the weathering process, the mechanism for removing CO₂ from the atmosphere on a geologic time scale (Schuiling and Krijgsman, 2006). In this section, I evaluate the possibility of utilizing surficial enhanced weathering in Washington, addressing the requirements for the reaction, the resources available in Washington, the potential negative environmental impacts, and the potential benefits aside from CO₂ sequestration.

Enhanced weathering relies on the dissolution of silicate minerals like olivine or pyroxene that contain divalent metal cations (Mg²⁺, Ca²⁺), thus mafic and ultramafic rocks are the main rocks types of interest. The minerals dissolve by reaction with carbonic acid, formed by the dissolution of atmospheric CO₂ in rainwater or groundwater, releasing the metal cations (Dessert et al., 2003; Schuiling and Krijgsman, 2006). This reaction is an intermediate step of equation 4.1, where olivine is dissolved and magnesite precipitates. The full reaction is shown in equations 5.1 and 5.2:



In equation 5.1, olivine is dissolved and the Mg²⁺ and CO₃²⁻ ions are released into solution. In equation 5.2, these ions come together to precipitate magnesite (MgCO₃). While mineralization relies on equation 5.2, enhanced weathering relies on equation 5.1. Reactions 5.1 and 5.2 show the dissolution of olivine; reactions are similar for other silicate minerals that contain divalent metal cations.

Once reaction 5.1 occurs, the dissolved bicarbonate ions and metal cations will flow into stream systems or the groundwater, only precipitating with certain soil chemistry not common in

Washington. The eventual goal is for the ions to reach the ocean, where they will increase ocean alkalinity, offset ocean acidification, and possibly precipitate into a carbonate shell or mineral and, at that stage, permanently sequester the CO₂ (Dessert et al., 2003; Schuiling and Krijgsman, 2006). A schematic of the enhanced weathering process is shown in Fig. 5.1. On geologic time scales, this method naturally removes CO₂ from the atmosphere, storing it as buried carbonate minerals.

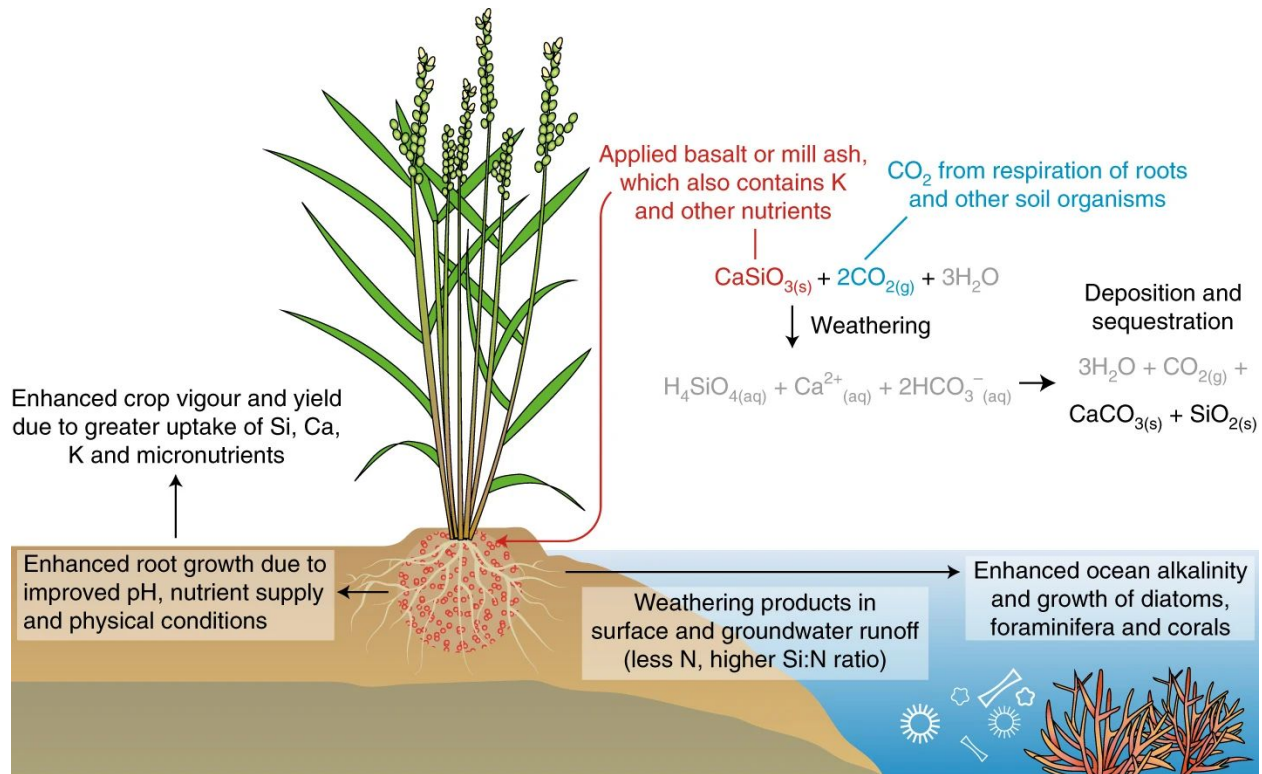


Figure 5.1. A schematic of the silicate weathering process. As silicate minerals weather, they provide nutrients to soils. Divalent metal cations can bond with dissolved CO₂, entering water systems and precipitation into carbonate minerals. Reproduced from Beerling et al. (2018).

Enhanced weathering depends on the dissolution of olivine in water or in the soil column, which, in turn, depends primarily on temperature, humidity, and grain size (Hangx and Spiers, 2009). The kinetics are not fully clear, with laboratory tested kinetics not fully matching results of small-scale field experiments (Amann et al., 2020; Dietzen et al., 2018; Peters et al., 2004; Renforth et al., 2015; ten Berge et al., 2012). However, it is clear that reaction rates are increased with higher temperature and humidity and with decreased grain size, which increases surface

area (Hangx and Spiers, 2009; Renforth et al., 2012). Outside the laboratory, temperature and humidity are limited by local climate and can only be fully leveraged in humid tropical regions. On the other hand, comminution (the grinding of rock into fine powder) can be controlled and optimized to encourage enhanced weathering in locations like Washington (Renforth 2012, Strefler et al., 2018).

A practical approach to implementing enhanced weathering may be the distribution of crushed rock on agricultural land, as there is established infrastructure to spread crushed rock (Renforth, 2012), and there are suspected benefits besides CO₂ sequestration, like promoting crop growth (ten Berge et al., 2012). To reduce the cost of implementing enhanced weathering, agricultural land must be sufficiently close to quarries where the crushed rock is sourced. A cost analysis by Strefler et al. (2018) suggests that agricultural land should be no more than 300 miles from the rock quarries. According to maps from the Washington State Department of Agriculture (WSDA) and the Washington State Department of Natural Resources (WA DNR), all agricultural land in Washington is within 300 miles of a mafic or ultramafic aggregate mine. This makes Washington a candidate for the implementation of enhanced weathering.

Experimental Evidence

There have been no large-scale field studies to test enhanced weathering as a CO₂ removal tool, only small-scale experiments, focusing mainly on the dissolution rate of olivine in soils, the effects of dissolved olivine on plant growth and soil chemistry, and the sequestration of CO₂. The major small-scale experiments were conducted by Amann et al. (2020), Dietzen et al. (2018), Renforth et al. (2015), and ten Berge et al. (2012). In all cases, they found that dissolution of olivine into soil increased soil pH, and there was no evidence of reduced plant growth. Most cases showed increased levels of nickel and chromium in the soil. Amann et al. (2020) observed a concentration above World Health Organization safe drinking-level standards but within the recommended limits for agricultural irrigation water. In all studies, smaller doses of olivine weathered faster, and more CO₂ was removed per unit of rock. Renforth et al. (2015) also focused on the optimal grain size for dissolution, considering differences in grain size has the largest impact on cost. They, and other studies (Beerling et al., 2018; Hangx and Spiers, 2009; Renforth et al., 2012; Strefler et al., 2018), recommend a grain size ranging from ~5 – 50

µm. While the dissolution of olivine in general is well studied, further research needs to be done to fully understand the dissolution rate of olivine in these circumstances.

In addition to testing for carbon sequestration potential, Dietzen et al. (2018) compared basalt to standard liming practices (using calcium oxide). Basalt contains olivine but also other silicate minerals that contain calcium. They determined that weathering of basalt increased soil pH to safe levels while still combating aluminum toxicity, and that there was no release of CO₂ from basalt-treated plots, compared to the 221% increase in CO₂ release from the lime-treated plots. They determined that basalt could possibly be an adequate replacement for lime, but that it may be preferable to apply smaller amounts of olivine over a larger area, maximizing CO₂ consumption rather than prioritizing liming replacement.

Relevant Resources in Washington

Potential implementation in Washington state is dependent on three things: abundance of mafic and ultramafic rock, abundance of accessible cropland, and financial support. According to the WA DNR (2011), there is ~380 km² of ultramafic outcrop (including dunite) and ~31,000 km² of mafic rock outcrop, primarily basalt (Fig. 5.2). Along with that, there are currently 169 aggregate mines mining mafic or ultramafic rock in Washington (WA DNR, 2020). A cost analysis from Renforth (2012) suggests that extraction is financially viable for depths up to only 50 m, and the majority of mines in question have permits up to or exceeding that depth (WA DNR, 2020). By analogy to Renforth's (2012) study in the UK, many of these mining facilities would also likely have the machinery required for comminution. The maximum CO₂ storage potential for dunite is 1.1 ton of CO₂ per ton of rock; for basalt, it is 0.3 tons of CO₂ per ton of rock (Strefler et al., 2018). This is the possible maximum storage and assumes optimal conditions like high humidity and high temperatures (Hangx and Spiers, 2009), so it can be assumed that storage potential will be somewhat less in the temperate, arid climate of eastern Washington.

As shown in Fig. 5.3, there is ~26,600 km² of currently used agricultural land in Washington (WSDA, 2020). Strefler et al. (2018) suggests a maximum of 15 kg rock/m² spread over agricultural land, spread out over multiple annual applications. This amount corresponds to an ~8 mm layer of crushed rock spread over the agricultural land. To give a maximum, though

highly unlikely value, almost 4 billion tons of rock could be spread, using all available cropland in Washington. If all agricultural land is used, then over the time it takes for all the rock to dissolve, a rough estimate of 4.4 Gt of CO₂ can be stored through ultramafic weathering, or 1.2 Gt of CO₂ can be stored through basalt weathering (approximately 10 years of Washington state emissions at current rates). This could be increased further by utilizing the ~450 km² agricultural land in Washington that is either left fallow or untended (WSDA, 2020), though actively growing agricultural land should be used whenever possible, as weathering of basalt and release of magnesium is increased (Hinsinger et al., 2001). However, the 4 billion tons that can be spread over agricultural land dwarfs the ~14,400 tons of aggregate mined yearly in Washington (NSSGA, 2015). Agricultural land is not the limiting factor for enhanced weathering. The limiting factors are the amount of aggregate mined per year, the cost of mining, comminution, and distribution, and environmental concerns (Renforth, 2012).

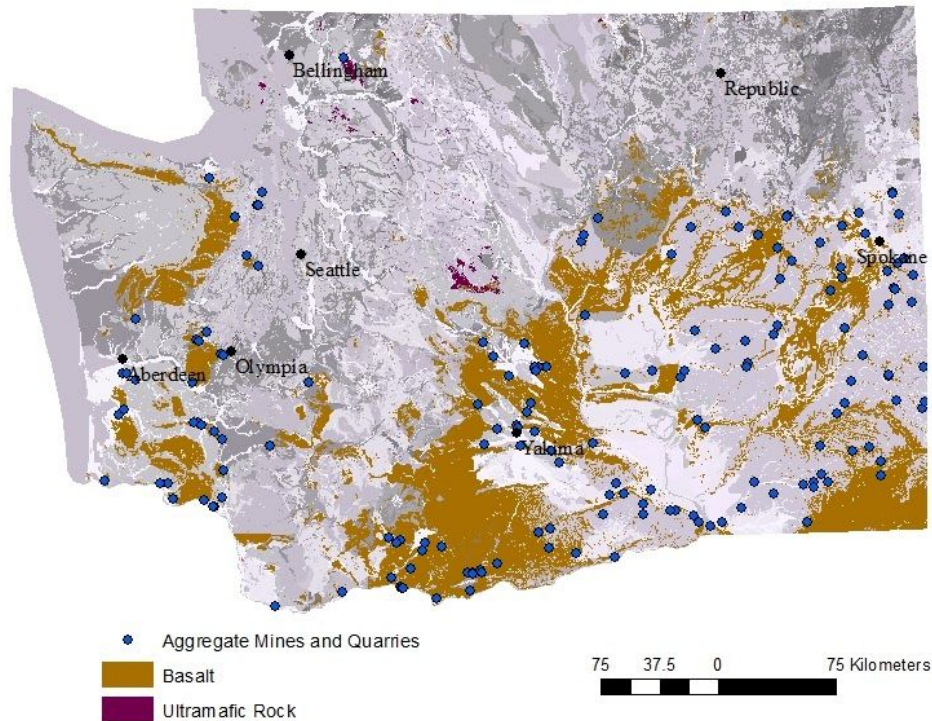


Fig. geologic map of Washington. There are ~31,000 km² of mafic rock (orange), ~380 km² of (purple), and 169 aggregate mines in either of these rock types (blue dots). Data from WA DNR (2011) and WA DNR (2020).

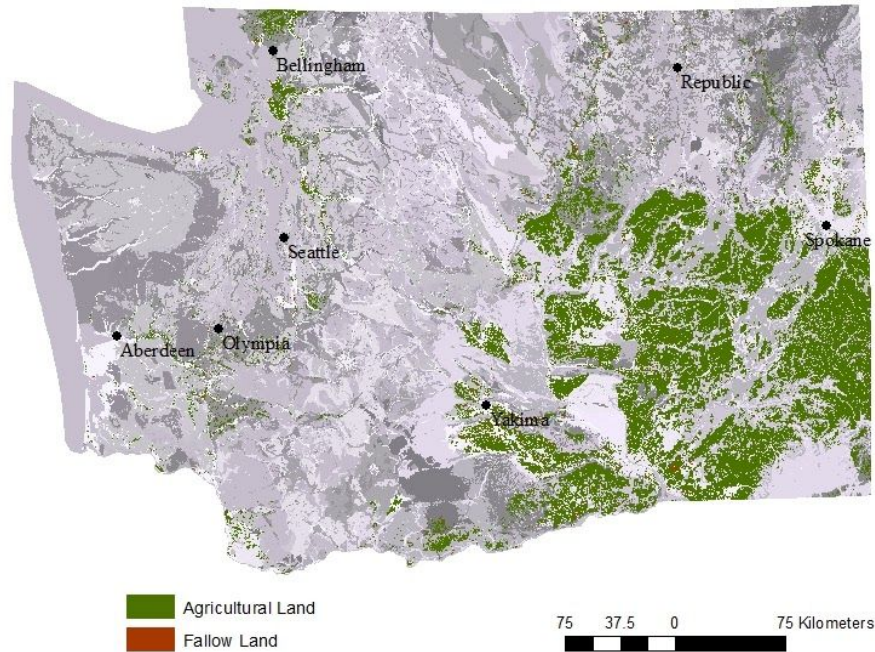


Figure 5.3. The distribution of cropland in WA state overlain on the geologic map of Washington. There is ~26,600 km² of actively used cropland (green) and ~450 km² of fallow or unused cropland (brown). Geology from WA DNR (2020), cropland distribution from with data from WSDA (2020).

Ancillary Benefits

Along with the potential for CO₂ sequestration, there are other benefits of an enhanced weathering scheme. Primarily, enhanced weathering would lead to increased ocean alkalinity and that could combat ocean acidification (Schuiling and Krijgsman, 2006; Taylor et al., 2016). Spreading of crushed basalt for enhanced weathering also has the potential to reduce soil erosion and replenish nutrients (Amann et al., 2020; Gillman et al., 2001) and to enhance crop growth (Dietzen et al., 2018; Hinsinger et al., 2001; Kantola et al., 2017; ten Berge et al., 2012). Also, it might be possible for basalt to be used as a replacement for limestone in the liming process, as pH is raised enough to combat aluminum poisoning without the drawback of CO₂ emissions from the dissolution of carbonates (Dietzen et al., 2018). Using basalt in the liming process also has the potential to reduce emission of N₂O, another greenhouse gas, from soils (Kantola et al., 2017).

Conclusion

Enhanced weathering is still a new technology ripe for exploration, and the Pacific Northwest is perfectly positioned to investigate its possibilities. Washington state has the relevant geology and agricultural industry to utilize enhanced weathering to sequester atmospheric CO₂. There is potential for significant sequestration of CO₂ and other ancillary benefits, like increased ocean alkalinity and a replacement for lime, but a large-scale study needs to be conducted to establish the viability of this potential tool.

VI. Improved Efficiency and Utilization of Abundant Seawater

Ryan W. Daniels

Introduction

Any of the methods introduced in the prior sections could be made more effective with improved technology. As reviewed in sections III and IV, in situ carbon remineralization is possible in Washington, and has shown promising long-term results in other settings (Islam et al., 2016). Methods to increase the efficiency will drive down cost and make it more viable to sequester carbon for long term or permanent storage, especially if we involve new techniques from research and development. Possibilities include sequestration using CO₂-enriched seawater, reducing pH with hydrofluoric acid (HF), the reduction of sulfate (SO₄²⁻) in injection fluids, and controlling injection thermodynamics.

Seawater

Sequestration by injection of CO₂-seawater is promising. CO₂ may be sequestered in the form of in situ mineralization, and this proceeds with the generation of carbonate minerals in the form of X_n(CO₃)_m (Fig. 6.1; Kelemen et al., 2019), including calcite, magnesite, dolomite, dawsonite, siderite, and strontianite. Divalent cations are naturally abundant in seawater, and enriching seawater with CO₂ will enhance the mineralization process (Wolff-Boenisch et al., 2011).

Wolff-Boenisch et al. (2011) investigate multiple injection solutions, ranging from freshwater to seawater, acidified freshwater to acidified seawater, and more complex variants. They find that silicate release rates (r_{Si}) of basaltic glass and crystalline basalt were comparable with that of peridotite when using seawater. Results suggest that for the purposes of in situ carbonate sequestration, CO₂-charged seawater injected into basalt might be nearly as efficient as injection into peridotite (Wolff-Boenisch et al., 2011).

Because Washington is a coastal state, seawater is readily available, especially for projects west of the Cascades or offshore, and seawater could be piped to dry areas of the state. For dry areas in Washington, seawater or synthetic solutions also may be transported by truck.

Seawater may even be pumped directly from the ocean to CO₂ processing plants for enhancement.

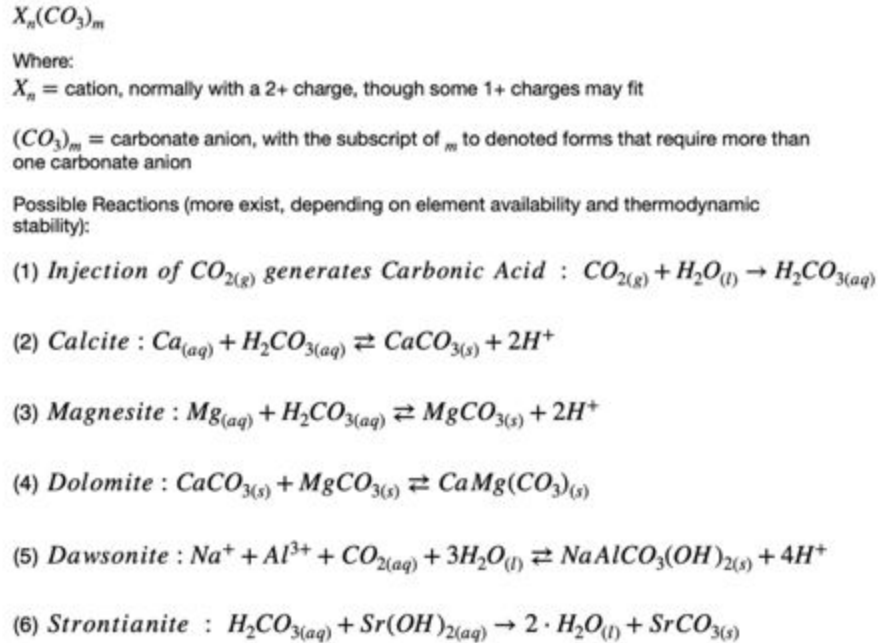
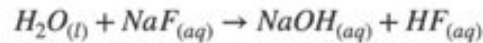


Figure 6.1. Reactions that produce carbonate species precipitated during in situ mineralization.

Hydrofluoric Acid

One method to increase storage space and available divalent cations for in situ geological carbon sequestration is to introduce hydrofluoric acid. Mineral reactions include the formation of carbonic acid (H₂CO₃) from the interaction of CO₂ plus H₂O (Wolff-Boenisch et al., 2011). The addition of sodium fluoride (NaF) to the naturally occurring fluoride in seawater allows the generation of HF when H₂O reacts with NaF (Fig. 6.2). As described in prior sections, rocks may be dissolved by carbonic acid to release divalent cations for reaction and react with HCO₃⁻ to form carbonates, but HF is far more efficient at dissolution because it is a much stronger acid (Wolff-Boenisch et al., 2011). Cations are normally the limiting reagent in these carbonate mineral reactions during sequestration. Additional host rock dissolution provides more cations and additionally may even increase the net hydraulic conductivity of the host rock (Wolff-Boenisch et al., 2011).

Sodium Fluoride Reaction Produces Strong Hydrofluoric Acid



Sodium Sulfate Reacts with Calcium Chloride to Produce Anhydrite

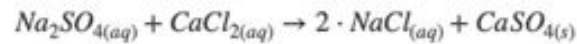


Figure 6.2. Referenced chemical reactions.

Wolff-Boenisch et al. (2011) found that the addition of NaF in seawater inlet solutions allows for a 1.70 to 1.86% enhancement of r_{Si} (Fig. 6.3), and it prevents the formation of anhydrite, $CaSO_4$, that would normally absorb available Ca^{2+} ions and block pore spaces (Fig. 6.2; Wolff-Boenisch et al., 2011). One problem is the NaF concentration would need to vary based on the availability of fluorine already in the seawater, from any seawater extraction location. To achieve this experimental result, Wolff-Boenisch et al. added 2.8-9.9mg NaF per 1 kg of seawater. Conversion of this cost to large scale production is an important issue, and more research on supply cost of NaF is needed to understand if it is a viable option.

Silica Release Rates:

$$r_{Si,geo} = \frac{F \cdot C_{Si}}{A_{geo} \cdot m}$$

$r_{Si,geo}$ = Si release rate per geometric surface area

F = pump flow rate

C_{Si} = concentration of Si in injection fluid

A_{geo} = specific geometric surface area

m = initial mass of material

Figure 6.3. Above are experimental silica release rates from the synthetic seawater-rock dissolution experiment by Wolff-Boenisch et al. (2011).

There are concerns around the presence of hydrofluoric acid in the natural environment. HF is a very strong acid that is normally used in many industrial applications, but it is toxic to

humans and wildlife. It is quite often used in geological and earth science laboratories to break bonds of silicates. The production of HF comes from its violent reaction with water, and the large amount of energy released. However, the final solution pH of chemical reactions in the laboratory using HF to enhance host rock dissolution rises to ~5.14 to 5.66, making it safe for the environment (Wolff-Boenisch et al., 2011). Even though HF has a potential to leak into the environment, unreacted HF will mainly be sequestered into the host rock pore spaces or equilibrated by natural alkalinity in the seawater, again raising the pH (Wolff-Boenisch et al., 2011). This reduces the concentration of HF to levels that are deemed safe, while utilizing the acidity it provides to dissolve minerals in subsurface wells. More investigation into the hazards of hydrofluoric acid in the subsurface needs to be evaluated.

Controlling Sulphate

An enhancement method discovered by Flaathen et al. (2011) concentrates on the control of sulphate as Na_2SO_4 in injection water to limit permeability reduction with mineralization. Carbonate precipitation may occur before ideal conditions are met in a reservoir, and this can lead to blockages that do not allow for the full potential of pore space to be utilized in a given host rock (Flaathen et al., 2011). The presence of sulphate in a reservoir inhibits carbonate mineralization, to a controllable degree, and this allows for deeper and more laterally extensive pore space to be utilized for mineral storage of CO_2 (Flaathen et al., 2011).

Anhydrite is a mineral that precipitates as calcium sulphate, CaSO_4 , in subsurface wells and reservoirs (Bader et al., 2007). This occurs most in situ carbon sequestration methods due to the presence of sulphate in injection water (Fig. 6.2). The Ca^{2+} cation is one of the major ions found in riverine, lacustrine, and the oceanic settings, so it is available to spontaneously react with SO_4^{2-} (Mackenzie and Garrels, 1966). The injection of Na_2SO_4 needs to be restricted by modulating the concentration added to inlet solutions. This presents a limitation as premature precipitation restricts the full potential to access host rock pore space, just as the CaCO_3 would, but not as fast (Flaathen et al., 2011). If some of the pore space is not utilized or blockages persist, then host rock will not be able to sequester as much CO_2 as projected. Nonetheless, as we strip SO_4^{2-} from waste water and seawater, we can control the amount that is precipitating by removing enough SO_4^{2-} species. This technique stems from technology currently in use in the oil

and gas industry (Bader et al., 2007). This limitation applies to all injection solutions containing SO_4^{2-} naturally.

Flaathen et al. (2011) agree with Bader et al. (2007) as they find that sulfate may be controlled by stripping excess from seawater, and if the amount of sulfate reintroduced is modulated, then divalent cations required plus CO_2 in solution will generate the most efficient synthetic mineralizing fluid. Sodium can even be used to mineralize CO_2 at high enough alkalinity, gained from seawater, pH ~ 8.4 (Wolff-Boenisch et al., 2018).

Controlling Thermodynamics

Another method of enhancement is to change the thermodynamic conditions such that mineral precipitation rates increase. A study by Clark et al. (2019) found that the mineralization of water dissolved CO_2 injected into basaltic rocks occurs within two years in field-scale settings, at 50°C and 80 bar. This study finds that varying the pressure and temperature of CO_2 -enriched water precipitates minerals at extremely quick rates, and in the study, they found carbonate mineral saturation 12 hours after beginning reactions in the experimental setup. This is consistent with modeling results reported by Paukert et al. (2012) for ultramafic rocks (section IV).

Injection pulsing may provide enhanced in situ geological carbon sequestration by cyclical pressurization of the host rock. In an experiment by Galeczka et al. (2013), dissolved cation concentrations were 100 times greater 10 hours after injection for trials involving CO_2 pulses than those at constant injection rates. Releasing more cations only continues to enhance the process.

Applicable Infrastructure

New infrastructure may need to be built in order for the described techniques to be implemented. However, the use of existing or abandoned industrial localities may prove cost effective, including the possibility of utilizing dry or abandoned wells (Fig. 6.4). Potential to modify or expand existing pipeline networks in the state exists, and this could be a useful transportation network for seawater to injection facilities. The potential to pipe seawater

around the state makes it far more interesting in attempting to sequester large quantities, especially for regions lacking in water supply, such as eastern Washington.

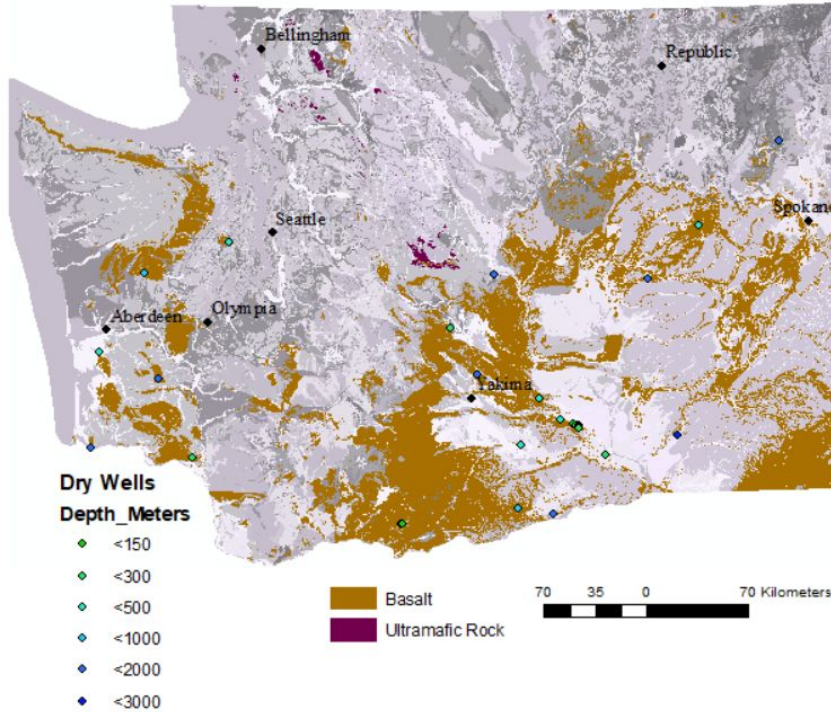


Figure 6.4. Exploratory dry wells that could be reevaluated for carbon sequestration (colored dots) in relation to mafic and ultramafic rocks, overlain on the geologic map of Washington. Data from WA DNR (2020).

Conclusion

Understanding what enhancements may influence in situ mineralization is an emerging field of geochemistry. Many experimental methods using technology such as CO₂-enriched seawater, reducing pH with hydrofluoric acid, the modulation of sulfate in injection fluids, and controlling injection thermodynamics show how adjustments to the in situ mineralization process can lead to increased mineralization. Quantifying the effects and comparison of different enhancements to geochemical injection solutions remains to be accomplished. However, the improved efficiency of in situ remineralization may be beneficial in the future through the progression of the geochemical discipline.

VII. Limitations And Concerns For Geologic Sequestration Approaches

Introduction

Many of the geologic carbon sequestration methods addressed above share common risks. Two of the most important concerns to consider are the impacts of natural and induced seismicity and the risk of contamination of the natural environment.

Natural seismicity

Natural seismicity can impact the integrity of a caprock keeping injected CO₂ safely stored. Faults can provide permeable pathways for CO₂ to leak out of the injection site and potentially be released into the groundwater or the atmosphere (Lewicki et al., 2007). The concern over leakage is compounded by the fact that western Washington is above an active subduction zone, one of the least favourable tectonic settings for sequestration due to the high natural seismic activity when compared to other tectonic settings (Baccu, 2003). This activity not only has the potential to fracture the caprock but may decrease the lateral extent of saline aquifers due to structural deformation, limiting the volume of CO₂ that can be sequestered in a basin (Maroto-Valer, 2010). Careful site selection avoiding active faults or regions of high seismic activity can help mitigate the risk of sequestration projects in active tectonic settings.

Induced Seismicity

Induced seismicity is one of the main concerns when injecting fluid into the subsurface. Wastewater disposal, hydraulic fracturing, and solution mining are processes that involve the injection of liquid into the earth and are known to trigger small-to-moderate earthquakes (e.g. Foulger et al., 2017). Just like these processes used in the oil and gas industry, injection of liquid or supercritical CO₂ or CO₂-charged water into bedrock has the potential to induce seismicity through the increase in pore pressure. The hazards include the annoyance of small felt earthquakes, structural damage from moderate earthquakes, or, possibly leakage of the sequestered carbon causing risks to the surrounding environment.

At least two demonstration projects demonstrate that induced seismicity can be avoided. One of the main goals of the JCCS Tomakomai CCS Demonstration Project was to establish a

reliable time series of regional earthquakes to understand the impact of injection on seismicity. This project found that there was no micro-seismicity around the reservoir site before or after the start of the carbon injection. While monitoring of the site will continue for another few years in order to establish data for post-injection seismicity, these results indicate that the injection process can be carried out with minimal increases in seismic activity. In addition, there was a natural earthquake that occurred during the project's duration. This event was found to have no impact on the temperature and pressure of the caprock (JCCS).

Injection of fluid into mafic rocks also poses a seismicity risk. During the initial phases of the CarbFix project, high injection rates induced seismicity, with the seismicity climax being two magnitude 4 earthquakes (Snæbjörnsdóttir et al., 2020). However, since that time, the injection rates have been lower and only one seismic event greater than magnitude 2 occurred in 2018 (Snæbjörnsdóttir et al., 2020). As mentioned above, injecting CO₂-charged water requires larger amounts of water than the injection of pure CO₂; that large fluid volume being injected into the subsurface poses a seismic hazard. This hazard can potentially be mitigated by drawing on subsurface water for the source of water to be injected (Snæbjörnsdóttir et al., 2020).

Leakage and Contamination

Many of the GCS approaches carry a risk of CO₂ leakage and the mobilization of heavy metals in the groundwater and soil. Both saline aquifer storage and sequestration in basalts carry the risk of leakage of the CO₂ out of the designated reservoir (Snæbjörnsdóttir et al., 2020). Although liquid CO₂ is easy to transport due to its low critical temperature, low pressure, and high density, it is buoyant, which is a difficulty injection schemes must overcome (Snæbjörnsdóttir et al., 2020). Due to its buoyancy, the CO₂ can migrate upward, out of the host rock, an issue difficult to mitigate when the host rock is a highly fractured volcanic formation such as the CRBG (Snæbjörnsdóttir et al., 2020). Leakage is normally a result of the physical and chemical trapping mechanisms being disrupted but can be exacerbated by increasing subsurface pressure with CO₂ injection (Newmark et al., 2010).

Research shows that it is unlikely that leakage will randomly occur, because there is usually some type of trigger that initiates sequestration failure. Unsealed faults, fracture zones, and improperly constructed or abandoned wells are ways for the CO₂ to migrate towards the

surface. Careful site selection can mitigate this risk: the identification of suitable caprocks for the Wallula project successfully prevented upward migration of CO₂ (Section III; McGrail et al., 2017b). New research has shown that the mineralization process may be faster than previously understood (Takaya et al., 2012). Over time, solubility and mineral trapping increases, decreasing the risk of leakage as the CO₂ is immobilized in an aquifer (IPCC, 2005; Jun et al., 2013).

One mitigation strategy for preventing leakage through upward migration is to dissolve the CO₂ in water before or during injection, as is done in the CarbFix project. This strategy prevents upward migration because CO₂-charged water is denser than subsurface water (Snæbjörnsdóttir et al., 2020). However, when charging water with CO₂, one environmental concern may be the large amounts of water required to dissolve significant amounts of CO₂. To mitigate this concern in some regions, the required water may be sourced from the target reservoir, which would have ancillary benefits of facilitating the monitoring of subsurface water chemistry and maintaining subsurface water pressure (Snæbjörnsdóttir et al., 2020).

Because CO₂ saturated water is highly acidic (Equation 2.1), there is the potential in sequestration projects for surrounding minerals containing hazardous heavy metals to be dissolved and mobilized (Xu et al., 2007). In the early stages of dissolution, metal ions released from dissolving basaltic minerals can reach higher concentrations than recommended for drinking water (Snæbjörnsdóttir et al., 2020). Consequently, leakage could be a problem for sites near important groundwater supplies, such as the saline aquifer sites in western Washington (USGS National Water Information System, 2003). However, over time, with the precipitation of carbonates and other minerals, the metals will be scavenged from the groundwater and the concentrations of dangerous metals will decrease, which will limit contamination of subsurface water or soils (Snæbjörnsdóttir et al., 2020). In the rare case of a leak reaching groundwater reported in the literature, the water remained potable after contamination (Lewicki et al., 2007).

The leading environmental concern regarding enhanced weathering is also the release of toxic trace elements, particularly nickel and chromium during the dissolution of olivine (ten Berge et al., 2012). Small scale experiments suggest that chromium and nickel concentrations in the soil column are increased during the enhanced weathering process. Due to the propensity of

nickel and chromium to stay in the soil column, prolonged implementation of enhanced weathering techniques may increase trace metal concentrations to unhealthy levels (Amann et al., 2020; Dietzen et al., 2018; Renforth et al., 2015; ten Berge et al., 2012). Research over an extended time period is required to evaluate the environmental risk (Amann et al., 2020; Dietzen et al., 2018). Ultramafic rocks, like dunite, are known to have much higher concentrations of nickel and chromium than mafic rocks, like basalt (Dessert et al., 2003, Hangx and Spiers, 2009), so we suggested the use of basalt in larger scaled experiments, even though it is less efficient at sequestering CO₂ (see section VI).

Other environmental concerns relate to the runoff of crushed rock used for enhanced weathering entering stream systems. Effects from increased turbidity and increased pH due to enhanced weathering are currently unknown (Beerling et al., 2018). However, in a large scale experiment involving weathering of powdered wollastonite (a calcium silicate mineral), researchers reported no ill effects on the nearby stream, with only a minor shift in pH (Peters et al., 2004).

In summary, with careful site selection, controlled injection, and careful monitoring, the major risks of geologic carbon sequestration related to seismicity and soil or water contamination can be successfully mitigated.

VIII. Estimating Implementation Costs

An accurate estimate of the costs is needed before a proper financial incentive can be created for any of these sequestration approaches. Estimates of the costs of sequestering carbon vary widely depending on which GCS method is used. While the costs of sequestering carbon in deep saline aquifers or by in situ remineralization of basalt can be estimated relatively accurately due to previous and on-going projects, the cost for other methods and proposed enhancements are less well constrained.

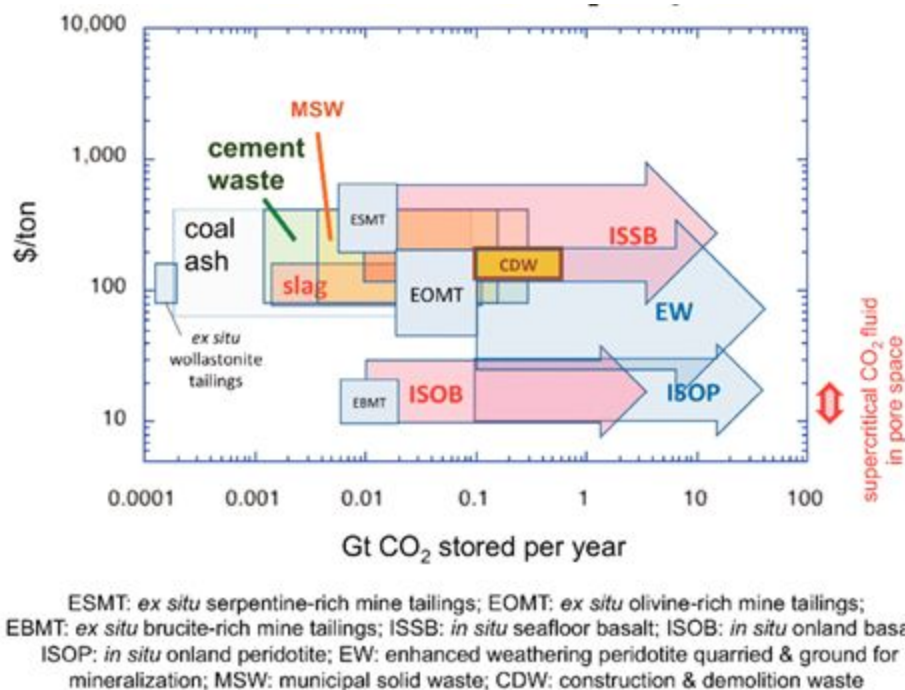


Figure 8.1: Cost of carbon sequestration via mineralization in various rock types and settings. Source: National Academies of Sciences Engineering and Medicine (2019). Sequestration in in-situ, on-land basalt (ISOB), such the Columbia River Basalt Group, is estimated to be one of the least expensive alternatives.

Enhanced oil recovery operations have given researchers accurate estimates for the costs of sequestering carbon in the deep saline aquifers. A recent National Academies of Sciences Engineering and Medicine report puts the costs of this GCS method at \$10-20 per ton of CO₂ (NASEM 2019). For the in situ remineralization of basalts, the costs of a future sequestration

project can be estimated from the CarbFix project in Iceland, which provides a good model for what could be implemented in Washington. Gunnarsson et al. (2018) report that the CarbFix project is sequestering CO₂ at a cost of \$25/ton. This is consistent with the NASEM (2019) report, which found that the in situ remineralization of on-land basalts and ultramafic rocks is the least expensive solid storage method at ~\$20/ton CO₂ (Fig. 8.1). The same report estimates the remineralization of seafloor basalts to cost an order of magnitude more, \$100-500/ton CO₂ (Fig. 8.1). According to Snæbjörnsdóttir et al. (2020), injecting CO₂-charged water is likely less expensive than injecting liquid CO₂ provided sufficient water is freely available, since less pressurization energy is required prior to injection.

The cost estimates for surficial enhanced weathering schemes range from ~\$60 per ton of CO₂ if using ultramafic rocks to ~\$200 per ton of CO₂ for if basalt is used, making it significantly more expensive than established in situ methods (Strefler et al., 2018). The main expense for this method is the comminution of the rock, with mining and transport costs being negligible (Strefler et al., 2018). The CO₂ sequestration potential is maximized with smaller grain sizes due to greater surface area and reactivity, but the cost increases with decreasing grain size (Schuiling and Krijgsman, 2006). Fig. 8.2 shows a summary of cost estimates for mining, comminution, and transportation. It is important to note that Fig. 8.2 takes into account transportation between mines and cropland with a maximum distance of 300 km, a criterion that all agricultural land in Washington meets (WA DNR, 2020; WSDA, 2020).

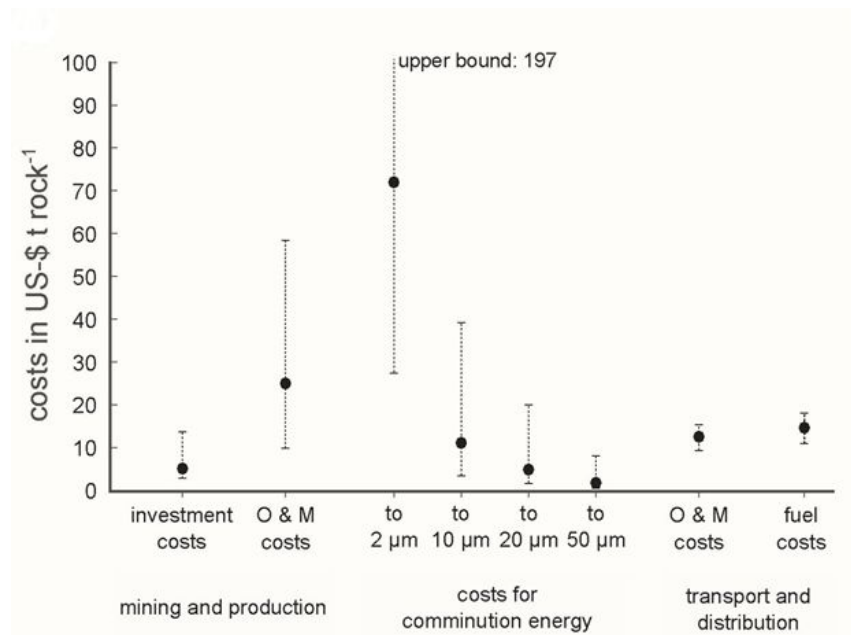


Figure 8.2. Cost estimates to implement enhanced weathering of mafic and ultramafic rock, per ton of rock. O&M: operation and maintenance. Cost for transportation is assuming a maximum distance of 300 km. Total cost is the sum of mining, comminution to selected size, and transportation, or about \$80/ton for 10mm particles. From Strefler et al. (2018).

Using these estimates, we can make a general estimate of the cost to achieve net-zero carbon emissions in Washington state. Assuming no further reduction in emissions from present values of 90 Mt annually, to sequester all the CO₂ that Washington produces with either in situ remineralization of basalt or injection of CO₂ into deep aquifers would cost in excess of \$2 billion, approximately 10% of Washington's FY2020 operating budget. This would clearly be a significant financial burden. Therefore, although the sequestration capacity exists, it is not economically feasible for the state to meet its net-zero target with geologic sequestration alone. If CO₂-emission reductions efforts increase, offsetting the final 4.5 Mt of emissions (set as a target for 2050) would cost ~\$90 million, less than 1% of the state's FY2020 budget (Wilson, 2020).

IX. Summary and Recommendations

In an effort to mitigate climate change, Washington state has an emissions target of no more than 50 million metric tons by 2030. By 2050, the goal is to achieve net-zero emissions by emitting less than 4.5 million metric tons and offsetting the CO₂ that is emitted (Department of Ecology, 2020). Carbon sequestration will be part of this strategy. Based on our review of geologic carbon sequestration methods, we make the following recommendations:

GCS should be incorporated into part of the emissions management portfolio to offset remaining emissions in Washington, but it is not financially viable, nor sustainable in the long term, to solely rely on GCS to offset carbon emissions. The combined CO₂ storage capacity of the CRBG and saline aquifers in Washington is over 100Gt, which is enough capacity to sequester 4.5 Mt of annual emissions almost indefinitely. With GCS strategies that focus on storage of CO₂ by converting it into carbonate minerals, all remaining CO₂ emissions in Washington can be permanently sequestered.

Carbon sequestration in saline aquifers and in situ basalt remineralization should be implemented as the main GCS strategies in Washington. These two methods have been proven viable, most recently in the Tomakomai and the CarbFix projects, which focus on sequestration in saline aquifers and in situ remineralization of basalt, respectively. Washington state is an ideal location for both of these methods and has extensive storage capacity with the presence of multiple saline aquifers and an abundance of basalt in the CRBG. This capacity can be increased with the potential use of off-shore sequestration in the Juan de Fuca basalts. With the proven success of these methods and Washington's geologic capacity, we believe that these two methods can play an integral role in achieving the state's emissions goals.

More research and large-scale field studies must be conducted to investigate in situ remineralization in ultramafic rock, surficial enhanced weathering, and enhancements using seawater before we can recommend their implementation. These methods have the potential to increase CO₂ storage capacity in Washington, passively sequester CO₂, or lower the cost and efficiency of GCS strategies. Washington state has the relevant resources to test these methods on a large scale, and upon completion of these large-scale tests, the role of these experimental methods in the strategy to mitigate CO₂ emissions can more accurately be determined. If any of

these methods are proven successful, we believe they could be valuable tools in mitigating emissions in Washington. Depending on future incentives for CO₂ reduction, these methods may even prove profitable by sequestering more CO₂ than is emitted from Washington alone.

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