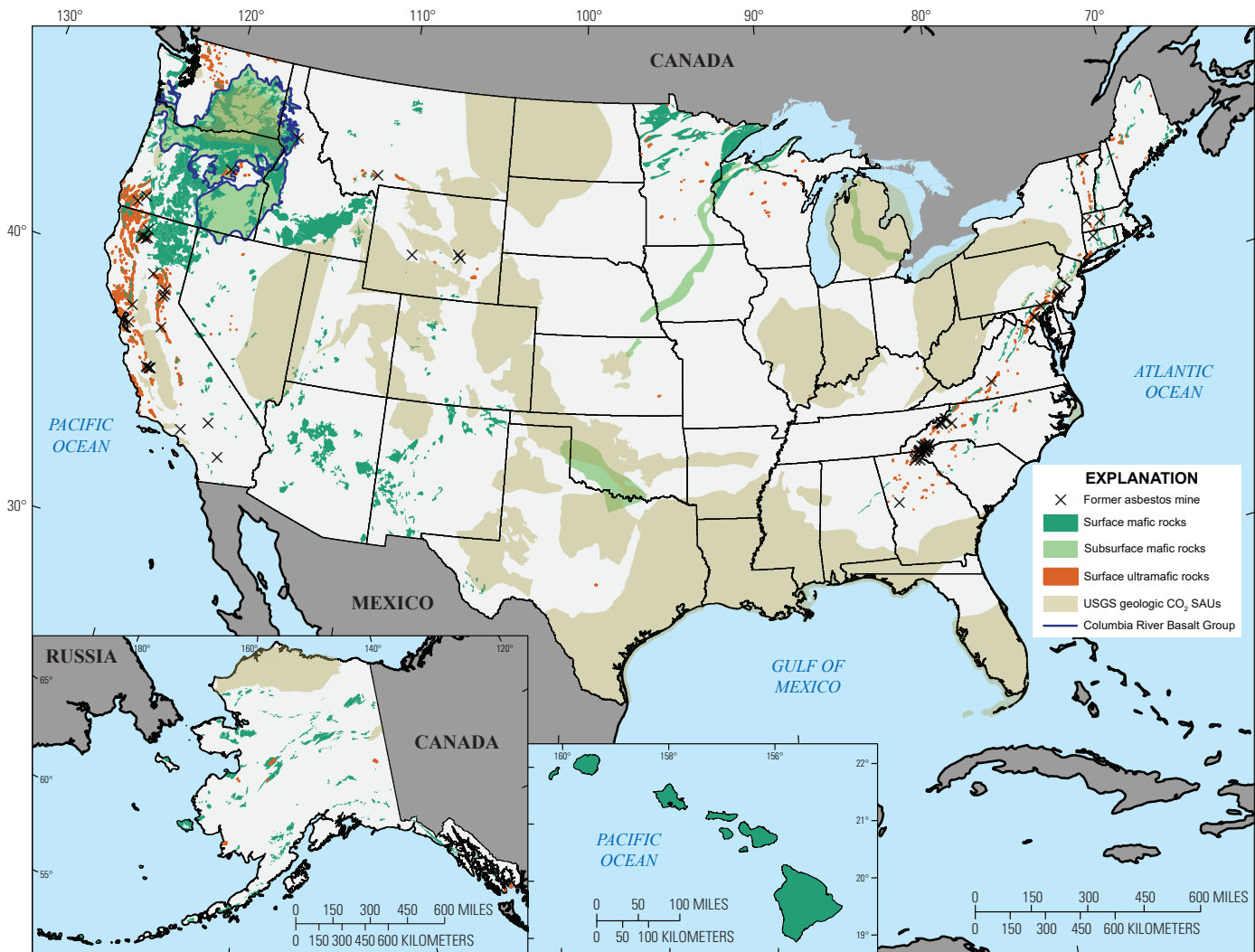


# Carbon Dioxide Mineralization Feasibility in the United States



Scientific Investigations Report 2018–5079

**Cover.** National map of mafic and ultramafic rocks in the United States. See figure 1.  
[USGS, U.S. Geological Survey; CO<sub>2</sub>, carbon dioxide; SAU, storage assessment unit]

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By Madalyn S. Blondes, Matthew D. Merrill, Steven T. Anderson,  
and Christina A. DeVera

Scientific Investigations Report 2018–5079

**U.S. Department of the Interior**  
**U.S. Geological Survey**

**U.S. Department of the Interior**  
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**U.S. Geological Survey**  
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U.S. Geological Survey, Reston, Virginia: 2019

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## Preface

The U.S. Geological Survey (USGS) has decades of experience in devising and testing methods for objectively assessing energy and mineral resources. Geologic storage is one potential method for sequestering atmospheric and anthropogenic CO<sub>2</sub>. The injection of CO<sub>2</sub> in sedimentary reservoirs is the most commonly discussed form of geologic CO<sub>2</sub> storage. This report discusses the feasibility of an alternative form of geologic CO<sub>2</sub> storage: CO<sub>2</sub> mineralization. In this method, storage is achieved by the formation of stable, solid carbonate minerals. This report was prepared in response to a request from the Senate Interior, Environment, and Related Agencies Subcommittee.<sup>1</sup>

The Survey is urged to conduct a study on the feasibility of carbon mineralization for permanent sequestration of carbon dioxide emissions, as described in the recently published report, “Rapid Carbon Mineralization for Permanent Disposal of Anthropogenic Carbon Dioxide Emissions,” with specific focus on the technical feasibility and potential sequestration sites. The Survey should consult with EPA, the Department of Energy, and other relevant agencies.

—Senate Committee on Appropriations, 2016, p. 37

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<sup>1</sup>Senate Committee on Appropriations, 2016, Department of the Interior, environment, and related agencies appropriations bill, 2017: U.S. Congress, 114th, Senate Report 114–281, to accompany S. 3068, 153 p., accessed July 1, 2017, at <https://www.congress.gov/congressional-report/114th-congress/senate-report/281/1>.



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## Conversion Factors

International System of Units to U.S. customary units

<b>Multiply</b>	<b>By</b>	<b>To obtain</b>
<b>Length</b>		
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
<b>Area</b>		
square kilometer (km <sup>2</sup> )	0.3861	square mile (mi <sup>2</sup> )
<b>Volume</b>		
cubic meter (m <sup>3</sup> )	35.31	cubic foot (ft <sup>3</sup> )
cubic kilometer (km <sup>3</sup> )	0.2399	cubic mile (mi <sup>3</sup> )
<b>Mass</b>		
kilogram (kg)	2.205	pound, avoirdupois (lb)
metric ton (t) [1,000 kg]	1.102	ton, short [2,000 lb]
metric ton (t) [1,000 kg]	0.9842	ton, long [2,240 lb]

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$^{\circ}\text{F} = (1.8 \times ^{\circ}\text{C}) + 32.$$

## Datum

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).



## Abbreviations

CAMP	Central Atlantic magmatic province
Ga	giga-annum, billion ( $10^9$ ) years before the present (A.D. 1950)
Gt	gigaton
km	kilometer
m	meter
<i>M</i>	molar
Ma	mega-annum, million ( $10^6$ ) years before the present (A.D. 1950)
$p\text{CO}_2$	partial pressure of carbon dioxide
ppm	part per million
t	metric ton
USGS	U.S. Geological Survey

## Chemical symbols used in this report

Ca	calcium
CaOH	calcium hydroxide
CO <sub>2</sub>	carbon dioxide
CO <sub>3</sub>	carbonate
Fe	iron
FeO	iron (II) oxide
Fe <sub>2</sub> O <sub>3</sub>	iron (III) oxide
H	hydrogen
H <sub>2</sub> S	hydrogen sulfide
Mg	magnesium
MgO	magnesium oxide
MnO	manganese (II) oxide
NaHCO <sub>3</sub>	sodium bicarbonate
NaCl	sodium chloride

### Minerals discussed in this report

$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	kaolinite
$\text{CaAl}_2\text{Si}_2\text{O}_8$	anorthite
$\text{CaCO}_3$	calcite
$\text{Ca}(\text{Fe},\text{Mg})(\text{CO}_3)_2$	ankerite
$\text{CaMg}(\text{CO}_3)_2$	dolomite
$(\text{Ca},\text{Mg},\text{Fe})_2\text{Si}_2\text{O}_6$	pyroxene
$(\text{Ca},\text{Na})(\text{Al},\text{Si})\text{AlSi}_2\text{O}_8$	labradorite
$\text{CaSiO}_3$	wollastonite
$\text{FeCO}_3$	siderite
$\text{FeCr}_2\text{O}_4$	chromite
$\text{FeS}_2$	pyrite
$\text{MgCO}_3$	magnesite
$(\text{Mg},\text{Fe})_2\text{SiO}_4$	olivine
$\text{Mg}_2\text{SiO}_4$	forsterite
$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$	chrysotile, antigorite, and lizardite pseudomorphs of serpentine
$\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$	talc
$\text{NaAlSi}_3\text{O}_8$	albite
$(\text{Na},\text{Ca})\text{Al}(\text{Si},\text{Al})\text{Si}_2\text{O}_6$	plagioclase
$\text{SiO}_2$	quartz

# Carbon Dioxide Mineralization Feasibility in the United States

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## Abstract

Geologic carbon dioxide (CO<sub>2</sub>) storage is one of many methods for stabilizing the increasing concentration of CO<sub>2</sub> in the Earth's atmosphere. The injection of CO<sub>2</sub> in deep subsurface sedimentary reservoirs is the most commonly discussed method; however, the potential for CO<sub>2</sub> leakage can create long-term stability concerns. This report discusses the feasibility of an alternative form of geologic CO<sub>2</sub> storage: CO<sub>2</sub> mineralization. In this method, CO<sub>2</sub> reacts with rocks and minerals to form solid and stable carbonate rocks. New pilot projects and laboratory-based kinetics experiments have revealed that this method, both in situ and ex situ, may be a viable option for storage. In situ storage targets in-place rocks at the surface or subsurface. Ex situ storage targets industrial byproducts at the surface like mine tailings. Environmental risks include induced seismicity for in situ methods if pressure is not managed properly, as well as potential water and land use effects. However, there are fewer long-term CO<sub>2</sub>-leakage concerns for mineralization methods compared to saline storage methods and therefore potentially lower long-term monitoring costs. The costs and benefits of CO<sub>2</sub> mineralization are compared to those of CO<sub>2</sub> storage in saline reservoirs using estimates of pressure-limited dynamic storage capacity. This report highlights the regional potential of areas in the United States for in situ and ex situ storage, as well as their proximity to potential sources of CO<sub>2</sub>. Especially suitable targets include asbestos or other ultramafic mine tailings, in situ ultramafic rocks on the East and West Coasts, the Columbia River basalts in the Pacific Northwest, the Midcontinent Rift basalts in the midcontinent, and the basaltic Hawaiian Islands.

## 1. Introduction

The capture and storage of carbon dioxide (CO<sub>2</sub>) has the potential to reduce CO<sub>2</sub> concentrations in the Earth's atmosphere. At significant scales, CO<sub>2</sub> storage may facilitate carbon-neutral electric power generation or provide other sectors with the ability to utilize existing industrial and mine waste. CO<sub>2</sub> capture, the process of obtaining the CO<sub>2</sub> to be stored, uses various industrial methods at point-source locations or, in the case of direct air capture, at any location.

CO<sub>2</sub> can be isolated from the atmosphere by physical, biological, or geochemical means (Stephens and Keith, 2008). Capture processes are not the focus of this report, though they are mentioned for context where appropriate. The U.S. Geological Survey developed a method for assessing storage resources for physical CO<sub>2</sub> storage, which is the injection of CO<sub>2</sub> into saline reservoirs and depleted oil and gas fields, leading to buoyant and residual trapping of supercritical CO<sub>2</sub> (U.S. Geological Survey Geologic Carbon Dioxide Storage Resources Assessment Team, 2013). When using physical trapping mechanisms in these types of reservoirs, CO<sub>2</sub> leakage can be a concern because the buoyantly trapped liquefied gas in depleted oil and gas fields needs to be physically stopped from reaching overlying aquifers or the surface through fractures or permeable rock. The residually trapped gas in saline formations can be improperly engineered, become buoyant, and subsequently leak from the storage reservoir.

An alternative to CO<sub>2</sub> storage as a fluid in sedimentary reservoirs is a geochemical method, CO<sub>2</sub> mineralization, which occurs via carbonation reactions. This method is akin to the natural weathering process whereby rocks and minerals with high magnesium (Mg), calcium (Ca), or iron (Fe) content react with CO<sub>2</sub> to form a stable and inert carbonate rock. In this process, the CO<sub>2</sub> is captured from the atmosphere or oceans during the reaction or supplied in a more concentrated form via injection into subsurface reservoirs or piping into surface industrial waste piles to significantly increase the rate of reaction. This report focuses primarily on the storage potential of mineralization reactions and does not discuss the capture or source of the CO<sub>2</sub>.

CO<sub>2</sub> mineralization reactions can utilize various materials in different settings and include the in situ CO<sub>2</sub> mineralization of basalts or ultramafic rocks (Matter and Kelemen, 2009; Gadikota, 2014; Gadikota, Matter, and others, 2014), the ex situ mineralization of alkaline mine tailings (Harrison and others, 2013; Gadikota, Natali, and others 2014; Gadikota and Park, 2014; Power and others, 2014; Pan and others, 2015), and reactions that produce other materials that have the potential to be used as mineral resources (Bobicki and others, 2012; Olajire, 2013). Kinetic experiments have shown fast reaction rates (review in Kelemen and others, 2011; more recent studies include Gadikota and others, 2014a), and recent pilot injection tests have shown greater than 95 percent CO<sub>2</sub>

## 2 Carbon Dioxide Mineralization Feasibility in the United States

mineralization in less than 2 years (Matter and others, 2016; McGrail and others, 2017). Building upon these results, the U.S. Department of Energy, National Energy Technology Laboratory, through its CarbonSAFE program, is funding a feasibility study on CO<sub>2</sub> mineralization of the oceanic basalts in the offshore Cascadia basin (Goldberg, 2017). The United States has extensive resources of suitable rocks, and here we investigate how these experimental studies and pilot projects apply to the mineral resources in the United States and whether CO<sub>2</sub> mineralization is an economically feasible technology.

Rock types suitable for large-scale CO<sub>2</sub> mineralization are those that contain Mg<sup>2+</sup>, Fe<sup>2+</sup>, or Ca<sup>2+</sup> cations. Suitable rocks that are abundant in the United States include the ultramafic rocks dunite, peridotite, and serpentinite and the mafic rock basalt. Dunite and peridotite consist principally of the magnesium iron silicate mineral olivine ([Mg,Fe]<sub>2</sub>SiO<sub>4</sub>), and serpentinite consists principally of the magnesium end-member serpentine-group pseudomorphs antigorite, lizardite, and chrysotile (Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>[OH]<sub>4</sub>). Basalt consists principally of plagioclase (anorthite-albite, CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>-NaAlSi<sub>3</sub>O<sub>8</sub>), pyroxene minerals ([Ca,Mg,Fe]<sub>2</sub>Si<sub>2</sub>O<sub>6</sub>), and olivine. Locally, very quickly chilled basaltic lava forms amorphous glass, which has available Mg<sup>2+</sup>, Fe<sup>2+</sup>, and Ca<sup>2+</sup> cations, rather than a crystalline rock. Although these minerals and basaltic glasses can vary in composition, we show here examples of only the most reactive simple endmember reactions to form carbonate minerals (table 1). We show the example reactions as simplified one-step processes; however, carbonate mineralization of silicates can occur by several multistep methods. In some

natural environments and engineered settings, a pH-swing process is used, which is a three-step reaction: (1) hydration of CO<sub>2</sub> to release hydrogen (H<sup>+</sup>) and carbonate (CO<sub>3</sub><sup>2-</sup>) ions into solution; (2) mineral dissolution to release Mg<sup>2+</sup>, Fe<sup>2+</sup>, and Ca<sup>2+</sup> cations into solution; and (3) ionic reactions to form insoluble carbonates (Gadikota, Swanson, and others, 2014). Alternatively, water dissolved into liquid or supercritical CO<sub>2</sub> is also highly reactive with Mg<sup>2+</sup>, Fe<sup>2+</sup>, and Ca<sup>2+</sup>-bearing materials (McGrail and others, 2009).

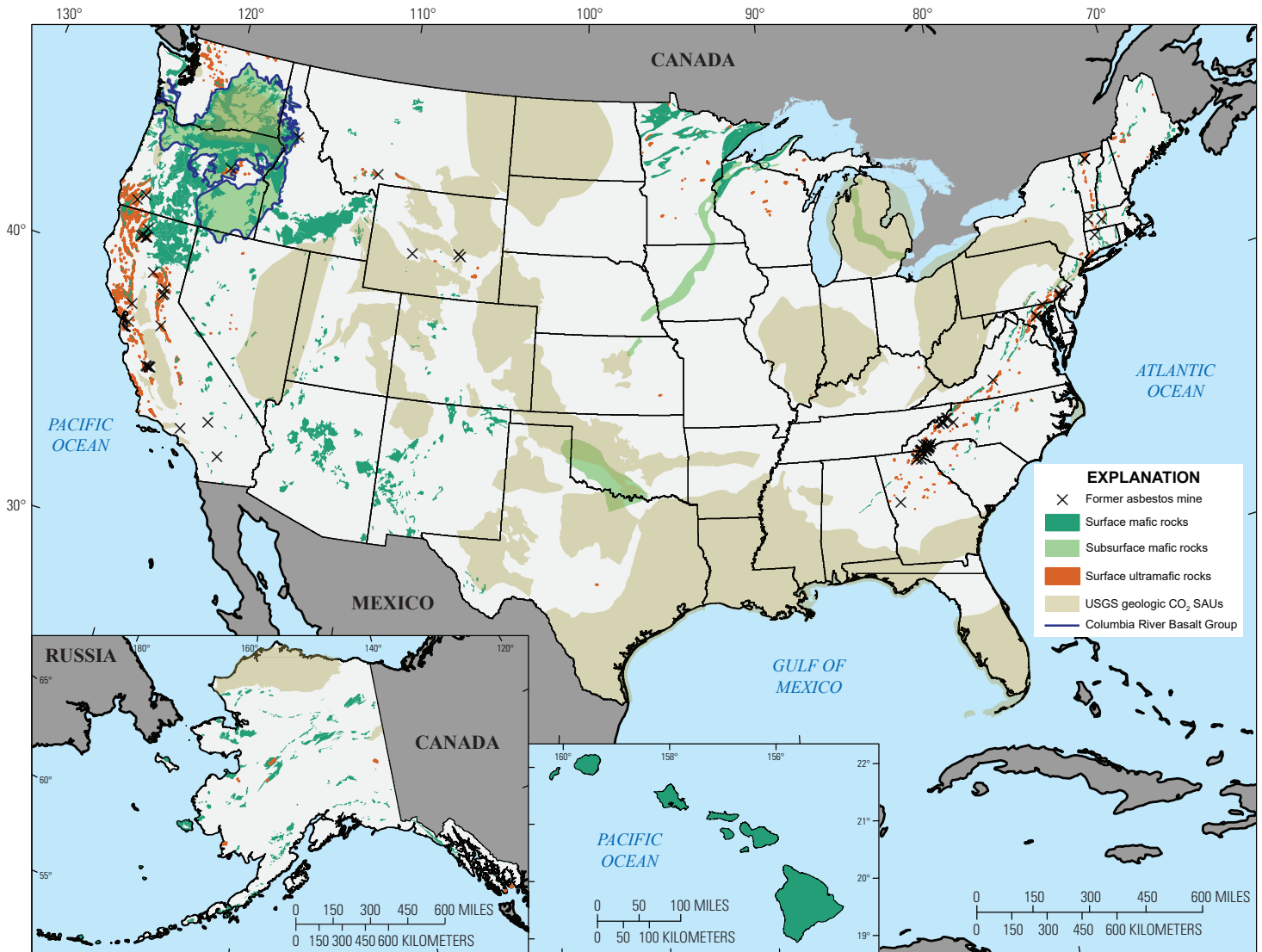
The feasibility of CO<sub>2</sub> mineralization in the United States is a function of many factors. First, it is necessary to know the location and abundance of the suitable rock types and the specific mineralogy and chemistry of these rocks. This is addressed in figure 1 (which shows location and areal extent of the appropriate rock types) and in greater detail in section 8 of this report, “Regional Carbon Dioxide Mineralization Feasibility in the United States.” It is then imperative to understand the extent and timing of the carbonation reactions, which are dependent on the kinetics of reaction for different minerals, as well as variables like temperature, pressure, the addition of catalysts, and the phase of CO<sub>2</sub>. Reaction rate experiments are discussed in section 2, “Reaction Rate Experiments and Models.” The extent and timing of carbonate formation are also highly dependent on the surface area of reaction. The surface area of reaction is itself a function of the porosity and permeability of the rock for in situ CO<sub>2</sub> mineralization, or the grain size of crushed rock for ex situ CO<sub>2</sub> mineralization. These properties are addressed in sections 3 and 4, “In Situ Carbon Dioxide Mineralization” and “Ex Situ Carbon Dioxide Mineralization,” respectively. To test

**Table 1.** Simplified carbonate mineralization reactions of common mafic and ultramafic minerals and basaltic glass.

Mineral or phase	Reaction	Notes
Olivine	$\text{Mg}_2\text{SiO}_4 + 2\text{CO}_2 \rightarrow 2\text{MgCO}_3 + \text{SiO}_2$ (olivine [forsterite]) + (carbon dioxide) → (magnesite) + (quartz)	Peridotite rocks consist of 40 to 90 percent olivine. Dunite rocks consist of >90 percent olivine. Olivine is also often found as crystals in basaltic lavas. Forsterite is the magnesium endmember.
Serpentine	$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 3\text{CO}_2 \rightarrow 3\text{MgCO}_3 + 2\text{SiO}_2 + 2\text{H}_2\text{O}$ (serpentine [for example, antigorite]) + (carbon dioxide) → (magnesite) + (quartz) + (water)	Serpentine is a hydrated form of olivine found in serpentinite rocks.
Plagioclase	$\text{Ca}_2\text{Al}_2\text{Si}_2\text{O}_8 + \text{CO}_2 + 2\text{H}_2\text{O} \rightarrow \text{CaCO}_3 + \text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ (plagioclase [anorthite]) + (carbon dioxide) + (water) → (calcite) + (kaolinite)	Plagioclase is the most abundant mineral in basaltic rocks. Anorthite is the calcium end-member.
Basaltic glass	$(\text{Mg}, \text{Fe}, \text{Ca})^{2+} + \text{CO}_2 + \text{H}_2\text{O} \rightarrow (\text{Mg}, \text{Fe}, \text{Ca})\text{CO}_3 + 2\text{H}^+$ (divalent cations in basaltic glass) + (carbon dioxide) + (water) → (magnesite, siderite, or calcite) + (hydrogen ions that can be consumed by dissolution reactions [Gislason and others, 2010])	Basaltic glass is an amorphous material formed during rapid cooling of basaltic lava with high concentrations of Mg <sup>2+</sup> , Fe <sup>2+</sup> , and Ca <sup>2+</sup> cations.

whether CO<sub>2</sub> mineralization techniques will work in natural settings, multiple large-scale pilot tests have been undertaken, all by in situ injection of CO<sub>2</sub> into basaltic rocks. These tests are discussed in section 5, “Pilot Projects.” To evaluate the feasibility of CO<sub>2</sub> mineralization, it is also important to discuss environmental effects, as well as economic constraints and

risk, which are sections 6 and 7, respectively. Lastly, in section 8, “Regional Carbon Dioxide Mineralization Feasibility in the United States,” we take a regional approach to understanding the strengths and weaknesses of different techniques of CO<sub>2</sub> mineralization, taking into account the availability of CO<sub>2</sub> from industrial sources.



**Figure 1.** National map of mafic and ultramafic rocks in the United States. Geologic data were compiled and, in some instances, simplified from Lambert and others (1988), Ojakangas and others (2001), Reed and others (2005), Sherrod and others (2007), Garrity and Soller (2009), Krevor and others (2009), and Barry and others (2013). Outlines of ultramafic-rock polygons were used to increase visibility. Asbestos mines were compiled from past producing mines described in Van Gosen (2006a,b; 2007a; 2008; 2010) and Van Gosen and Clinkenbeard (2011). U.S. Geological Survey (USGS) geologic carbon dioxide (CO<sub>2</sub>) storage assessment units (SAUs) were defined in the USGS national assessment of geologic CO<sub>2</sub> storage resources (U.S. Geological Survey Geologic Carbon Dioxide Storage Resources Assessment Team, 2013) to estimate CO<sub>2</sub> storage potential in saline reservoirs within sedimentary basins. The western Aleutian Islands in Alaska that do not contain relevant rock types are not shown. Base map was compiled from data from Environmental Systems Research Institute, Inc. (2007a,b) and Jarvis and others (2008).

## 2. Reaction Rate Experiments and Models

To understand whether carbonate mineralization is technically and economically feasible for storing CO<sub>2</sub> on reasonable time scales and to large extents, it is important to understand the rates of reaction for the relevant minerals. Kinetic dissolution and precipitation experiments on minerals (olivine and serpentine) and reactive whole rocks (basalt, serpentinite, peridotite, and dunite) have been performed for varying conditions, such as using a range of grain sizes, whole rock versus crushed samples, catalysts, different phases of CO<sub>2</sub> (aqueous versus supercritical), and a range of experimental designs.

CO<sub>2</sub> mineralization is commonly envisioned as a two-step process consisting of the dissolution of Mg-, Ca-, and Fe-bearing silicates (or basaltic glass) and the subsequent precipitation of Mg, Ca, and Fe carbonates. Dissolution is often thought to be the rate-limiting step; therefore, the kinetics of CO<sub>2</sub> mineralization for different minerals have been compared mostly by measuring the dissolution rates for different materials under varying conditions (Kelemen and others, 2011). Aside from the relatively uncommon mineral wollastonite (CaSiO<sub>3</sub>), olivine, particularly the Mg endmember forsterite, has the fastest dissolution rates and requires the least mass to store a given mass of carbon (Oelkers and others, 2008). Much of the experimental work on determining reaction rates for CO<sub>2</sub> mineralization has thus been done on olivine; for example, analyzing the effects of the partial pressure of CO<sub>2</sub> (pCO<sub>2</sub>), temperature, pH, dissolved sodium bicarbonate (NaHCO<sub>3</sub>), and dissolved sodium chloride (NaCl) (O'Connor and others, 2004; Hänchen and others, 2006; Chizmeshya and others, 2007; Prigiobbe and others, 2009; Gadikota, Matter, and others, 2014) and the addition of catalysts to increase dissolution rates (for example, Krevor and Lackner, 2011). Pyroxene in basalts has also been shown to be an important source of divalent cations for carbonation (Wells and others, 2017).

Other studies have focused on processes other than dissolution that can affect the rate of CO<sub>2</sub> mineralization. Saldi and others (2012) showed that under certain conditions magnesite precipitation can be three to four orders of magnitude slower than olivine dissolution and is the rate-limiting step. The formation of a passivating silica (SiO<sub>2</sub>) layer on mineral grain boundaries may inhibit dissolution and therefore restrict carbon mineralization (Béarat and others, 2006; Daval and others, 2011). However, precipitation rates increase in high-NaHCO<sub>3</sub> and high-pCO<sub>2</sub> fluids, perhaps because the passivation layers do not have a strong influence on inhibiting carbon mineralization in these environments (Chizmeshya and others, 2007; Kelemen and others, 2011). Direct carbon mineralization studies by Gadikota, Matter, and others (2014) showed that more than 70 percent of olivine was converted to

magnesium carbonate in 3 hours by reacting in a fluid bearing 1.0-molar (*M*) NaCl and 0.64-*M* NaHCO<sub>3</sub> in the temperature range of 150 to 200 degrees Celsius.

Serpentine dissolution kinetics are significantly slower than those of olivine (Park and others, 2003; O'Connor and others, 2004; Park and Fan, 2004; Daval and others, 2013) unless serpentine is heat treated (Gerdemann and others, 2007). In addition, it is possible to engineer a pH-swing process to optimize dissolution of serpentine at low pH and precipitation at higher pH (Park and Fan, 2004). Serpentine and serpentinites are much more mineralogically variable than olivine and peridotites, so a wide range of materials and techniques have been used in kinetic experiments. Experiments on serpentine have shown that both fast and slow dissolution regimes may exist (Gadikota, Swanson, and others, 2014). The CO<sub>2</sub> mineralization of existing serpentine mine tailings, including asbestos mine tailings, is discussed in section 4, "Ex Situ Carbon Dioxide Mineralization."

Basalt mineralization kinetics can be measured on whole rocks, volcanic glass, or plagioclase crystals that are the dominant phase of basalt. Of the phases analyzed by Oelkers and others (2008), plagioclase has the slowest dissolution rates and requires the most mass to store a given mass of carbon. Basaltic glass dissolution, however, is faster than the dissolution of serpentine, pyroxenes, and even olivine under some conditions (Oelkers and others, 2008; Kelemen and others, 2011). Schaef and others (2009) reacted mildly acidic, CO<sub>2</sub>-saturated water with rock samples collected from flood basalt provinces around the world, and they found that CO<sub>2</sub> was converted to carbonates in all samples. The proportion of volcanic glass was positively correlated with the reaction rate and was the dominant controlling factor. Experiments using supercritical CO<sub>2</sub> showed faster mineralization rates than those using aqueous CO<sub>2</sub> (Schaef and others, 2011). Though the rates and extents of reactions for basalts are less than those for olivine and serpentine, basaltic material is more widely distributed across the near surface and therefore is potentially more available for mineralization.

Reaction rates for minerals found in mafic and ultramafic rocks can be optimized by adjusting the temperature, pressure, and pCO<sub>2</sub>; using additives; and crushing rock to generate carbonates in both in situ and ex situ settings within a few years. These reaction rates are much faster than the CO<sub>2</sub> mineralization time scales (thousands of years) considered in earlier review papers that compared the implementation feasibility of various CO<sub>2</sub> storage methods (Benson and others, 2005, fig. 5.9). In section 5, "Pilot Projects," we describe two pilot projects using basalts that have shown nearly complete mineralization of injected CO<sub>2</sub> in less than 2 years (Matter and others, 2016; McGrail and others, 2017), though they have injected relatively small amounts of CO<sub>2</sub> compared to the reservoir volumes. Continued work on these projects may provide more applicable information on reaction rates as more CO<sub>2</sub> is injected.

### 3. In Situ Carbon Dioxide Mineralization

One potential method to store CO<sub>2</sub> through mineralization is by injecting CO<sub>2</sub> fluids into subsurface rocks without first mining or crushing the rocks. This process is known as in situ CO<sub>2</sub> mineralization and is possible in basalts and ultramafic rocks. Though the difference between basalts and ultramafic rocks, from a chemical standpoint, is a small change in the amount of Mg and Fe relative to silicon (Si), in general, these rock types have very different emplacement origins. The different origins lead to distinct textures, fracture densities, and porosities, which, in addition to the abundance of mafic elements, results in varied mechanisms of reaction. Most mafic rocks in the United States are extrusive igneous lava flows or near-surface sills and dikes, which exhibit high porosity and permeability in their vesicular flow boundaries and fractures (Reidel and others, 2002). Mafic rocks have a total mafic component (magnesium oxide [MgO], iron [II] oxide [FeO], manganese [II] oxide [MnO], and iron [III] oxide [Fe<sub>2</sub>O<sub>3</sub>]) content of 15 to 28 weight percent and SiO<sub>2</sub> content of 46 to 54 weight percent (Sen, 2014).

Ultramafic rocks in the United States are generally intrusive or deeply buried igneous or metamorphic rocks. These rocks have a higher total mafic component content of 35 to 46 weight percent and lower SiO<sub>2</sub> content of 42 to 48 weight percent (Sen, 2014). Overall, ultramafic rocks exhibit more variable reactivity than mafic rocks: ultramafic rocks contain the most energetically favorable minerals for reaction with CO<sub>2</sub>, but they generally have lower porosity and permeability and wide-ranging crystal sizes. Though more common in ultramafic rocks, if mafic or ultramafic rocks contain minerals that have been naturally carbonated, they become minimally reactive. The reactivities of olivine are much higher than those of alumino-silicate-bearing minerals and rocks (such as basalt, labradorite, and anorthosite) (Gadikota, 2014).

#### 3.1. Mafic Rocks (Basalt)

Mafic rocks, particularly large basaltic lava flows, offer voluminous potential sinks for CO<sub>2</sub> mineralization (Gislason and Oelkers, 2014). Abundant Mg<sup>2+</sup> and Ca<sup>2+</sup> cations are available for reaction in basaltic glass, plagioclase crystals, and olivine crystals (table 1). Large-volume basalts exist in the Columbia River Basalt Group, a continental flood-basalt province primarily in Washington, Oregon, and southern Idaho (figs. 1 and 2A) that erupted between 17 and 6 million years ago (Ma) (Tolan and others, 1989). These lava flows have abundant fractures as well as high-permeability and -porosity flow boundaries, interbedded with low-porosity flow interiors (Reidel and others, 2002). The high-porosity flow boundaries create a large surface area of reaction to form carbonates, and the low-porosity flow interiors act as vertical barriers to keep mineralization in the preferred zones (McGrail and others,

2006). The Columbia River basalts have been the focus of many studies on CO<sub>2</sub> mineralization (such as McGrail and others, 2006; Schaefer and others, 2009). Estimates of storage potential range from 10 metric gigatons (Gt) to 100 Gt (McGrail and others, 2006; Gislason and others, 2010). Recent results from a pilot project at the Wallula basalt pilot project in Washington (McGrail and others, 2017) are described in section 5.

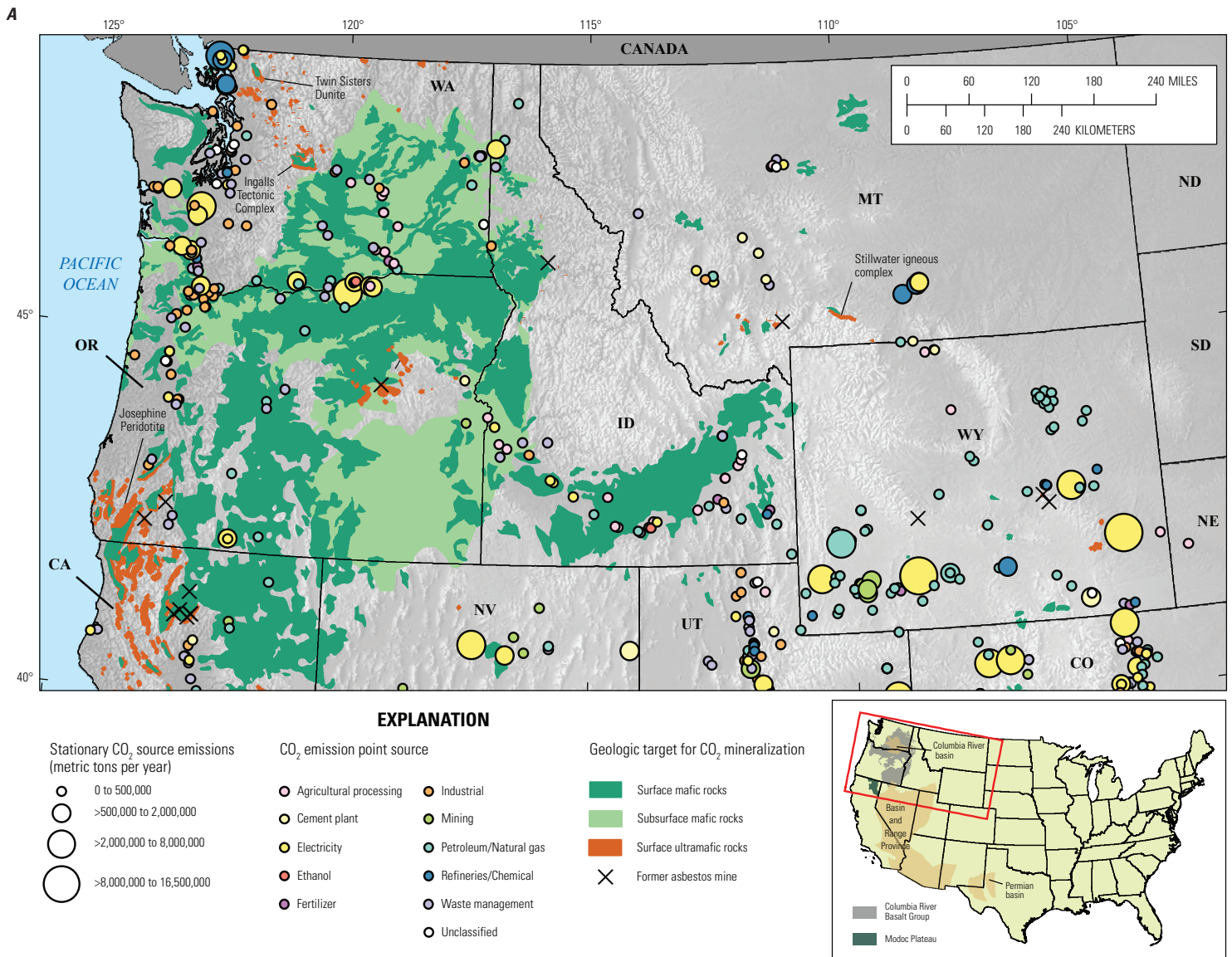
Older surface volcanic and subsurface intrusive basalts exist in the United States, including the Keweenaw Supergroup basalts (that formed at 1.1 Ga, or 1.1 billion years before the present) in the Midcontinent Rift (Cannon and others, 1989) and the Central Atlantic magmatic province (CAMP) basalts (that formed at 200 Ma, or 200 million years before the present) in Mesozoic rift basins both onshore and offshore of the East Coast (Goldberg and others, 2010). These older basalts, particularly those buried at depth, are likely less favorable for storage than younger surface lava flows because metamorphic and metasomatic reactions at depth generally reduce porosity and permeability and therefore reduce the availability for carbonation.

Deep-sea basalts, which are the main constituent of oceanic crust, are another potential target for CO<sub>2</sub> mineralization in basalts. At oceanic ridges, basalt is generated by sea-floor spreading and is highly fractured and permeable. Older basalts that are farther from spreading ridges lose porosity because of chemical weathering (Goldberg and Slagle, 2009). The estimated storage potential in basalts flanking the Juan de Fuca Ridge in an area roughly 200 to 1,000 kilometers (km) off the coast of the Pacific Northwest of the United States is 134 to 668 Gt CO<sub>2</sub> (Goldberg and Slagle, 2009). Other oceanic basalts include most of the Hawaiian Island chain and therefore the entire State of Hawaii (fig. 3). The recent CarbFix pilot project in Iceland (Matter and others, 2016), described in more detail in section 5, can be used as an analog for CO<sub>2</sub> storage in the Hawaiian Islands.

#### 3.2. Ultramafic Rocks (Dunite, Peridotite, and Serpentinite)

Unlike basalts, which have relatively homogenous whole-rock compositions, ultramafic rocks are compositionally heterogeneous and have highly variable porosity and permeability. Most ultramafic rocks near the surface are, or once were, peridotites or dunites that originated from the upper mantle beneath oceanic spreading ridges and were obducted onto convergent plate margins (Coleman, 1977). Since emplacement, these rocks have undergone variable levels of retrograde metamorphism (Kelemen and Hirth, 2012). Retrograde metamorphism can include hydration to form serpentinite and (or) carbonation to form retrograde-metamorphic assemblages that include Mg carbonates, commonly carbonate-veined peridotites, soapstone (magnesite and talc), and listvenite (magnesite and quartz). Dunite, peridotite, and serpentinite are all ultramafic targets for CO<sub>2</sub>

## 6 Carbon Dioxide Mineralization Feasibility in the United States



**Figure 2.** Maps of stationary point sources of carbon dioxide (CO<sub>2</sub>) emissions within 100 miles (160 kilometers) of mafic and ultramafic rocks in the Northwest (A) and Southwest (B) United States. Geologic data were compiled and, in some instances, simplified from Reed and others (2005), Garrity and Soller (2009), Krevor and others (2009), and Barry and others (2013). Outlines of ultramafic-rock polygons were used to increase visibility. Asbestos mines, also shown, were compiled from past producing mines described in Van Gosen (2007a, 2008, 2010) and Van Gosen and Clinkenbeard (2011). CO<sub>2</sub> point-source data are from National Energy Technology Laboratory Carbon Storage Atlas (2015). Base maps were compiled from data from Environmental Systems Research Institute, Inc. (2007a,b) and Jarvis and others (2008). Modoc Plateau outline is from Fram and Shelton (2014). Central Valley outline is from Faunt (2012).



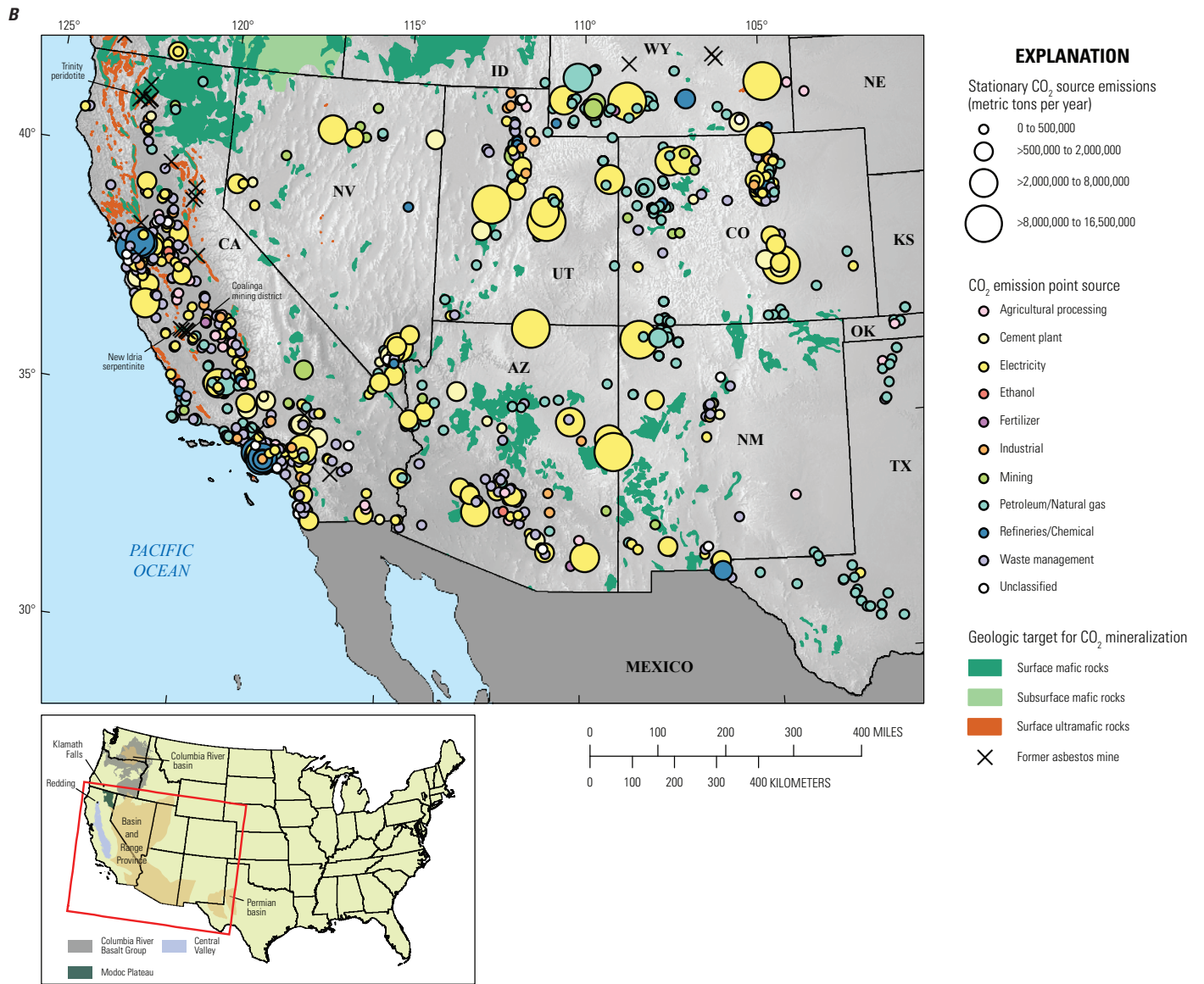
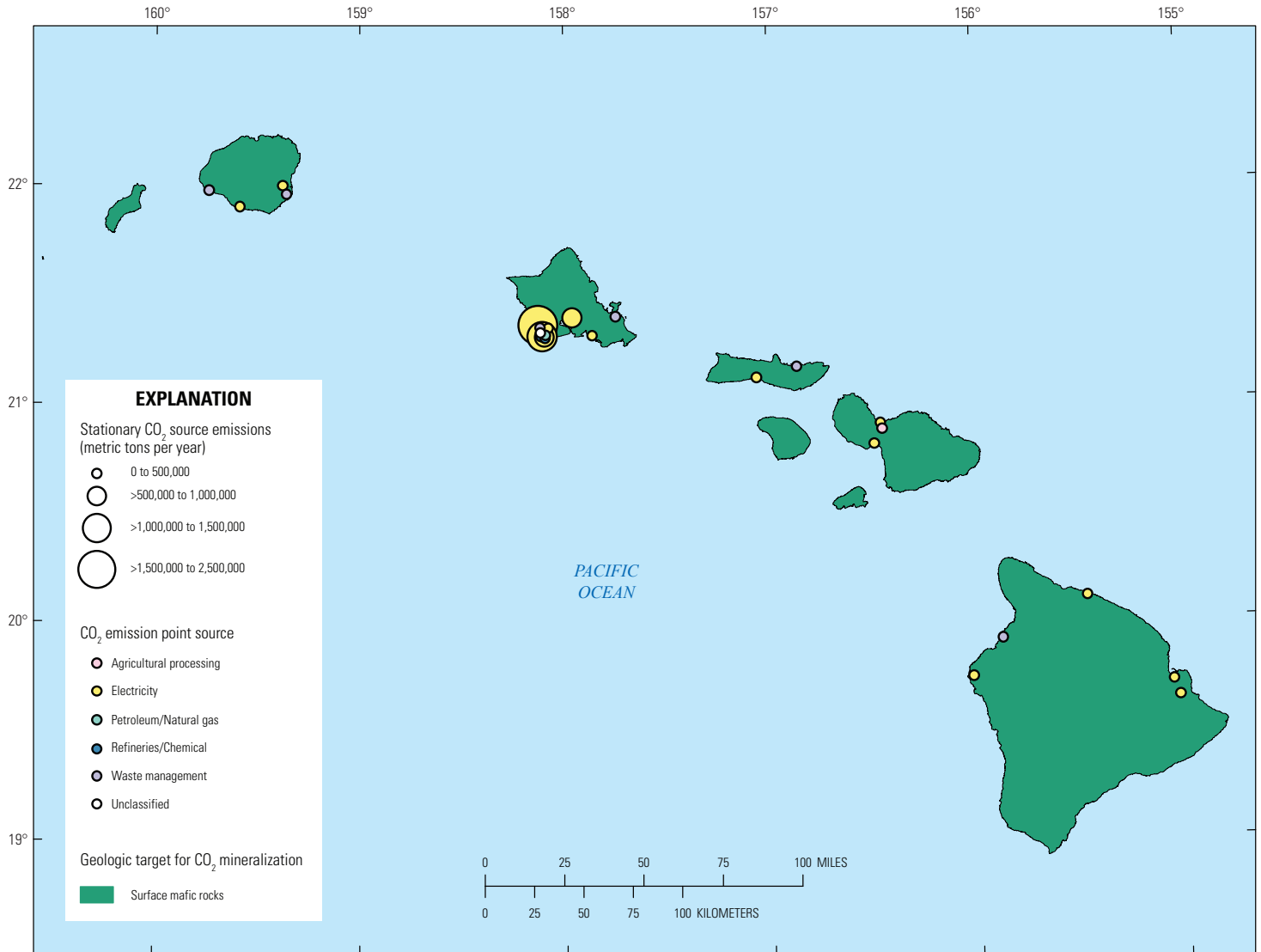


Figure 2. —Continued.

## 8 Carbon Dioxide Mineralization Feasibility in the United States



**Figure 3.** Map of stationary point sources of carbon dioxide (CO<sub>2</sub>) emissions and mafic rocks in the State of Hawaii. Geologic data were compiled and, in some instances, simplified from Sherrod and others (2007). CO<sub>2</sub> point-source data are from National Energy Technology Laboratory Carbon Storage Atlas (2015). Base map was compiled from data from Environmental Systems Research Institute, Inc. (2007a).

mineralization. Dunite and peridotite are typically partially hydrated (serpentinized) but still contain tens of percent Mg olivine. Fully hydrated peridotites (serpentinites) contain abundant Mg-serpentine minerals. Dunites, peridotites, and serpentinites that have naturally reacted with CO<sub>2</sub> to form soapstone or listvenite are not ideal targets for further CO<sub>2</sub> storage, particularly if that reaction has gone to completion. In the United States, ultramafic rocks suitable for CO<sub>2</sub> storage are generally found along the East and West Coasts in orogenic settings or as layered intrusions in intracratonic settings such as the Stillwater igneous complex in Montana (Goff and Lackner, 1998; Goff and others, 2000; Krevor and others, 2009).

To date, there have been no field-scale pilot projects of in situ CO<sub>2</sub> storage in ultramafic rocks, but one can gauge the feasibility of this technique by studying the laboratory reaction-rate experiments described in section 2 and particularly by investigating natural analogs (Matter and Kelemen, 2009; Kelemen and others, 2011). By examining natural settings where CO<sub>2</sub> has reacted with ultramafic rocks in near-surface environments, we can estimate reaction rates, extents of reaction, changes in porosity and permeability, and potential risks. It is particularly important to determine permeability and porosity changes in ultramafic outcrops because the reaction of CO<sub>2</sub> with olivine or serpentine to form

magnesite is both an exothermic and volume-increasing reaction. Whether the volume expansion inhibits or enhances the carbonation reaction is an open question. A laboratory-scale study showed that increasing magnesite formation reduced the accessible pore space in the olivine precursor (Gadikota, Matter, and others, 2014). This volume expansion may either clog the existing pore space, restricting the surface area of reaction such that the carbonation reaction ceases, or it may increase pore pressures beyond the fracture pressure, creating new fractures, greater permeability, and a self-propagating reaction-driven cracking system with ever increasing surface area of reaction controlled by CO<sub>2</sub> injection (Rudge and others, 2010; Kelemen and Hirth, 2012; Zhu and others, 2016). Lisabeth and others (2017) postulated that the propagating reaction is a function of chemo-mechanical coupling, whereby stress-induced dissolution leads to a reduction in permeability. Following mineral carbonation, the change in pore space in any given setting is dependent on a number of variables, including the pressure and temperature of the system and the mineralogy of the rock.

Examples of ultramafic rocks that have naturally reacted with CO<sub>2</sub> to form carbonates are in most convergent-margin settings where ophiolites exist. The Samail ophiolite in Oman and the United Arab Emirates shows evidence of both high-temperature mineralization in the listvenites and low-temperature mineralization in the carbonate veins (Kelemen and others, 2011; Falk and Kelemen, 2015). The fractures that contain the listvenite assemblage of magnesite and quartz are randomly oriented, suggesting that the fracturing is not tectonically driven but rather is derived from carbonate mineralization reaction-driven cracking (Falk and Kelemen, 2015). Listvenites in Atlin, British Columbia, Canada, show that the carbonation of serpentinites extends tens of meters from CO<sub>2</sub>-filled fractures. Though the intermediate reaction to soapstone generates increased fracture permeability, the full reaction to magnesite and quartz significantly reduces the porosity by filling the existing pore space, ending the ability to react more rock with CO<sub>2</sub> unless new fractures form (Hansen and others, 2005). Bruni and others (2002) and Cipolli and others (2004) analyzed natural springs in Genova, Italy, where high-pH, calcium hydroxide (CaOH) spring waters are indicative of serpentinite aquifers at depth that have reacted with CO<sub>2</sub> to form magnesite and chalcedony (SiO<sub>2</sub>) or amorphous silica (also SiO<sub>2</sub>). Their models showed a reduction of effective porosity in the aquifers, which was more pronounced if amorphous silica precipitated rather than chalcedony. Serpentinite ophiolite fragments in Norway also show evidence of a large-scale carbonation reaction to soapstone (magnesite and talc) and listvenite (magnesite and quartz) (Beinlich and others, 2012). Though carbonation reaction rates are slower in serpentinite than in peridotite, the resulting volume expansion is lesser, so there is a reduced possibility of clogging the pore space. Porosity and mass-balance calculations showed that the conversion of serpentine to soapstone proceeded isovolumetrically, and the reaction fronts were controlled by the CO<sub>2</sub> supply from alteration of the surrounding schist,

not by a reduction in porosity (Beinlich and others, 2012). Other examples include ultramafic-rock-hosted magnesite and talc deposits in Vermont (Robinson and others, 2006), the Advocate ophiolite to the northeast in Newfoundland, Canada (Bédard and Escayola, 2010), and the listvenites found in the Sartohay and Luobusa ophiolites in China (Robinson and others, 2005).

## 4. Ex Situ Carbon Dioxide Mineralization

Another possible method to store CO<sub>2</sub> through mineralization is by ex situ reaction with crushed material at the surface. Available, crushed solid reactants include mine tailings derived from mafic or ultramafic rocks and alkaline industrial wastes (Gerdemann and others, 2007; Renforth and others, 2011; Kirchofer and others, 2013; Power and others, 2013; Gadikota, Matter, and others, 2014; Power and others, 2014). When using crushed material instead of whole rock, the surface area of reaction is greater; therefore, the kinetics of the reaction are significantly faster, but the volume of resource available for carbonate mineralization is orders of magnitude less than that of in situ resources like basalts and ultramafic rocks. The faster reactions and readily available material, however, may prove to be more economic on small scales when attempting to create carbon-neutral electric powerplants or utilizing existing mine-waste material. The cost per metric ton (t) of CO<sub>2</sub> stored would be smaller for ultramafic rocks with abundant Mg<sup>2+</sup> and Ca<sup>2+</sup> cations available for reaction and larger for lithologies with fewer Mg<sup>2+</sup> and Ca<sup>2+</sup> cations, such as felsic rocks. Depending upon demand and cost, rocks could be mined and crushed for the specific purpose of ex situ carbon mineralization. If mining is done specifically for this process, however, there will be an increase in surface impacts, such as increased noise, truck traffic, and visibility of the mining operation and carbonate piles.

### 4.1. Carbon Dioxide Mineralization of Mine Tailings

Mafic and ultramafic mine tailings are generated during the extraction and production of asbestos, chromite, diamond, nickel, platinum-group metals, and talc. These tailings react passively with atmospheric CO<sub>2</sub> to produce carbonate minerals under natural conditions (Levitan and others, 2009; Wilson, Dipple, and others, 2009; Wilson, Raudsepp, and others, 2009; Pronost and others, 2012). Piping CO<sub>2</sub> into the tailings and designing tailings storage methods to favor increased reactions would significantly increase the carbonation reaction rate (Gerdemann and others, 2007; Harrison and others, 2013; Power and others, 2013; Power and others, 2014). Power and others (2014) and references therein reviewed strategies to increase tailings-hosted reaction rates, including bioleaching

of Mg silicates, increasing the CO<sub>2</sub> supply through heterotrophic oxidation, using bioreactors to induce precipitation of carbonate minerals, and other methods.

The most well-known and largest asbestos deposits in the United States and worldwide are those that have replaced or formed by metasomatic alteration of an ultramafic rock (Ross and Nolan, 2003; Van Gosen, 2007b). Most commonly, the host ultramafic rock is a dunite, peridotite, amphibolite, pyroxenite, or the hydrated equivalent, serpentinite. In the United States, large amounts of mine wastes composed of ultramafic rocks and serpentinite rock have been left behind at abandoned asbestos mines, which could offer potential sites and sources of material for pilot projects of ex situ CO<sub>2</sub> mineralization. Prominent examples occur in Vermont and California (Krevor and others, 2009), such as the abandoned Belvidere asbestos mine near Eden Mills in northern Vermont and the former asbestos mines of the Coalinga mining district in San Benito and Fresno Counties of west-central California.

## 4.2. Carbon Dioxide Mineralization of Alkaline Industrial Wastes

In addition to mining, many industrial processes also generate high-surface-area materials that could be potential sources of Mg<sup>2+</sup>, Fe<sup>2+</sup>, and Ca<sup>2+</sup> cations. Industrial byproducts such as fly ash, cement kiln dust, and iron and steel slag have been evaluated for their mineral carbonation potential (Huijgen and Comans, 2005; Gerdemann and others, 2007; Renforth and others, 2011; Gadikota, Matter, and others, 2014). Kirchofer and others (2013) found that 7.6 million metric tons of CO<sub>2</sub> (0.1 percent of U.S. total annual emissions) could be prevented or sequestered each year using these byproducts. Gerdemann and others (2007) noted that the small amount of total byproducts would restrict their use to niche applications where a hazardous waste could be remediated by carbonation or to help a company meet emissions targets.

Brines also have high concentrations of cations that could react to form carbonates. As a CO<sub>2</sub>-storage solution, the injection of a brine and CO<sub>2</sub> mixture into disposal wells could create scaling within the well and also mineralize the pore space at depth, creating pressure-management issues. However, there might be economic uses for the carbonates formed in reactions using salts from desalination processes. Haywood and others (2001) provided some discussion of these potential reactions.

## 5. Pilot Projects

Subsurface carbonate mineralization following the injection of CO<sub>2</sub> into mafic rocks has been demonstrated in two locations using different injection methods. Both projects concluded after 2 or more years of postinjection research that the injected CO<sub>2</sub> mineralized, a first to be demonstrated in the field (Matter and others, 2016; McGrail and others, 2017). Summaries of the results of these two projects, as well as discussions of past and potential injection projects, are provided in the following sections. It should be noted that the laboratory-based carbon dioxide mineralization research described in section 2 forms the foundation on which the pilot projects are based and is the source of the new technologies that developing projects may implement.

### 5.1. CarbFix and SulFix Projects, Iceland

CO<sub>2</sub> and combined CO<sub>2</sub>-hydrogen sulfide (H<sub>2</sub>S) gas-injection pilot projects at the Hellisheiði geothermal powerplant in Iceland have tested two injection systems named CarbFix and SulFix at neighboring sites. CarbFix mixes water with pure CO<sub>2</sub> or CO<sub>2</sub> and H<sub>2</sub>S at depth within the injection well, and SulFix mixes the gases with water at the surface before injection. The gas dissolution common to both systems limits buoyancy and the potential for leakage along fractures.

CarbFix 1, phase 1 injected 175 t of pure CO<sub>2</sub> during January through March 2012 (Matter and others, 2016). The CO<sub>2</sub>-water mixture was injected at a depth of about 500 meters (m) in the 2,000-m-deep injection well. The basalt storage reservoir lithology is present between depths of 400 and 800 m. Low-permeability hyaloclastites above the storage formation act as a sealing unit because of their low vertical permeability (Aradóttir and others, 2015). There are eight monitoring wells between 150 and 1,300 m in depth (Matter and others, 2016). CarbFix 1, phase 2 injected 73 tons of a CO<sub>2</sub>-H<sub>2</sub>S mixture, of which 55 tons were CO<sub>2</sub> from June through August 2012 at the same location.

SulFix 1 was similar to CarbFix except that the gas mixture was dissolved into the water at the surface and the amount of water used was greater. Concentrations of the gases in the water were between 350 and 550 parts per million (ppm) for CO<sub>2</sub> and 200 and 300 ppm for H<sub>2</sub>S compared to concentrations of 26,000 ppm and 6,800 ppm, respectively, for CarbFix 1, phase 2 (Aradóttir and others, 2015).

Isotopic and nonreactive but volatile chemical tracers were used to track the movement of dissolved gases in these pilot projects. After 2 years, the observed dissolved inorganic carbon and carbon-14 isotope spike concentrations were much lower than what would be expected for simple mixing between injected CO<sub>2</sub> and ambient groundwater (Matter and others, 2016). These results suggest that the precipitation of carbonates along the flow path accounted for more than 95 percent of the injected CO<sub>2</sub>. The presumed carbonate mineralization was corroborated by significant amounts of carbonate precipitation on a submerged pump that was recovered from a monitoring well. The reaction times appear to have been rapid. A larger industrial-scale injection project called CarbFix 2 began at the SulFix site in 2014. The results are not yet published in peer-reviewed journals.

## 5.2. Wallula Basalt Pilot Project, Washington

The Wallula basalt pilot project began in 2009 with the drilling of an injection well and extensive characterization of the Grande Ronde Basalt of the Columbia River Basalt Group in southeastern Washington State. A summary of the current results of the project noted that a 59-m-thick interval between depths of 828 and 887 m was selected for injection (McGrail and others, 2017). The potential reservoirs within the interval include three porous, low-density basalt flow tops. The upper two reservoirs are in the Indian Ridge member, and the lower reservoir is in the Ortlely member of the Grande Ronde Basalt. The sealing units are interflows with low permeability and high density, and they serve as flow barriers to the buoyant CO<sub>2</sub>. A total of 1,000 t of supercritical CO<sub>2</sub> was injected into the porous units over a 3-week period ending in August 2013. A daily limit of 40 t was established to keep the pressure within 30 percent of the hydrostatic reservoir pressure (McGrail and others, 2017). Comparison of preinjection reservoir rock and waters with 2-year post-injection samples indicated that a new carbonate mineral phase formed in the pore space of the reservoir as a result of the injected CO<sub>2</sub> rather than the dissolution and precipitation of preexisting carbonates.

Pore-water chemistry data indicated increases in total dissolved solids with major cation concentrations over initial baseline data. Fifty postinjection sidewall cores were taken in 2015 to investigate potential mineralization. Globular deposits of ankerite (a carbonate mineral in the dolomite group) were found in vugs and vesicles in an unknown number of those cores. Such deposits were not present in preinjection well cuttings or sidewall cores taken in 2009. Trends in Ca and Fe cation concentrations in the vugs showed higher Ca in the cores and increasing Fe moving outward, indicating dissolution of primary minerals and glass in the reservoir. Stable

isotope data from the ankerite nodules were similar to those from the source CO<sub>2</sub> and postinjection formation water but differed from calcite veins and other carbonates found during preinjection rock characterization (McGrail and others, 2017).

## 5.3. Other Pilot Injection Projects

In addition to the two injection projects discussed above, some smaller-scale field experiments have been completed, and researchers have plans for large-scale injection of CO<sub>2</sub> into mafic rocks. To evaluate the dissolution rate of Ca and Mg silicates, Matter and others (2007) injected 1,400 liters of CO<sub>2</sub>-saturated acidic water at a depth of 230 m into an 8-m-thick dolerite and sedimentary rock boundary layer at the bottom of the Palisades Diabase sill in New York. The Palisades Diabase sill is a thick sequence of Jurassic basalts intruded into the Triassic sedimentary sequence of the Newark Supergroup in the Mesozoic Newark basin. The acid was neutralized within hours, and the release rates of Ca<sup>2+</sup> and Mg<sup>2+</sup> available for mineralization were comparable to laboratory experiments.

A project funded by the U.S. Department of Energy to investigate the potential for CO<sub>2</sub> mineralization in oceanic basalts in the Cascadia basin, offshore Washington and British Columbia, may provide insight into offshore CO<sub>2</sub> injection and mineralization. At actively rifting (seismic) oceanic ridges, basalt is generated by sea-floor spreading and is highly fractured and permeable (Goldberg and Slagle, 2009) and therefore is a potential reservoir for injected CO<sub>2</sub>. Older basalts farther from spreading ridges lose porosity because of chemical weathering. The cited goal of the project is to complete an “integrated pre-feasibility study to characterize an ocean basalt reservoir for safe and permanent storage of 50 MMT [million metric tons] of CO<sub>2</sub>” (Goldberg, 2017, slide 3). The subsediment sea floor is composed of basaltic rock, and its use as a stable reservoir for CO<sub>2</sub> sequestration would be a significant development if proven feasible, successful, and economically viable.

## 6. Possible Environmental Effects

The most conspicuous advantage of carbonate mineralization is the stable sequestration of CO<sub>2</sub> gas. McCafferty and others (2016) noted that in general and compared to other methods of storage, the formation of stable minerals such as magnesite, dolomite, and calcite to store CO<sub>2</sub> gas does not have certain legacy issues like acid mine drainage and would require significantly less long-term monitoring. However, no industrial process is without potential hazards, and some potential environmental concerns have been raised, described below.

Induced seismicity is a potential hazard whenever fluids are injected into the subsurface (Zoback and Gorelick, 2012). Pressure-management techniques will be needed for subsurface CO<sub>2</sub> mineralization. This potential hazard is mostly dependent on the likelihood of the interaction between the pressure perturbation caused by CO<sub>2</sub> injection and existing faults. The injected CO<sub>2</sub> could also react with the minerals present in faults to alter the friction and permeability properties (White and Foxall, 2016). Site characterization within the expected areal extent of the CO<sub>2</sub> plume may identify faults near the injection well that should be avoided (Birkholzer and others, 2015).

Reports of induced seismicity directly associated with CO<sub>2</sub> injection have been limited to a few instances of measurable microseismicity and very few felt seismic events (Gan and Frohlich, 2013; White and Foxall, 2016). The largest induced seismic events (estimated moment magnitude greater than M3) have been documented in cases of wastewater injection into deep sedimentary formations when the injections were in the basement or lowermost sediments (Ellsworth, 2013; Keranen and others, 2014; McGarr and others, 2015). The depth of CO<sub>2</sub> injection, the proximity to the basement, and the potential for the resulting pressure front to invade deeper units are all relevant factors for CO<sub>2</sub> storage project operators to consider (White and Foxall, 2016).

Lisabeth and others (2017) suggested that the precipitation of certain minerals along fault surfaces may affect frictional behavior and the likelihood of induced seismicity. The authors also suggested that the mineral dissolution from reaction with injected CO<sub>2</sub> could concentrate stress and cause microcracking, which could maintain or increase the permeability of the rock. Although the microcracking could improve storage through permeability enhancement, it could also result in induced seismicity.

Injection of CO<sub>2</sub> into basalts has been accomplished only at the pilot project scale, but the limitations imposed on the CO<sub>2</sub> injection rate at the Wallula basalt pilot project closely approximate the pressure limitations typically assumed as necessary to model pressure-limited CO<sub>2</sub> storage capacity in sedimentary formations (Birkholzer and others, 2015; Jahediesfanjani and others, 2017). Some lessons may be gleaned from the geothermal industry. In a survey of the induced seismic responses to fluid injection in geothermal and CO<sub>2</sub> storage reservoirs, Evans and others (2012) found that fluid injection in geothermal wells in Iceland generally took place at very low pressure (or were even gravity driven) because the basalt reservoirs tended to have high natural transmissivity. High transmissivity generally promotes deep penetration of the pressure front generated by the fluid injection, which could increase the risk of induced seismicity. The authors also reported that these Icelandic basalt reservoirs were shallow, and the deepest injection well surveyed was 2.8 km deep. Despite the shallow depth of injection and low injection pressure, seismic responses to fluid injection were documented at four geothermal project sites. It is uncertain how analogous the fluid injection for these geothermal projects

is to the injection of CO<sub>2</sub> into basalts for the purpose of in situ storage via mineralization.

Aside from induced seismicity, other potential effects of CO<sub>2</sub> mineralization are currently under investigation. Guyot and others (2011) noted that disposal of CO<sub>2</sub> in the subsurface may change the behavior of microorganisms at depth and may lead to increased production of methane and H<sub>2</sub>S gases. The authors noted that these outcomes are not known results of injection but rather areas where the science could be improved through experiments in natural CO<sub>2</sub> accumulations, CO<sub>2</sub>-enhanced hydrocarbon production fields, and injection pilot projects.

In a review of the potential reaction-based pitfalls of industrial CO<sub>2</sub> mineralization, Haywood and others (2001), following methods from Newall and others (2000), documented the input requirements and byproducts generated by six ex situ mineral carbonation procedures. Aside from the energy and therefore financial cost required by many of the candidate procedures, environmental concerns such as large water requirements and the production of potentially harmful byproducts such as hydrochloric acid were noted. Ex situ CO<sub>2</sub> mineralization also may have drawbacks related to surface impacts. These include increased noise and traffic, as well as visibility of the mining operations.

As noted in the CarbFix injection project in Iceland and other locations, H<sub>2</sub>S gas may be associated with industrial CO<sub>2</sub>. Separating H<sub>2</sub>S from the CO<sub>2</sub> waste stream before injection is an additional cost burden that can be remedied through the co-injection of the gases. Schaefer and others (2010) found that co-injecting CO<sub>2</sub> and H<sub>2</sub>S in basalts resulted in combined carbon and sulfur mineralization, forming carbonate minerals plus pyrite and (or) sulfate minerals. When subjected to oxidative alteration, pyrite weathering can form acidic waters. Schaefer and others (2010) noted that although the pyrite began to inhibit carbonate formation as it grew on reaction surfaces, the carbonates eventually formed, and sulfur was incorporated in sulfate minerals.

## 7. Economic Constraints and Risk

The often-cited Intergovernmental Panel on Climate Change (2005) range of estimated costs for buoyant and residual saline storage (\$0.5–\$8 per t of stored CO<sub>2</sub>) is commonly compared with the estimated costs of in situ CO<sub>2</sub> mineralization (\$50–\$100 per t of stored CO<sub>2</sub>) (Intergovernmental Panel on Climate Change, 2005; 2007). The economic value of the benefits of carbon mineralization's long-term stability and lower leakage risks, costs of monitoring, and costs of long-term liability coverage are suggested as possible reasons why in situ mineralization could be preferred to storage in saline aquifers. However, the benefits of carbon mineralization are not well quantified, and economic comparisons based solely on estimated costs, without considering the relative risks, clearly favor storage in saline aquifers.

Given the lack of valuation estimates for in situ mineralization's stability benefits, it is useful to compare the costs of in situ mineralization with the costs of lower-risk saline storage. Zoback and Gorelick (2012) and others (Birkholzer and others, 2015; White and Foxall, 2016; Anderson, 2017b) have suggested that pressure buildup is a major source of potential risk associated with CO<sub>2</sub> injection into saline aquifers. Therefore, cost estimates for pressure-limited storage could be more appropriate for comparison with the estimated costs of CO<sub>2</sub> storage via in situ mineralization. Unfortunately, even recent studies do not account for the costs of the pressure management that will likely be necessary for storage in saline aquifers (Rubin and others, 2015; Anderson, 2017a).

Dynamic storage capacity can be defined as the maximum volume or tonnage of CO<sub>2</sub> that can be injected (stored) per year (Gorecki and others, 2014). On the basis of the results of saline storage-reservoir simulations, Jahediesfanjani and others (2017) reported estimates of pressure-limited dynamic storage capacity for a case study of saline storage (without brine extraction) in the Mount Simon Sandstone in the Illinois basin, and Anderson and Jahediesfanjani (2017) listed estimates of the potential costs of pressure-limited storage for the case study. Representative capacity and cost estimates from this ongoing work are listed in table 2 for comparison with the costs of CO<sub>2</sub> storage via in situ mineralization. In addition, the costs of brine extraction estimated by Harto and Veil (2011) have been added to the estimated costs of sedimentary reservoir storage without brine extraction (Morgan and Grant, 2014), and the estimated costs are listed in table 2. Harto and Veil (2011) assumed extraction of brines at a ratio of 1:1 per volume of CO<sub>2</sub> injected, which could nullify pressure buildup, minimize associated risk, and make the entire volumetric storage capacity accessible to CO<sub>2</sub>. The dynamic capacity is greatest for storage in saline reservoirs with brine extraction (table 2; sections 1 and 2). When brine extraction or other pressure-management techniques, such as reducing injection rates, are not considered, the cost is seemingly low. However, when the costs of pressure management are considered, the estimated costs for saline storage are comparable to CO<sub>2</sub> mineralization (table 2).

The significant economic advantages of CO<sub>2</sub> storage via ex situ mineralization over the other storage options in table 2 are mostly due to the location of storage operations being on (or very near) the surface. These advantages include an absence of costs for injecting CO<sub>2</sub> underground, zero expected reservoir pressure buildup or underground migration of CO<sub>2</sub>, low costs to recover marketable mineral carbonate commodities produced as a result of mineralization, and less dependence on the spatial location of geologic deposits of reactive material. The main constraint on the dynamic capacity of CO<sub>2</sub> storage via ex situ mineralization is the availability of reactive material near stationary sources of CO<sub>2</sub> on the surface. However, CO<sub>2</sub> reacts with a wide variety of industrial waste materials and can contribute to the remediation of such wastes. The potential for remediation of wastes suggests environmental and economic

benefits of ex situ mineralization in addition to the potential economic, environmental, and social benefits (such as the potential mitigation of CO<sub>2</sub> concentrations in the atmosphere) that are common across all storage options listed in table 2. There are also drawbacks such as visible activity at the surface and environmental effects from the operation of a plant.

Carbon mineralization is receiving increased interest as an option to complement saline CO<sub>2</sub> storage. Interpreting the evidence in table 2 as suggesting that in situ carbon mineralization could be cost-comparable with other storage options is not to argue that any of these CO<sub>2</sub> storage options is economically feasible. Still, the information in table 2 might be helpful to those responsible for determining the economic incentives that could be necessary to encourage greater deployment of the various CO<sub>2</sub> storage options. Specifically, table 2 presents a comparison of the dynamic storage capacity, costs, relative benefits, and potential risks of some CO<sub>2</sub> storage options. Table 2 includes recent estimates of the potential costs of pressure-limited sedimentary reservoir storage, which could be more appropriate for comparison with the costs of in situ CO<sub>2</sub> mineralization than the estimated costs of sedimentary reservoir storage that do not include the costs of pressure management required to mitigate risk. According to the comparisons in table 2, in situ carbon mineralization could be a more cost-competitive CO<sub>2</sub> storage option than has been suggested by other comparisons (such as Olajire, 2013).

## 8. Regional Carbon Dioxide Mineralization Feasibility in the United States

Abundant resources for CO<sub>2</sub> mineralization are present in the United States in the form of in situ mafic and ultramafic rocks, as well as mine wastes derived from them. Whether the use of these resources is technically or economically feasible, however, is a function of multiple variables, including the size of the target rock body or mine tailings pile, composition and reaction rates of relevant minerals, the permeability and porosity of the material, the availability of CO<sub>2</sub>, environmental risks, and mitigation costs. In this section, we take a regional approach to describe the pros and cons of CO<sub>2</sub> mineralization in different regions of the United States and address potential storage sites. Figure 1 is a map of mafic and ultramafic rock resources in the United States that could be potential sinks for CO<sub>2</sub> mineralization. To generate figure 1 and the regional maps referenced in this section, geologic data were compiled and, in some instances, simplified from the following sources: Lambert and others (1988), Ojakangas and others (2001), Reed and others (2005), Sherrod and others (2007), Garrity and Soller (2009), Krevor and others (2009), and Barry and others (2013). The baseline requirement for rock types suitable for CO<sub>2</sub> mineralization is a high concentration of Mg<sup>2+</sup>, Ca<sup>2+</sup>,

## 14 Carbon Dioxide Mineralization Feasibility in the United States

**Table 2.** Summary of potential costs, benefits, and risks for carbon dioxide mineralization and saline storage methods.

[t, metric ton; CO<sub>2</sub>, carbon dioxide; yr, year; km, kilometer; m<sup>3</sup>, cubic meter; CaO, calcium oxide; MgO, magnesium oxide]

Dynamic capacity	Costs (in U.S. dollars)	Benefits and risks
<b>1. Sedimentary reservoir storage assuming no pressure limitations or associated risks</b>		
About 1,070,000 t CO <sub>2</sub> /yr/injection well <sup>1</sup>	• \$7 to \$13/t CO <sub>2</sub> (or less). <sup>1,2</sup>	<ul style="list-style-type: none"> <li>• Lowest apparent cost because estimates do not include management of pressure, water, or risk.</li> <li>• Dynamic capacity is equal to technically feasible capacity.</li> <li>• Storage capacity is widely distributed (reservoirs may exist within 80 km of more than 70 percent of the major stationary CO<sub>2</sub> emitters in the United States [Dahowski and others, 2005]).</li> </ul>
<b>2. Sedimentary reservoir storage with pressure management (either by reducing injection rates or brine extraction)</b>		
About 300,000 to 1,070,000 t CO <sub>2</sub> /yr/injection well <sup>1,3</sup>	• \$20 to \$80/t CO <sub>2</sub> (or more). <sup>3,4</sup>	<ul style="list-style-type: none"> <li>• Lower risk (but higher cost) than non-pressure-limited estimates.</li> <li>• Pressure-limited dynamic capacity without brine extraction could be significantly greater than for mineralization, and with brine extraction it could approach technically feasible storage capacity.<sup>5</sup></li> <li>• Storage capacity is widely distributed (reservoirs may exist within 80 km of more than 70 percent of the major stationary CO<sub>2</sub> emitters in the United States [Dahowski and others, 2005]).</li> </ul>
<b>3.a. In situ CO<sub>2</sub> mineralization in basaltic rocks</b>		
About 14,600 t CO <sub>2</sub> /yr/injection well <sup>6</sup>	<ul style="list-style-type: none"> <li>• \$50 to \$100/t CO<sub>2</sub> (Intergovernmental Panel on Climate Change, 2005, 2007).</li> <li>• \$54/t CO<sub>2</sub> (Olajire, 2013).</li> <li>• \$30/t CO<sub>2</sub><sup>7</sup> (Zalzal, 2017).</li> </ul>	<ul style="list-style-type: none"> <li>• Lower leakage risk than saline storage options and may not require decades-long postinjection monitoring.</li> <li>• Pressure management is necessary to control for the risk of induced seismicity.</li> <li>• Lower dynamic capacity and not as widely distributed (not as close to as many CO<sub>2</sub> sources) as saline storage options.</li> </ul>
<b>3.b. In situ CO<sub>2</sub> mineralization in ultramafic rocks</b>		
Offshore: 1,000 t CO <sub>2</sub> /yr/injection well Onshore: 1 t CO <sub>2</sub> /yr/m <sup>3</sup> in olivine-hosted aquifers <sup>8</sup>	<ul style="list-style-type: none"> <li>• Storage cost data are not available.</li> <li>• As a proxy, a dated Intergovernmental Panel on Climate Change (2005) estimate of \$0.50 to \$8/t CO<sub>2</sub> for injection into deep saline-filled formations is used (for example, Power and others, 2014).</li> </ul>	<ul style="list-style-type: none"> <li>• Injection into ultramafic rocks could pose a significant risk of induced seismicity.</li> <li>• Pressure-management costs are not included in cost estimates.</li> <li>• Dynamic storage capacity in onshore ultramafic rocks could be orders of magnitude less than that in basalts (but data are lacking).</li> </ul>
<b>4. Ex situ CO<sub>2</sub> mineralization</b>		
7,000,000 t CO <sub>2</sub> /yr <sup>7</sup>	<ul style="list-style-type: none"> <li>• \$8/t CO<sub>2</sub> stored in CaO- and MgO-rich industrial wastes or ultramafic mine tailings (Power and others 2014). The estimated cost is the same for CO<sub>2</sub> stored in steel slag.<sup>9</sup></li> <li>• \$54/t CO<sub>2</sub> for mined material already at the surface (Olajire, 2013).</li> <li>• \$100/t CO<sub>2</sub> (or more) associated with mining and processing of feedstock, additional processing at the surface, and the energy required for each processing stage (Power and others, 2013).</li> </ul>	<ul style="list-style-type: none"> <li>• No risky pressure buildup because there are no underground injections.</li> <li>• Economically feasible only at the local scale.</li> <li>• Can provide environmental benefits such as mitigating the effects of alternative treatments, emissions, or disposal of industrial wastes.</li> <li>• Environmental concerns include surface impacts of mining operations.</li> <li>• Costs of storage can be offset by sales of mineral carbonates that are produced because these products will be available at the surface.</li> </ul>



**Table 2.** Summary of potential costs, benefits, and risks for carbon dioxide mineralization and saline storage methods.—Continued

[t, metric ton; CO<sub>2</sub>, carbon dioxide; yr, year; km, kilometer; m<sup>3</sup>, cubic meter; CaO, calcium oxide; MgO, magnesium oxide]

<sup>1</sup>Based on baseline results from the National Energy Technology Laboratory's Saline Storage Cost Model (Morgan and Grant, 2014).

<sup>2</sup>For storage onshore in the United States in 2013 U.S. dollars (Rubin and others, 2015).

<sup>3</sup>Based on preliminary results of simulations of pressure-limited CO<sub>2</sub> storage capacity and costs for the Mount Simon Sandstone (Anderson and Jahediesfanjani, 2017; Jahediesfanjani and others, 2017).

<sup>4</sup>Brine extraction costs based on Harto and Veil (2011), who assumed extraction of brines at a ratio of 1:1 per volume of CO<sub>2</sub> injected.

<sup>5</sup>Gorecki and others (2014, p. 8).

<sup>6</sup>Based on 3 weeks of injection at the Wallula basalt pilot project, where the rate of injection was controlled to maintain pressure within 30 percent of hydrostatic conditions (McGrail and others, 2017).

<sup>7</sup>Based on injection of CO<sub>2</sub> and hydrogen sulfide mixed with water into basalts in Iceland. The injection rate was not pressure limited (Zalzal, 2017).

<sup>8</sup>Data were not available for the maximum rate of injection of CO<sub>2</sub> into ultramafic-hosted aquifers per well. The estimated injection rate per cubic meter of aquifer volume is from Power and others (2013).

<sup>9</sup>Based on the average availability per year of fly ash, cement kiln dust, and iron and steel slag in close proximity to stationary sources of CO<sub>2</sub> emissions in the United States (Kirchofer and others, 2013).

or Fe<sup>2+</sup> cations, which are required for the reactions to form Mg, Ca, or Fe carbonates. In the regional maps, point sources of CO<sub>2</sub> from various industrial sectors are shown within a 100-mile (160-kilometer [km]) buffer of potential mafic and ultramafic reservoirs. The 100-mile buffer is representative of a distance in which long distance pipelines would not be needed to transport CO<sub>2</sub> (Dahowski and others, 2005).

Use of the terms “surface mafic rocks” and “surface ultramafic rocks” comes with the understanding that these rocks actually may be concealed at the ground surface by soil and surficial sediments like alluvium, or by unconsolidated glacial sediments which in some places are hundreds of feet thick.

## 8.1. Pacific Northwest

The Columbia River basin contains 53,700 cubic miles (223,832 cubic kilometers) of reactive flood basalt (Reidel and others, 2002) (fig. 2A) and is estimated to be able to store between 10 and 100 Gt CO<sub>2</sub> (McGrail and others, 2006; Gislason and others, 2010). The kinetics of CO<sub>2</sub> mineralization for basalts are well studied (examples include Gislason and others, 2010; Schaef and others, 2010; Rosenbauer and others, 2012). The results of a recent pilot project involving the injection of 1,000 t of CO<sub>2</sub> in the Columbia River basin indicated that carbonate mineralization was complete in 2 years (McGrail and others, 2017). A similar study of CO<sub>2</sub> injection in Icelandic basalts showed greater than 95 percent mineralization in less than 2 years (Matter and others, 2016). Because of the high porosity and permeability, there is low risk of reaching fracture pressures in the basalt that could lead to induced seismicity and leakage. Compared to other parts of the country, the Pacific Northwest has fewer stationary sources of CO<sub>2</sub> (fig. 2A) (most of which are related to electric power generation and agricultural processing), lower CO<sub>2</sub> emissions,

and less infrastructure for well injection (National Energy Technology Laboratory, 2015).

Large expanses of ultramafic rocks are present in Washington and Oregon, and ultramafic complexes also occur in southwestern Montana. The ultramafic rocks of Washington and Oregon are ophiolite complexes, sections of oceanic plates (crust and uppermost mantle) that have been accreted to and thrust over portions of the western margin of the North American plate through convergent tectonic collisions. Krevor and others (2009) noted that northwest Washington's Ingalls Tectonic Complex and neighboring bodies are dominantly dunite and harzburgite, whereas the Twin Sisters Dunite contains large volumes of unaltered olivine (fig. 2A). The latter would be a good target for mineralization because of its greater potential reactivity and inherent permeability. The Twin Sisters Dunite also hosts an active olivine mine and therefore is a potential source of mining waste for ex situ mineralization. The Josephine Peridotite and Canyon Mountain Complex in Oregon (fig. 2A) are also regional ultramafic bodies that could potentially host mineralization operations, though they are not near any significant point sources of CO<sub>2</sub>.

The Stillwater igneous complex in southwestern Montana (fig. 2A) is a mined, layered intrusive deposit that is the only U.S. producer of platinum-group elements. A peridotite layer in the ultramafic zone of the intrusion is composed mostly of olivine and some pyroxene minerals that are 20–80 percent serpentinized (Page, 1976). The ultramafic zone of the intrusion is only minimally mined, which precludes using tailings or processed materials for ex situ mineral carbonation. The region around the Stillwater platinum-palladium mine in the Stillwater igneous complex has some CO<sub>2</sub> sources, but it may be more economically feasible to capture the emissions from the mine's onsite ore-refining operations.

## 8.2. California

California contains abundant ultramafic rocks for CO<sub>2</sub> storage within multiple ophiolite and pluton assemblages (Krevor and others, 2009). The Trinity peridotite in the eastern Klamath Mountains of northern California (fig. 2B) crops out in approximately half the area of a region roughly 50 km x 75 km (Quick, 1981; Irwin, 2003). Seismic data indicate these rocks extend as far east as Mount Shasta in the subsurface (Fuis and others, 1987). The Trinity peridotite is one of the largest in the United States. Point sources of industrial and agricultural CO<sub>2</sub> are small in this area, and they are mostly limited to the regions around Redding, California, and Klamath Falls, Oregon (fig. 2B).

The Central Valley of California contains numerous industrial, agricultural, and electric power generation point sources of CO<sub>2</sub> emissions. Ultramafic rocks in close proximity surround the entire Central Valley (fig. 2B). These rocks could potentially serve as stable storage reservoirs for the emissions from these industrial sectors. The potential to activate known or unknown faults is a risk that must be taken into account for any CO<sub>2</sub> injection project, especially those in the seismically active regions of the Western United States.

The chrysotile asbestos mines of the Coalinga mining district in California (fig. 2B) operated from 1917 to 2002 and were the last mines in the United States devoted to asbestos production. The remaining mine waste and the surrounding ultramafic complex may be suitable targets for CO<sub>2</sub> mineralization of chrysotile asbestos. The locations of the individual asbestos mines in the district are provided in Van Gosen and Clinkenbeard (2011). The chrysotile asbestos exploited by these mines occurs within the New Idria serpentinite mass (fig. 2B), a serpentinitized peridotite that crops out for approximately 117 square kilometers. The New Idria serpentinite mass contains a large number of serpentine-group minerals (Wise and Moller, 1995).

Large amounts of asbestos-bearing waste rock and serpentinite rock remain in the Coalinga district, and some of the mining areas have been designated as Superfund sites (U.S. Army Corps of Engineers, 2016). Carbonation of the tailings would have the added benefit of mitigating the potential asbestos exposures to recreational users of the Coalinga area. Preventing such exposures has been a challenge for land management and environmental protection agencies (Coleman, 1996; U.S. Environmental Protection Agency, 2008; U.S. Army Corps of Engineers, 2016).

Mafic lavas are present in northeastern California's Modoc Plateau (fig. 2B) and also in southern Oregon. These Miocene- through Quaternary-age volcanics occupy the transition zone between the Cascade Mountains to the west and Basin and Range Province to the east and underlie an area many times larger than the Trinity peridotite (Gay and Aune, 1958). Seismic studies of the plateau indicate layered flood basalts and associated sedimentary deposits are present to a

depth of 3.5 km below sea level (Fuis and others, 1987). Like the nearby Trinity peridotite, there are few major sources of CO<sub>2</sub> in the Modoc Plateau region.

Peridotite-hosted alkaline springs in California (Barnes and O'Neil, 1969) offer the possibility of producing CaOH-rich water to directly capture CO<sub>2</sub> from air to form calcite. This process is an accelerated version of the natural process that forms travertine terraces in peridotites at the surface and carbonates in peridotites at depth (Cipolli and others, 2004).

## 8.3. Hawaii

Basalts in Hawaii, like the Columbia River basalts, represent a volumetrically large potential CO<sub>2</sub> sink. Hawaii has significant volcanic stratigraphy with highly porous vesicular flow boundaries and high permeability within natural fractures. Compared to the potential pressure-management issues associated with low-porosity and -permeability ultramafic rocks, there is less risk of induced seismicity in these mafic basalts. Hawaii's history of major landslides (Moore and others, 1989), however, indicates that extensive, site-specific characterization efforts would be needed to avoid these risks. The naturally high heat flow in Hawaii may increase reaction rates. The kinetics of the reaction would be most similar to those reported by Matter and others (2016) for Iceland, another active volcanic setting with high heat flow, as a low-end estimate, because those proof-of-concept pilot injections were CO<sub>2</sub> limited and not designed to maximize mineralization. Ongoing efforts in Iceland using faster rates of injection may provide more appropriate estimates of the potential mineralization rates in high-heat-flow basalts. Hawaii has low CO<sub>2</sub> emissions, but there are local stationary sources of CO<sub>2</sub>, mostly from electric power generation (fig. 3). The State has less well and pipeline infrastructure to utilize (National Energy Technology Laboratory, 2015) but has considerable basalt resources for mineralization.

## 8.4. Desert Southwest

The Tertiary basalts of the Basin and Range Province in the desert Southwest (fig. 2B) are widespread over a high-heat-flow extensional environment, but they are volumetrically minor compared to the Columbia River and Hawaiian basalts. Most of the rocks are volumetrically small, dispersed extrusive basalts in volcanic fields and likely have subsurface mafic dikes and sills. There is perhaps an increased leakage risk for injection of volatiles into regions with extensive preexisting volcanism and faulting. Well and pipeline infrastructure and stationary sources of CO<sub>2</sub> are sparse here, although they increase toward southeastern New Mexico and western Texas where numerous oil and gas wells and CO<sub>2</sub> infrastructure exist in the Permian basin.

## 8.5. Midcontinent

The midcontinent region and upper Midwest contain abundant subsurface basalts, potentially as much as 20 km thick, within the Mesoproterozoic Midcontinent Rift (Cannon and others, 1989). This region includes surface exposures in Wisconsin, Michigan, and Minnesota, including the Duluth Complex, and subsurface plutonic basalts farther south in Wisconsin and Minnesota as well as in Michigan, Iowa, Nebraska, and Kansas (fig. 4). The subsurface Glen Mountains Layered Complex of early Paleozoic age is found even farther south in Oklahoma and Texas (Lambert and others, 1988). Not only are there large volumes of subsurface basalts in the midcontinent region, but they also fall in an area with abundant stationary sources of CO<sub>2</sub> (fig. 4). The highest-CO<sub>2</sub>-output stationary sources include electric powerplants in northern Minnesota and along the Nebraska-Iowa border. Other sites include industrial sources of CO<sub>2</sub> near the Duluth Complex of northern Minnesota and in Michigan and ethanol plants in Minnesota, Iowa, and Nebraska. The subsurface basalts in Oklahoma and Texas are much closer to regions of existing well infrastructure for petroleum and natural gas production. Unlike the highly porous, near-surface Hawaiian and Columbia River basalts, these deep basalts are likely to be significantly less porous and to have unknown amounts of previous carbonation. Induced seismicity from wastewater injection is a known hazard in this region (for example, see Ellsworth, 2013), particularly for fluids injected into basement rocks; therefore, injection pressures would need to be regulated. Because of the economic cost of accessing rocks at great depth, the significant volumes of mafic rocks below the shallow subsurface may not be viable storage options unless the economics of storage change significantly.

## 8.6. Eastern States

The Eastern States contain both basalt and ultramafic resources for CO<sub>2</sub> storage. The Mesozoic rift basins in the Eastern United States are a source of igneous and metaigneous rift basalts that exist both in the subsurface and as surface exposures. These basalts are part of the CAMP, a large continental flood-basalt province that formed during rifting of the supercontinent Pangea (Goldberg and others, 2010). Like the subsurface Midcontinent Rift basalts, eastern Mesozoic rift basalts may be partially metamorphosed and may have low porosity and permeability. Point sources of CO<sub>2</sub> are abundant here, particularly electric powerplants, cement plants, and other industrial sites. There is well infrastructure in the nearby Appalachian basin but little to none within the CAMP.

The Eastern States also have resources of ultramafic rocks in sparse outcrops from Georgia to Maine (fig. 1). These include the dunites of North Carolina, as well as peridotite and metamorphic serpentinites. The asbestos mines shown in figure 5 are derived from these ultramafic serpentinite outcrops. Dunites, peridotites, and serpentinites, though

ultramafic and the most CO<sub>2</sub>-reactive of common natural materials, generally have very low porosity and permeability and therefore a small surface area of reaction. They also have highly variable mineralogies and degrees of natural carbonation, meaning that the amount of CO<sub>2</sub> that can be stored is highly variable and dependent on the specific subtypes of ultramafic rock.

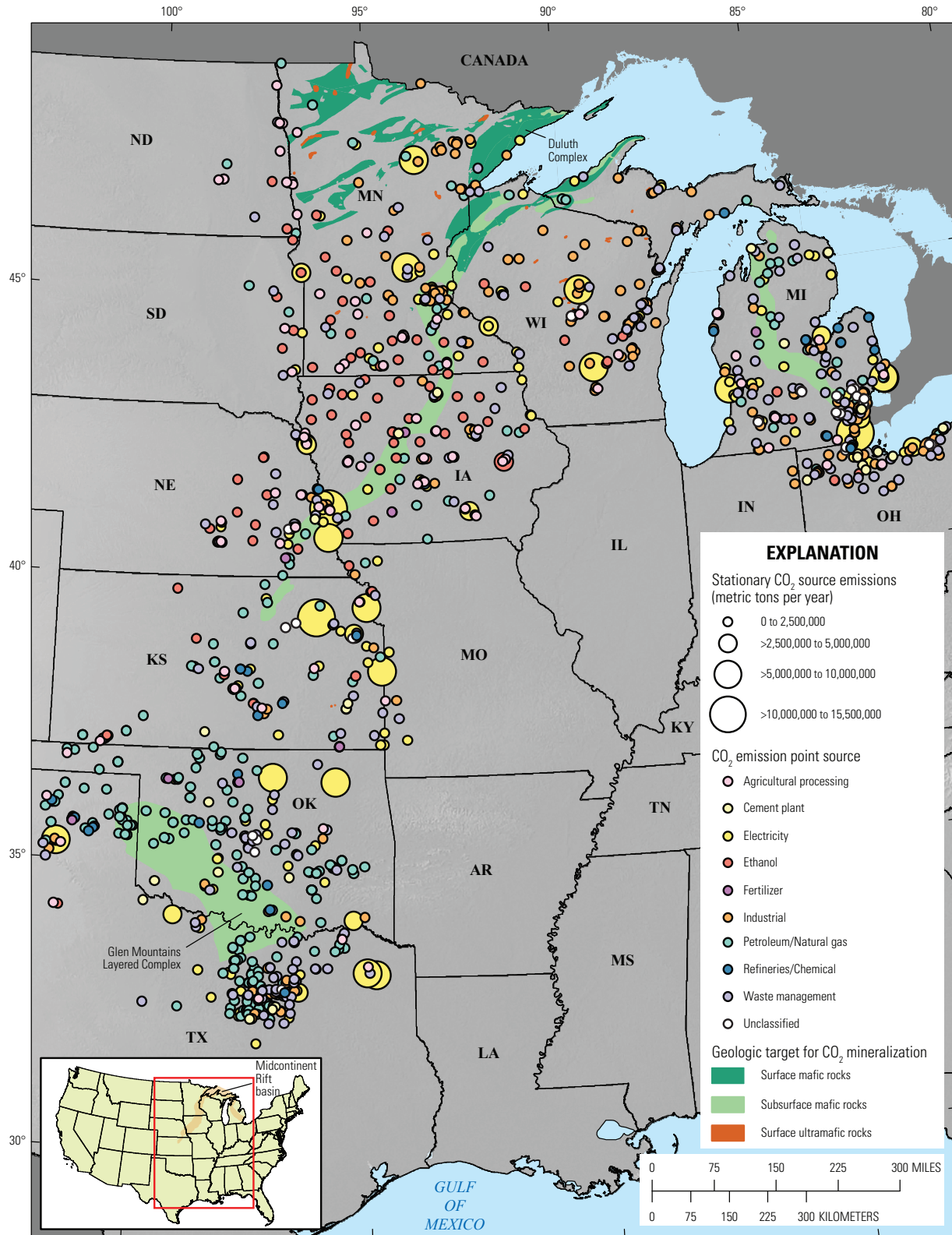
The Belvidere mine in northern Vermont is an example of a potential site for carbonate mineralization of asbestos with CO<sub>2</sub>. Until its closing in 1993, the Belvidere mine was a site of mining of veins of chrysotile asbestos developed within metasomatized dunite and serpentinite (Cady and others, 1963; Chidester and others, 1978; Van Baalen and others, 1999). The ultramafic body that forms Belvidere Mountain and hosts the chrysotile deposits is approximately 3,400 m (11,000 feet [ft]) long, 1,800 m (6,000 ft) wide, and as much as 450 m (1,500 ft) thick (Chidester and others, 1978). Large piles of waste rock of metasomatized dunite and serpentinite, some containing remnants of chrysotile, remain at the Belvidere mine site and the nearby processing plant (Krevor and others, 2009).

## 9. Conclusions

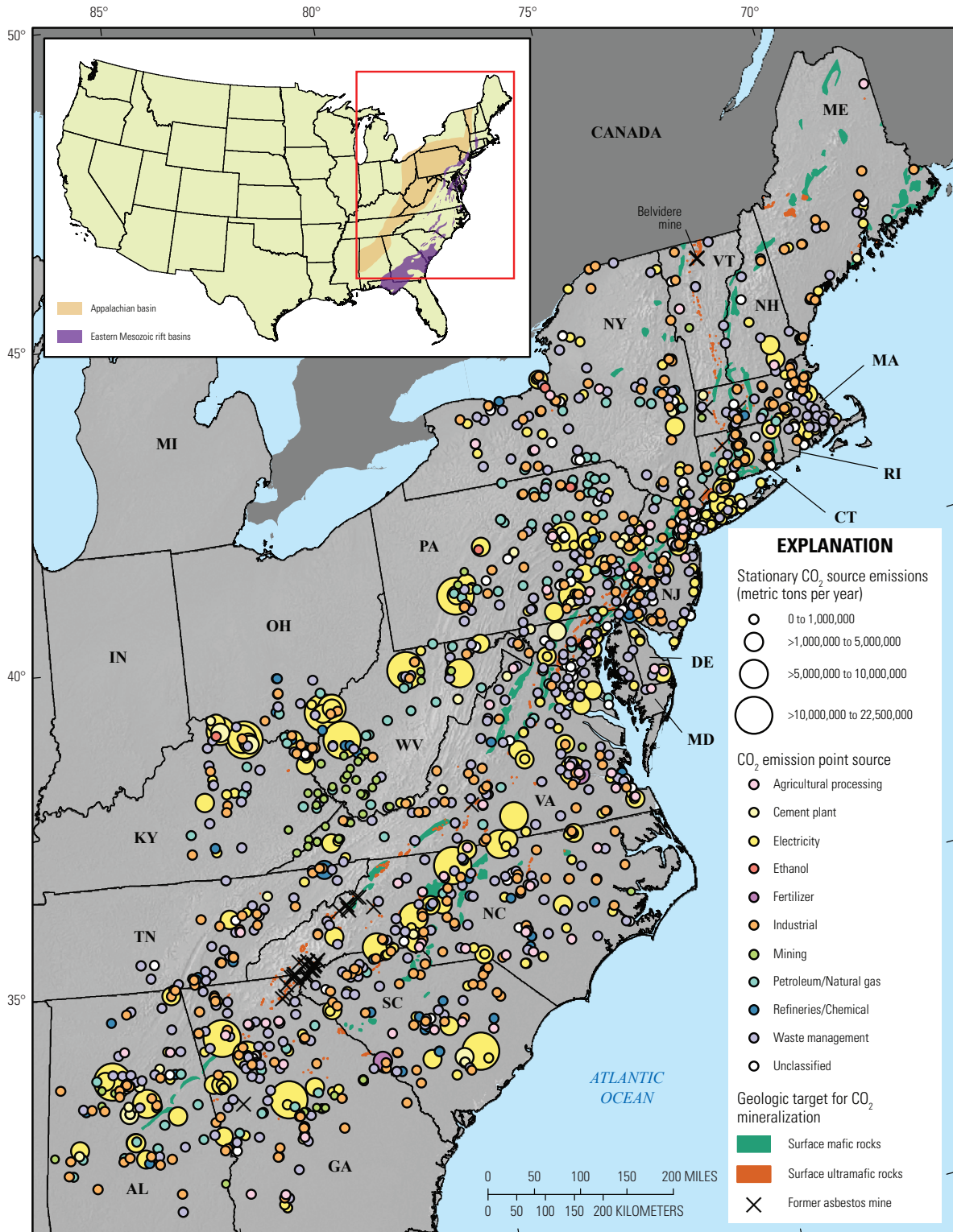
CO<sub>2</sub> mineralization has the potential to provide a stable storage mechanism for anthropogenic or atmospheric CO<sub>2</sub>. Mineral carbonation requires sources of Ca<sup>2+</sup>, Mg<sup>2+</sup>, and (or) Fe<sup>2+</sup> cations, such as mafic and ultramafic rocks, to react with the CO<sub>2</sub>. The United States contains abundant resources of mafic basalts and ultramafic rocks that could be used for mineral carbonation to capture and store CO<sub>2</sub> through subsurface injection or surface reactions with industrial byproducts. Particularly suitable targets include asbestos or other ultramafic mine tailings, in situ ultramafic rocks on the East and West Coasts, the Columbia River Basalts in the Pacific Northwest, and the Midcontinent Rift zone basalts in the midcontinent because of their proximity to major sources of CO<sub>2</sub>. Though not a major emitter of CO<sub>2</sub>, Hawaii is endowed with significant volumes of potential reservoir rocks that could be utilized to mitigate local CO<sub>2</sub> emissions.

CO<sub>2</sub> storage by mineralization is achieved by the formation of stable, solid carbonate minerals that capture and lock up the CO<sub>2</sub> in mineral form and thus do not pose the leakage risks seen with fluid injection methods. Additionally, the burden of long-term monitoring of a fluid plume of CO<sub>2</sub> in the subsurface is significantly reduced. However, there are risks and potential environmental effects associated with CO<sub>2</sub> mineralization technologies. Injection of CO<sub>2</sub> into subsurface environments carries the risk of induced seismicity just as with any subsurface injection activity. Depending on the mineralogy and conditions, it is possible that there could be unwanted byproducts of the reaction. There also may be increases in noise and traffic in these settings and potentially large water requirements.

Cost, risk, and the availability of suitable rock are the main restrictions to the adoption of mineral carbonation



**Figure 4.** Map of stationary point sources of carbon dioxide (CO<sub>2</sub>) emissions within 100 miles (160 kilometers) of mafic and ultramafic rocks in the midcontinent United States. Geologic data were compiled and, in some instances, simplified from Lambert and others (1988), Ojakangas and others (2001), Reed and others (2005), Garity and Soller (2009), and Krevor and others (2009). Outlines of ultramafic-rock polygons were used to increase visibility. CO<sub>2</sub> point-source data are from National Energy Technology Laboratory Carbon Storage Atlas (2015). Base map was compiled from data from Environmental Systems Research Institute, Inc. (2007a,b) and Jarvis and others (2008).



**Figure 5.** Map of stationary point sources of carbon dioxide (CO<sub>2</sub>) emissions within 100 miles (160 kilometers) of mafic and ultramafic rocks in the Eastern United States. Geologic data were compiled and, in some instances, simplified from Reed and others (2005), Garrity and Soller (2009), and Krevor and others, (2009). Outlines of ultramafic-rock polygons were used to increase visibility. Asbestos mines were compiled from past producing mines described in Van Gosen (2006a,b). CO<sub>2</sub> point-source data are from National Energy Technology Laboratory Carbon Storage Atlas (2015). Base map was compiled from data from Environmental Systems Research Institute, Inc. (2007a,b) and Jarvis and others (2008).

processes, and these issues are active areas of research. The costs and benefits of CO<sub>2</sub> mineralization are compared here to CO<sub>2</sub> storage in saline reservoirs using estimates of pressure-limited dynamic storage capacity. Investigations based on kinetic experiments, as well as demonstration pilot projects, indicate that reaction rates for basalts and ultramafic rocks are much faster than initial projections suggested. Increased technological developments combined with larger pilot projects may help to decrease costs and establish carbon mineralization as a cost-effective CO<sub>2</sub> sequestration method in the future.

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# Glossary

**asbestos** A fibrous serpentine or amphibole silicate mineral, common in some ultramafic rocks.

**aqueous CO<sub>2</sub>** Carbon dioxide dissolved in water.

**basalt** A fine-grained, extrusive igneous rock with a silica (SiO<sub>2</sub>) content of 45 to 55 weight percent.

**brine** Natural high salinity water found in the pore space of sedimentary basin reservoirs

**carbonate** A class of minerals with the CO<sub>3</sub><sup>2-</sup> anion.

**dunite** An ultramafic rock with an olivine content greater than 90 weight percent.

**ex situ mineralization** Storage processes that occur in rocks removed from their original position.

**felsic** An adjective used to describe rocks with a SiO<sub>2</sub> content greater than 69 weight percent.

**hyaloclastites** A brecciated and hydrated volcanic rock composed of glass.

**in situ mineralization** Storage processes that occur in rocks in their original position.

**listvenite** A completely carbonated ultramafic rock consisting of carbonate and quartz.

**mafic** An adjective used to describe rocks with a total MgO, FeO, MnO, and Fe<sub>2</sub>O<sub>3</sub> content of 15 to 28 weight percent and SiO<sub>2</sub> content of 46 to 54 weight percent.

**mineral assemblage** A collection of minerals that define a rock.

**olivine** A magnesium iron silicate mineral with the formula (Mg,Fe)<sub>2</sub>SiO<sub>4</sub>.

**ophiolite** A portion of the oceanic crust revealed at the surface due to tectonic thrusting.

**peridotite** An ultramafic, coarse-grained igneous rock with an olivine content greater than 40 weight percent.

**quartz** A silica oxide mineral with the formula SiO<sub>2</sub>.

**retrograde metamorphism** A form of rock metamorphism that occurs as pressure and temperature decrease.

**serpentine** A group of magnesium phyllosilicate minerals with the formula Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub> including those of the asbestiform variety.

**serpentinite** A low-temperature metamorphic rock containing serpentine minerals produced from the alteration of ultramafic rocks.

**supercritical CO<sub>2</sub>** Carbon dioxide in a fluid state where it is held at or above its critical temperature and critical pressure, imparting both gas and fluid characteristics.

**ultramafic** An adjective used to describe rocks with a total MgO, FeO, MnO, and Fe<sub>2</sub>O<sub>3</sub> content of 35 to 46 weight percent and SiO<sub>2</sub> content of 42 to 48 weight percent.





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