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# 3-D Geological Mapping of Rocks Potentially Suitable for Capturing Carbon Dioxide from the Atmosphere

**Agterberg FP***

Geological Survey of Canada, Canada

***Corresponding author:** Agterberg FP, Geological Survey of Canada, 601 Booth Street, Ottawa, Ontario K1A 0E8, Canada, Tel: +1 6138439358; Email: frits@rogers.com

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## Abstract

Rapid increase of the Anthropocene ambient air $CO_2$ concentration value is the main cause of the 1.2 °C global temperature rise creating the recent adverse climate and weather changes. Promising new methods to capture $CO_2$ from the air are being developed but remain too expensive for worldwide applications. Mantle-derived rocks, primarily basalts, peridotites and serpentinites are likely to play an important role in future $CO_2$ reduction because of relatively rapid disintegration of minerals (including olivine and serpentine) in these rocks potentially resulting in widespread $CO_2$ capture. Examples to be discussed include artificially enhanced carbonization of water emitted from ophiolites, and acid dissolution of serpentinites resulting in indirect mineral carbonation by optimizing temperature and pressure conditions. Reconstruction of a large cone-shaped body of serpentinite situated within the Mount Albert peridotite intrusion in Québec is presented as an example of the rôle 3-D geologic mapping can play in future $CO_2$ reduction efforts.

**Keywords:** Global Value Chain; Carbon Reduction; Degree of Coupling Coordination

## Introduction

During most of the past 800,000 years, global atmospheric $CO_2$ concentration value was fluctuating between about 200 and 300 ppm. However, at the beginning of the Anthropocene in about 1850, it began its increasingly rapid increase to approximately 422 ppm. This increase is the main reason that global surface temperature became enlarged by about 1.2 °C. It is well-known that $CO_2$ in air and mean global temperature are linked because the Earth's heat radiation is intercepted by "greenhouse gases", primarily $CO_2$ (but also methane and nitrous oxide). As documented in the latest (2022) Assessment Report of the International Panel on Climate Change, the average global temperature increase has resulted in many adverse climate conditions. Weather is the short-term manifestation of climate. Multifractal theory offers new insights into the relationship between weather and climate [1]. Currently, various efforts are being made to reduce further annual increases in the mean ambient air $CO_2$ concentration value. A one-page summary of some material covered in this paper has been published as a Viewpoint [2].

Global atmospheric $CO_2$ concentration value has fluctuated significantly during the Phanerozoic that commenced 539 million years ago. It was relatively low (< 500 ppm) during the Earth's two periods of widespread continental glaciation and relatively high (>1000 ppm) during warmer periods. Esmeray-Senlet [3] has identified three major Phanerozoic climate-related mass extinctions: Late Ordovician (445 Ma), Late Devonian (372 Ma) and end-Triassic (201 Ma) (ages from [4]; also see [5]). These three events were accompanied by significant atmospheric

$CO_2$ increases. According to Royer [6], global atmospheric $CO_2$ concentration value reached about 3000 ppm at the Triassic-Jurassic boundary. Recently, Song [7] has published a seawater temperature variation curve for the Phanerozoic accompanied by a post-Silurian $CO_2$ variation curve. Rapid $CO_2$ emissions resulting from basaltic volcanism may have been the cause of other worldwide Phanerozoic discontinuities as well. Such events can reoccur in future. Atmospheric oxygenation history analysis may be a new technique to help predict sudden $CO_2$ emissions of this type (cf.) [8].

With $CO_2$ concentration values more than a hundred times greater at the Triassic-Jurassic and Silurian-Devonian boundary ages, global surface temperatures probably also greatly exceeded its currently low Anthropocene value of about 14$^o$ Celsius. In the geologic past, higher temperatures on Earth were primarily reduced by absorption of $CO_2$ from the air by vegetation. The rapid Anthropocene $CO_2$ increase of about 120 ppm is unusual because it has taken place over such a short period of geologic time.

Large-scale ambient air $CO_2$ concentration reduction technologies would involve expensive major changes in the management of natural resources in the upper parts of the Earth's crust. Nevertheless, there already exist several ways in which the current average global atmospheric $CO_2$ concentration value of 422 ppm. could be reduced. An example is Lackner's [9] method to capture $CO_2$ from ambient air. The $CO_2$ collection rate described by this author significantly exceeds those of trees and other photosynthesizing organisms that in the geologic past were primarily responsible for decreasing the air's $CO_2$ content. Another promising technique is to capture $CO_2$ in water and injecting it under high pressure into basaltic rocks. At the Hellisheiði Carbix site in Iceland this process results in the formation of stable, $CO_2$-free, carbonate minerals (mainly calcite) in less than 2 years [10]. The calcite is dissolved in water to form bicarbonate ions ($HCO_3^-$) that subsequently are incorporated into carbonate minerals for shells and skeletons in the ocean (Encyclopedia Britannica, 2020).

Other methods to reduce atmospheric $CO_2$ by capturing it in the form of carbonates (primarily dolomite) are also becoming increasingly effective. Chemical engineers are actively working to improve existing methods for this purpose, especially by carbonization of rocks rich in olivine or in serpentine derived from olivine. Geoscientists can help by 3-dimensional mapping of the upper part of the Earth's crust, outlining the shapes and compositions of rock units suitable for the capture and retainment of ambient $CO_2$. Potential widespread usage of peridotites and serpentinites for this purpose will be discussed later in this paper. First, new theory on the relationship between weather and climate will be discussed in some detail.

## Weather and Climate

Weather is a short-term random manifestation of long-term climate. During the past 10 years significant progress has been made in understanding the relationship between weather and climate. Lovejoy and Schertzer [1] use the following approach. In their 3-parameter α-model, α represents the Lévy index that, together with the "codimension" $C_1$ and the "deviation from conservation" $H$, characterizes a universal multifractal field $\rho_\lambda$ that can be a flux or density characterized by its probability distribution

$P(\rho_\lambda \lambda^\gamma \propto \lambda^{-C_1(\gamma)}$ with statistical moments $E\left(\rho_\lambda^q\right) \propto \lambda^{K(q)}$;

$\lambda = L/\epsilon$ is the so-called scale ratio with $L$ representing the largest scale that can be set equal to 1 without loss of generality as shown by Cheng and Agterberg [11]. The relations between $K(q)$, $C_1(\gamma)$ and the field order $\gamma$ are:

$$K\left(q\right) = \max_{\gamma}\left\{q\cdot\gamma - C_1\left(\gamma\right)\right\}; \quad C_1\left(\gamma\right) = \max_{q}\left\{q\cdot\gamma - K\left(q\right)\right\}$$

Detailed explanations of these concepts are provided elsewhere [1]. Time series for temperature and other variables associated with weather and climate have characteristic features involving chaotic events. The approach taken here is an example of non-linear modeling of extreme events in the geosciences as defined by Cheng [12, 13]; also see Agterberg [14], and advocated in geophysics by Lovejoy et al. [15].

Weather, which typically has a duration of about one week only and is clearly full of chaotic events, is followed by a relatively long period with relatively few chaotic events that can last as long as 50 years depending on geographic location. After this quiet period called "macroweather" by Lovejoy and Schertzer [1], the "climate" sees a renewal of the occurrence of reactively many chaotic events. Weather and climate have positive values of $H$, whereas $H$ is negative for the interval between weather and climate. The parameter $H$ is named after Hurst [16], an early climatologist who discovered that average number of consecutive years $m(t)$ of plentiful water in the river Nile reaching Egypt is not constant as had been assumed previously but proportional to $t^H$, where $t$ is time and $H > 1$ instead of $H = 1$. Before $H$ was termed the multifractal "deviation from conservation" by Lovejoy and Schertzer and can become negative, it was called "fractal dimension" with the property $H > 0$ by Mandelbrot [17].

As mentioned before, Lovejoy and Schertzer [1] have shown that what is commonly called "climate" covers two separate time-domains. The first of these two domains is much less chaotic than both the short-term weather and the long-term climate. One of their examples is a Greenland dataset of 17,551 points of GRIP (summit) ice-core $\delta^{18}(O)$ (a temperature proxy) location data spanning the period from the present to 91,000 years ago. These authors prefer to use the power spectrum for statistical analysis with $Log_{10}$ E($\omega$) plotted against $Log_{10}\,\omega$ in the frequency domain where E denotes mathematical expectation and $\omega$ is a measure of frequency, A major advantage of this approach is that regimes of events that take place at the same time occupy separate, consecutive domains along the frequency axis. The resulting log-log spectrum for the GRIP example as shown in Lovejoy and Schertzer [1] Fig. 1.9c consists of approximately three consecutive straight-line segments with slopes $\beta_w \approx 2$, $\beta_{mw} \approx 0.2$, and $\beta_c \approx 1.4$, where the subscripts *w*, *mw* and *c* denote weather, macroweather and climate, respectively. The $Log_{10}\,\omega$ values separating these domains are 1.5 and slightly less than 2. It is noted that the macroweather slope ($\beta_{mw} \approx 0.2$) in the power spectrum for the GRIP example is close to zero. A horizontal macroweather line would correspond to a signal-plus-noise model [18] for variations of the original measurements along the ice core. The signal would then be according to a smooth, continuous curve without the possibility of discontinuous, chaotic interruptions as can occur when $\beta > 0$.

Each slope parameter satisfies the equation $\beta$=1+2$H$-$K$(2) that is mainly determined by the value of $H$, which is positive during weather and climate but negative during the macroweather period. For a simple explanation of positive versus negative value of $H$ (see Fig. 2.10 in [17]). Macroweather value of $H$ depends strongly on geographic location. It is close to zero in the tropics and close to -0.5 in the Arctic, where macroweather variability would be according to a signal-plus-noise model without the possibility of catastrophic events (cf., Fig. 5.5) [19]. Statistical methods to estimate $H$ include the one developed by Ramanathan [20] for rainfall scenarios in parts of France. Clearly, the preceding 3-stage approach to weather and climate differs from the commonly used 2-stage approach. The likelihood of re-occurrence of major chaotic events after the relative calm post-weather period from about 10 to 50 years that is generally believed to constitute "climate" deserves further study. Post-macroweather climate would signify the re-emergence of new types of chaotic events associated with the Late Anthropocene increases in atmospheric $CO_2$ concentration value and global temperature. The recent excessive rain fall event in Pakistan (September, 2022) could be an example of a chaotic event of this type, although it might also constitute a chaotic weather or macroweather event amplified by the Anthropocene rise in global surface temperature of 1.2$^o$.

## Olivine in Peridotites and Basalts

Peridotites are mantle-derived rocks primarily consisting of olivine. During the 1960s the International Upper Mantle Project (IUMP) existed as a multinational scientific program directed toward research on ultramafic and mafic rock formation processes influencing the development of the Earth's crust. Olivine is a silicate mineral with chemical equation (Mg, Fe)$_2$ $SiO_4$ that is readily affected by weathering mainly because of absorption of $CO_2$ and water. The ratio of magnesium to iron in olivine varies between its two endmembers which are forsterite (Fo with Mg only) and fayalite (Fa with Fe only). The molar percentage of forsterite exceeds that of fayalite. For example, in the Mount Albert peridotite intrusion to be discussed in more detail in a later section, Fo-content of olivine fluctuates around 90% [21]. The olivine usually co-exists with other minerals, primarily orthopyroxene, that are also subject to relatively rapid weathering. It is noted that olivine can contain traces of other metals including Mn, Ni, and Ca. The chemical formula for dissolution of forsterite is:

$$Mg_2SiO_4 + 4\cdot CO_2 + 4\cdot H_2O \rightarrow 2\cdot Mg + 4\cdot HCO_3^- + H_4SiO_4$$

where $HCO_3^-$ represents water with dissolved bicarbonate ions. Fayalite can capture $CO_2$ by means of the "serpentinization reaction" [22]:

$$3\cdot Fe_2SiO_4 + 2\cdot H_2O \rightarrow 2\cdot Fe_3O_4 + 3\cdot SiO_2 + 3\cdot H_2$$

where $H_2$ is hydrogen gas. An example of forsterite weathering is given by Schuiling [23]. The Eifel Mountains in Germany along the river Rhine consist mainly of basalts. Sediments deposited at the bottom of the Rhine here contain olivine and augite phenocrysts as well as pieces of slate, but 150 km downriver all olivine has disappeared from the river-bottom sediments, whereas the augite and pieces of slate are preserved. The Rhine carries the $HCO_3^-$ ions produced by weathering of the olivine phenocrysts to the North Sea where they are later biomineralized to carbonite in the shells of organisms. Schuiling [23] as well as other authors have proposed various methods of enhanced weathering; e.g., by spreading crushed olivine and silicate minerals with similar properties on beaches or other land surfaces in order to capture and immobilize significant amounts of $CO_2$ from the air.

The Samail Ophiolite in Oman is the world's largest ophiolite outcrop. As described by Sanna, et al. [24], mineralized springs on it emit water enriched in carbonates, thus capturing $CO_2$ from the air by a slow natural process of the order of 26,000 years. By artificially enhancing the carbonization, processing time could be reduced a million times according to these authors, making it equivalent to about 10 days only. This kind of weathering process includes a change of serpentine into "pseudo-forsterite":

$$Mg_3Si_2O_5(OH)_4 \rightarrow MgO_3\,(SiO_2)_2 + 2\cdot H_2O$$

requiring heating above 630$^o$ [24, Eq. 17].

### Serpentinites

According to Debret [25], serpentinites constitute one of the major components of the oceanographic lithosphere and are stable in the slab and the Mantle wedge up to 100-150 km depth in subduction zones [26]. Oyanagi [27] have described serpentinization of forsterite to form serpentine: $Mg_3Si_2O_5(OH)_4$ and brucite: $Mg\,(OH)_2$ as follows:

$$2\cdot Mg_2SiO_4 + 3\cdot H_2O \rightarrow Mg_3Si_2O_5(OH)_4 + Mg\,(OH)_2$$

These authors have also conducted artificial serpentinization experiments at temperatures between 260 °C and 300 °C with $SiO_2$ in solution, for which:

$$3\cdot Mg_2SiO_4 + SiO_2 + 4\cdot H_2O \rightarrow 2\cdot Mg_3Si_2O_5(OH)_4$$

In both preceding cases, subsequent serpentine carbonation is achieved by the reaction:

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 \rightarrow 3\cdot MgCO_3 + 2SiO_2 + 2\cdot H_2O$$

where $MgCO_3$ is dolomite and $SiO_2$ is quartz Although this reaction has been known to exist for a long time [28], recent experiments on tailings and waste rock [29] indicate that acid dissolution of serpentinites resulting in indirect mineral carbonation can be achieved much more rapidly by optimizing temperature and pressure conditions. It is also noted that there exists a method to capture $CO_2$ from brucite [30]:

$$3\cdot Mg\,(OH)_2 ++ 3CO_2 \rightarrow 3\cdot MgCO_3 + 3\cdot H_2O$$

The new developments briefly described in this section indicate that serpentine carbonation is an active current research subject in which progress is being made to significantly reduce the currently abnormally high $CO_2$ content of ambient air.

### The Mount Albert Peridotite Intrusion

The Canadian IUMP contribution included detailed study of the Mount Albert peridotite intrusion that is located on the Gaspé Peninsula in Québec. After its intrusion about 530 my ago, much of the peridotite was changed into serpentinite because of subsequent absorption of water. The Mount Albert intrusion that measures approximately 44 $km^2$at the surface was mapped and sampled systematically by scientists of the Geological Survey of Canada in 1959. Prior to serpentinization, this intrusion consisted mostly (from 80% to 90%) of olivine, the remainder being primarily orthopyroxene with some chrome spinel (up to 1%) and diopsidic clinopyroxene. It was attempted to systematically collect rock specimens on the topographic surface at the nodes of a rectangular grid with 1000 feet (approx. 30.5 m) N-S and E-W spacings.

Polynomial trend surface analysis was performed on compositional data for olivine and orthopyroxene in these rock specimens [31]. The percentage forsterite in these specimens varies from 88%. to 92%. Orthopyroxene has chemical equation $(Mg,\ Fe)_2\ Si_2O_6$. Its two endmembers are enstatite ($Mg_2\ Si_2O_6$) and ferrosilite ($Fe_2\ Si_2O_6$). Other metals (especially Ca and Al) may be may be present in trace amounts. Percentage enstatite in the specimens varies from 86% to 93%. Variability of block average concentration values depends on block size. Sample variance is relatively large for the original specimens. From a detailed comparison of forsterite content of olivine with enstatite content of orthopyroxene [31], it can be inferred that for sufficiently large blocks of rock (potentially suitable for mining) the percentage forsterite in olivine varies from about 90% to 91% and the percentage of enstatite in orthopyroxene from 85% to 93% across the topographic surface of the Mount Albert peridotite intrusion. If $CO_2$ sequestration from peridotites were to become economically feasible in future, detailed block average mineral concentration value studies would provide important inputs.

The relationship between percent serpentine and rock density in the rock samples taken from the Mount Albert intrusion is approximately linear. In total, 359 data points were available for specific gravity measurements ranging from 2.5 (nearly pure serpentine) to 3.3 (unaltered peridotite). In most of the study area, average specific gravity is about 2.7 indicating that, about 75% of the original peridotite at the current topographic surface was changed into serpentinite. Because of relatively strong differences in topographic relief, it was possible to perform 3-dimensional polynomial trend analysis [32] allowing the construction of 2-D maps at 2000 and 3000 ft elevations showing that, geometrically, serpentinization of the peridotite body occurred along a northward dipping inverted pyramid. To-day's locations of two rivers in the Mount Albert Provincial Park above the serpentinized peridotite reflect past differences in degree of serpentinization of bedrock eroded away relatively recently. Through time, the axis of maximum serpentinization moved northward as well as upward. More detailed studies of this type would amplify the worldwide availability of 3-D serpentinite bodies possibly suitable for the permanent capture and storage of atmospheric $CO_2$ in the form of carbonates.

### Potential Storage of $CO_2$ at Depth in Sedimentary Basins

Independent of the methods described in previous sections, progress is being made to develop methods of storing $CO_2$ in sedimentary basins including investigations of the $CO_2$ storage capacity of Miocene and Pliocene strata in the Utsira Formation [33]. These results showed that $CO_2$ can be stored here in dense phase up to a depth of approximately 500 m below mean sea level, significantly shallower than assumed previously. Storage capacity was studied using static volume estimates and by computer simulated injection with up to 210 wells across the entire 25,000 $km^2$ occupied by the Utsira Formation leading to the conclusion that cost effective utilization of the reservoir could be between 20 to 60 gigatons [33]. The objective of the Utsira project was to develop new methods to effectively help reduce the annual increase in atmospheric $CO_2$ concentration value. If methods were to be developed to reduce the current $CO_2$ concentration value of approximately 422 ppm itself, sedimentary basins could provide another way to store $CO_2$ captured from the air.

In previous sections of this paper the emphasis was on the potential use of mantle-derived rocks for the potential storage of $CO_2$. Chemical reactions that were listed included extraction of $CO_2$ from the air by combining it with forsterite and fayalite. It should be kept in mind, however, that olivine crystals within peridotites contain both forsterite and fayalite in variable, mutually intertwined, proportions It should also be kept in mind that there exist at least two natural processes (dissolution of Eifel olivine phenocrysts, and disintegration of olivine crystals in the Samail Ophiolite). With respect to both ophiolites and serpentinites, it was pointed out that temperature and pressures are critical factors to speed up the chemical reactions required to capture $CO_2$ from the air. The Utsira example briefly described earlier in this section illustrates that important factors to be considered for $CO_2$ capture are mechanical engineering and transportation costs. For example, if crushed olivine crystals were to be used for $CO_2$ capture by spreading them on beaches or other land surfaces, the costs of mining peridotite bodies, transportation and rock crushing might be insurmountably high.

## Concluding Remarks

During the Anthropocene there has been rapid increase in global surface temperature by about 1.2 °C, primarily due to increase of the atmospheric $CO_2$ concentration value. This process has already resulted in numerous undesirable climate and weather changes. Multifractal weather-climate modeling indicates that short-term weather is followed by "macroweather" representing a 10-50 year long period of relative stability with fewer chaotic events before long-term climate sets in with re-occurrences of major chaotic events. These three periods take place simultaneously anywhere on Earth. Currently, geoscientists and chemical engineers are developing promising, increasingly less expensive methods of atmospheric $CO_2$ concentration value reduction that would reduce the occurrence of both short-term and long-term chaotic events. These important contemporaneous efforts will probably be intensified as weather and climate continue to worsen. In the 19$^{th}$ century geology's subscience of stratigraphy and the invention of geologic maps helped to find coal and, subsequently, hydrocarbon deposits. Geoscientists may now intensify efforts to identify and describe rock units potentially favorable for carbonization by 3-D mapping and compositional data analysis of Mantle-derived rocks.

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