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2008-U-R0776/B Desk study on the feasibility of CO_2 sequestration by mineral carbonation of olivine

Subsurface and Groundwatersystems Princetonlaan 6 P.O. Box 80015 3508 TA Utrecht The Netherlands

www.tno.nl

T +31 30 256 47 50 F +31 30 256 48 55 info-BenO@tno.nl

Date	July 2008
Author(s)	H. Veld G.D. Roskam R. van Enk
Assignor	VROM
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Approved by	L.A. van de Kuil

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Executive summary

Since fossil fuels are and will be the main source of energy production for at least several more decades, it is imperative to reduce CO_2 emissions in order to stabilise atmospheric CO_2 levels. Several technological applications of mineral carbonation are under investigation or in use already, but limited information is available on the possibility to enhance mineral carbonation under natural conditions. This study contains an analysis of factors that influence the weathering rate of olivine, a magnesium iron silicate that can sequester CO_2 by conversion to bicarbonate and eventually magnesium and calcium carbonate. Olivine is one of the most common minerals in the world and occurrences are present on all continents.

Two different scenarios, the spreading of olivine on the coastline and the application of olivine as a replacement of lime in agriculture, have been evaluated based on a model developed by Olsen (2007). A small grain size, low pH value, high temperature, and the presence of organic acids are favourable for the dissolution of olivine. Under tropical conditions 80% of the olivine will dissolve in 6 years when applied in an agricultural setting and in 28 years when spread along the coastline.

One factor that can not be accurately quantified is the difference between laboratory and field data: the model indicates that the dissolution of olivine occurs 100 to 1000 times slower in the field as compared to the rates that are determined in the lab. However, several of the factors that contribute to the difference between laboratory and field (reactivity of the surface, contact of the surface with solution, local saturation of the solution) are of limited or no relevance to these scenarios. Overall we may have to increase the time estimate for an agricultural setting by one or two orders of magnitude, but the reaction rate of olivine in seawater could actually be close to laboratory conditions, making spreading of olivine along the coastline under tropical conditions a viable option for CO_2 sequestration.

If the EU (counting 15 member states at the time) would like to meet its requirements under the Kyoto protocol by sequestration of CO_2 with olivine (at 1.25 tonnes of CO_2 per tonne of olivine), the worldwide production of olivine would need to increase by a factor 55. However, many large olivine occurrences are not yet in production, and several of those are located in tropical regions, so the expenses for transportation would be limited if the olivine was spread in the vicinity of the mine. Currently the cost of olivine (mined in Greenland) ranges from 15 to 75 Euro per tonne, which means that the price per tonne of CO_2 avoided is between 12 and 60 Euro.

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1 Introduction

Since fossil fuels will remain the main source of energy production for the foreseeable future, it is crucial to reduce CO_2 emissions in order to stabilise atmospheric CO_2 levels. Various CO_2 sequestration and recycling studies are currently conducted including underground storage (geological sequestration), ocean disposal, biological fixation, and using CO_2 as feed material to produce various environmentally benign products. Mineral sequestration is a relatively new and less explored method of sequestering CO_2 , which involves the reaction of CO_2 with minerals to form geologically stable carbonates, i.e. mineral carbonation.

In 2007, Prof. Dr. Olaf Schuiling submitted a number of articles and proposals to the Dutch Ministry of Housing, Spatial Planning and the Environment (VROM) in which CO_2 sequestration under natural conditions by carbonation of the mineral olivine is claimed to be a viable alternative for more expensive technological processes concerning CO_2 reduction. VROM has requested TNO to collect background information on the possible application of olivine under natural conditions in the global warming issue.

The objectives of this desk study are to prepare:

- Literature overview on mineral carbonation using olivine;
- Overview of the relevant chemistry of the reaction(s) of CO₂ with olivine;
- Overview of the world-wide olivine occurrences;
- Estimate of the amounts and costs involved.

The first two items are covered in Chapter 2. Chapter 3 describes the world-wide olivine occurrences; a complete overview is added in Appendix A. An estimate of the costs involved is given in Chapter 4 and Chapter 5 presents the conclusions of this desk study.

2 Chemistry of natural weathering of olivine

Mineral sequestration aims to trap carbon in the form of solid carbonate salts. Complex silicates are converted to magnesium and calcium bicarbonate and silica, ultimately followed by precipitation of limestone (CaCO₃) and dolomite (MgCO₃). This is a natural proces and is responsible for much of the surface limestone. Olivine is a magnesium iron silicate that forms a solid solution series between the two end members Mg_2SiO_4 (forsterite) and Fe_2SiO_4 (fayalite). Olivine is very vulnerable to chemical weathering: the isolated tetrahedra of the olivine crystal structure allow olivine to be easily dissolved by acidic groundwater and rainwater. Dissolution of the silicate tetrahedra releases the Fe^{2+} and Mg^{2+} ions held in the crystal lattice. Cracking in olivine crystals increases the surface area exposed to pore water and speeds up the process of chemical weathering.

The formation of carbonates can only take place after the dissolution of olivine (Chen, 2006), and this process is most likely the rate-limiting step in the sequestration of CO_2 , and not the precipitation of the carbonates (Baines & Worden, 2000). In the next paragraph we will discuss the factors that influence the dissolution rate of olivine, assuming that these will be the factors that dictate the rate of CO_2 sequestration. Several major factors are the grain size, temperature, solution chemistry (pH, concentration of carbonate, magnesium, silica, organic acids, ionic strength), composition of the olivine (the contribution of forsterite versus fayalite), and the formation of coatings on the grains. Moreover there is a large discrepancy between data from the field and laboratory that needs to be considered.

2.1 Grain size

Reduction of the grain size causes an increase of the surface area and that has a direct influence on the reaction rate. A particle with a 10 times larger radius will take an order of magnitude longer to fully react (Stopar, 2006; Olsen & Rimstidt 2007). Especially high attrition grinding, which creates imperfections into the crystal lattice, accelerates the reaction. However, each grinding step comes with an energy penalty: the initial crushing costs 2 kW·h/tonne, grinding to minus 75 μ m adds 11 kW·h/tonne and to minus 38 μ m another 70 kW·h/tonne, attrition grinding comes at a cost of 150 kW·h/tonne (Gerdemann, 2007). The energy consumption can be translated into costs involved (0.06 €/ kW·h (Huijgen, 2007) and production of CO2 (0.45 kg/kW·h for natural gas and 0.87 kg/kW·h for coal-powered power plants).

2.2 pH and chemical composition of the water

When it comes to solution chemistry, pH is the single most important factor to consider. In acidic solutions, the exchange of two hydrogen ions for a magnesium or iron atom on the olivine surface is followed by the decomposition of a silica-enriched (magnesium/iron-deficient) complex. Under alkaline conditions, magnesium hydrated sites decompose after the preferential release of silica (Prokovsky & Schott, 2000). The rates of both processes are controlled by proton adsorption, which clarifies the influence of pH. In an overview of existing data, Olsen and Rimstidt (2007) detect a clear break around pH 6; in an acidic environment the increase in dissolution rate with decreasing pH was stronger than at alkaline pH values. They were able to fit two separate equations to the data:

$$\log r_{pH<6} = -0.48 \times pH - 6.90 \tag{1}$$

$$\log r_{pH>6} = -0.18 \times pH - 8.8 \tag{2}$$

where *r* is the dissolution rate in mol/m²sec. These slopes are similar to those by Prokovsky and Schott (2000), at 0.5 and approximately 0.1 respectively, although they place the break between pH 8 and 9. Under alkaline conditions (pH higher than 8), dissolution is supposedly inhibited at increased bicarbonate and dissolved silica concentrations according to some authors (Prokovsky, 2000). Other authors detect no effect of carbonate and bicarbonate concentrations up to 0.01 M, a concentration far higher than anticipated in natural environments (Golubev, 2005). The magnesium concentration in solution has no effect on the dissolution rate (Prokovsky, 2000), as has the partial CO₂ pressure at natural levels (Giammar, 2005; Golubev, 2005). Organic acids, however, form a complex with magnesium and iron on the surface, thus destabilising the metal-oxygen bond and increasing the dissolution rate (Wogelius & Walther, 1991).

2.3 Composition of olivine and formation of passivating layers

The dissolution rate of fayalite, the iron end member of olivine, is about 50 times higher than that of its magnesium-containing counterpart forsterite. Westrich et al. (1993) plot the dissolution rate of several end member orthosilicate minerals and finds a clear relationship with the rate of water exchange from solution into the hydration sphere of the dissolved cation. The dissolution rate of silicates of intermediate composition can be calculated from the rates of the end members (Westrich et al., 1993). These calculations are confirmed by experimental results: based on the theory, Wogelius and Walther (1992) were supposed to find a factor nine difference in rate between the fayalitic (Fo₆Fa₉₄) and forsteritic (Fo₉₁Fa₉) olivine they used, in practice the fayalitic olivine dissolved six times faster. However, the released iron may rapidly be oxidized and/or precipitated as (hydr) oxide as is demonstrated by the presence of iron-oxide phases on olivine surfaces under natural conditions (Wogelius & Walther, 1992), and this will limit its functionality in the sequestration of CO₂ as no carbonate minerals are formed.

The formation of coatings due to precipitation or incongruent dissolution may create a passivating layer. Under technological conditions (high temperature and CO_2 pressure), carbonation increases in the presence of abrasive (quartz) particles. The layer consists of SiO_x with some crystalline magnesite (Bearat, 2006) and possibly iron(hydr)oxides (Wogelius and Walther, 1992). The layer is highly stressed, because the molar volume of the SiO₂ is only 62% of the volume of the original olivine. Even though part of this volume reduction is compensated by the presence of MgCO₃, which is 130% of the volume of the original feedstock, the process results in a layer that is easily fractured and exfoliated (McKelvy, 2003). The presence of magnesite crystals indicates that there is significant transport of reaction products (Mg²⁺, H⁺, H₂O, CO₂, and HCO₃⁻) through the layer (McKelvy, 2006).

2.4 Temperature

The effect of temperature is interrelated with several other factors: at higher temperatures the dissolution of forsterite is more pH-dependent (Chen & Brantley,

2000) and plant degradation, which results in organic acids, is faster (Kump, 2000). According to Olsen and Rimstidt (2007), the dissolution of olivine will proceed 10 times quicker at a temperature of 298 °K than at 273 °K. In section 2.6 the effect of temperature has been addressed in several scenarios by using the Arrhenius equation (Equation 3).

2.5 Differences between field and laboratory reaction rates

The difference in reaction rate between field and laboratory experiments is generally several orders of magnitude (Kowalewski & Rimstidt, 2003; Kump, 2000; Olsen & Rimstidt, 2007; Stopar, 2006; Velbel, 1993). Environmental factors that may cause (part of) this discrepancy are the fluid to mineral ratio (Casey, 1993), effective surface area (Gislason, 1993), 'aging' of the mineral surface (White & Brantley, 2003), a difference in composition between the solution in micro channels and macropores (Casey, 1993), and the saturation state (Gislason, 1993). Freshly ground surfaces are highly reactive, while field-weathered surfaces are relatively non-reactive (Kump, 2000), contain fewer surface defects (Stopar, 2006), and are depleted of energetically reactive surfaces (White & Brantley, 2003).

In an experimental set-up the fluid to solid ratio is generally high, the mineral is freshly ground, the system is well-mixed, so all surface area is in contact with the solution and all the fluids are far from rate-limiting equilibrium. In the field, preferential flow makes that only 0.1 to 10 % of the mineral surface area is participating in reactions (Velbel, 1993). The other areas may be in contact with pore water, but if the interaction with the main flow is limited, the concentration of dissolution products will be higher and the increased saturation will slow the dissolution rates down (Stopar, 2006). Under natural conditions, dissolution in the macropores is limited by the reactions on the interface, whereas the rates in the 'inactive' pores are limited by the transport of reactants. Besides, intermittent wetting, like in a natural environment, is thought to decrease reaction rates due to the accumulation of leached layers and precipitation of secondary phases and a higher saturation level of the solution (Stopar, 2006).

Column studies revealed that the weathering rates on freshly ground minerals decreased parabolically over 6 years (White & Brantley, 2003). The authors calculated that it would take several thousand years to approach natural weathering rates under identical experimental conditions, and even then the rates would still be orders of magnitude higher than natural rates due to favourable hydrological conditions in the experiments.

2.6 Lifetime diagram and scenario calculations

Olsen (2007) created a diagram which displays the lifetime of forsterite as a function of pH in combination with the influence of most of the above-mentioned factors (Figure 1). The diagram enables us to calculate the lifetime of olivine under different environmental conditions and the CO_2 consumption involved.



Figure 1 Forsterite grain lifetime diagram by Olsen (2007). Forsterite grain lifetimes are given as a function of pH for laboratory conditions at 298 K. Vertical scales show how changing variables increase (scales above graph) or decrease (scales below graph) olivine grain lifetimes. Scales have been placed at arbitrary pH values and can describe the effects at any pH.

The effect of pH value, temperature, DOC concentration, and grain size has been calculated in several combinations that represent to two main scenarios: the spreading of olivine on the coastline and the application of olivine as a replacement of lime in agriculture. In both scenarios temperatures have been selected that represent application under tropical conditions and in a cooler temperate climate. Equations one and two were applied to include the pH value of the solution and the Arrhenius equation (Equation 3) is used to calculate the influence of temperature on the time it takes to completely dissolve olivine:

$$\ln k_{T} = \ln k_{T,R} - \frac{E_{a}}{R} \left(\frac{1}{T} - \frac{1}{T_{R}} \right)$$
(3)

where k_T is the dissolution rate at temperature T, $k_{T,R}$ is the rate at the reference temperature (T_R), which is 298 °K is our calculations, E_a is the activation energy (63 kJ/mol (Olsen & Rimstidt, 2007)), and R is the universal gas constant (8.31 JK⁻¹mol⁻¹). The effect of dissolved organic carbon (DOC) was derived from figure 4.7 in Olsen (2007) to the following equation:

$$E_{DOC} = 2.2076 \times [DOC]^{0.306} \quad R^2 = 0.9966 \tag{4}$$

where E_{DOC} is the effect that DOC has on the dissolution rate (a dimensionless factor) and (DOC) is the DOC concentration in mg C/L. A shrinking core model can be used to calculate the percentage of olivine that dissolved after a certain period of time by using the equations:

$$X_{olivine} = \frac{d_0^3 - d(t)3}{d_0^3} \times 100\%$$
(5)

$$d(t) = d_0 - 2R_{diss}\Omega t \tag{6}$$

where $X_{olivine}$ is the percentage of olivine that is dissolved, d_0 the initial grain size (µm), d is the grain size at time t (µm), R_{diss} is the dissolution rate of olivine (molm⁻²s⁻¹), Ω is the molar volume of olivine (43.97 cm³/mole), and t is time (s). Filling out equations 5 and 6 reveals that, independent of dissolution rate and initial grain size, within 40% of the time that is needed to completely dissolve the particles the first 80% will be dissolved. From equation 6 also follows that there is a linear relationship between initial grain size and the time is takes to dissolve: a particle twice the size will take twice as long to dissolve.

The results of the scenario calculations are presented in Table 1. The rates for forsteritic olivine were calculated; if fayalitic olivine is used the dissolution rate will be higher, however, if the iron precipitates as hydr(oxide), no gain in terms of CO₂ sequestration will be realised. The grain size is chosen because of the known price for grinding to < 75 μ m, however, when the olivine is ground to < 75 μ m the average particle size will actually be smaller. The calculated lifetimes of the particles are consequently too high.

Table 1Calculation of the time in years it takes to dissolve 75 μm olivine grains under several
conditions. Do note that the difference in rates between experimental and field conditions has
not yet been taken into account. Additional scenarios (different grain sizes, additional pH
values and temperatures) are presented in Appendix A.

	Coast	line	Agriculture	
pH	7.9	7.9	5.5	5.5
DOC (mg/L)	10	10	40	40
Temperature (°C)	10	25	8	25
Time (100 % dissolved)	388	66	64	14
Time (80 % dissolved)	161	28	27	6

According to the diagram, we have to multiply the outcome by two or three orders of magnitude to translate these laboratory-based lifetimes into a natural situation, and that would render the carbonation of olivine under natural conditions unfeasible. However, several of the factors that contribute to the difference between laboratory and field are not applicable to this situation. For example, the olivine will be ground, so the decrease in dissolution rate that is caused by a decreased reactivity of the surface will be avoided. In case the olivine is spread along the coastline, all surface area will be in contact with solution, the fluid to mineral ratio is high, and there will be no difference in solution composition between macro- and micropores. Under agricultural conditions the fluid to mineral ratio will be lower, not all olivine will be in contact with water at any moment in time and intermittent wetting may decrease the dissolution rate. However, plants do have a positive influence the physical environment: they increase the surface area exposed to water by attaching fine particles, increase the residence time of water, and enhance the transport of water due to evapotranspiration (Kump 2000). Overall we may have to increase the time estimate for an agricultural setting, although the reactivity of the freshly ground surface is advantageous for quick dissolution, and the reaction rate of olivine in seawater could actually be close to laboratory conditions.

According to Kojima et al. (1997) the following reaction will take place:

$$Mg_2SiO_4 + 4CO_2 + 2H_2O \leftrightarrow 2Mg^{2+} + 4HCO_3^- + SiO_2 \tag{7}$$

While according to Schuiling and Krijgsman (2006) the reaction will be:

$$Mg_2SiO_4 + 4CO_2 + 4H_2O \leftrightarrow 2Mg^{2+} + 4HCO_3^- + H_4SiO_4$$
(8)

Both reactions use 1.25 tonne of CO₂ per tonne of olivine. An increase in the bicarbonate concentration in seawater will increase calcification and may be as much as double the skeletal growth of coral (Marubini and Thake, 1999). The increase in the bicarbonate concentration will change the carbon cycle in the ocean and could have unforeseen consequences. However, when we take the volume of the ocean into account $(1.3 \times 10^{18} \text{ tonnes})$ and assume that for the next 100 years all CO₂ (annual production worldwide in 2004 27.2x10⁹ tonnes) will be transformed into bicarbonate, the current bicarbonate concentration of 142 mg/L will increase with 2%. Due to incomplete mixing, this may be an underestimation for the coastal areas, but even then, it is based on complete transformation into bicarbonate during 100 years. The process will increase the pH of the seawater and will counteract the 0.1 decrease in the pH value since pre-industrial times (Herzog et al., 2001). Under agricultural conditions, an increase in the bicarbonate concentration may instigate chlorosis due to a reduced uptake of, for example, zinc, iron, or manganese (Yang et al. 1993). If calcite is deposited, half of the CO₂ in bicarbonate will be stored in the calcite and half will be released so the overall CO₂ sequestrations will be 0.6 tonnes of CO₂ per tonne of olivine.

3 Olivine occurrences

Olivine has a low silica content and occurs in mafic to ultramafic rocks. The mineral is the main component of peridotites. The most olivine rich member of the peridotites is dunite, a rock with an olivine content of more than 90%. Related rock types are harzburgite, wehrlite en lherzolite, all with olivine contents between 50 and 90%. Often these rocks occur in so-called ophiolite complexes and in layered intrusions. Generally, the olivine in peridotites is magnesium-rich with forsterite contents of 87 to 96% (Fo₈₇ to Fo₉₆). Other minerals that often co-exist with olivine are Ca/Mg/Fe-silicates like clino- en orthopyroxene. The specifications of commercial olivine are:

SiO ₂	40	-	43	wt%
MgO	45	-	51	wt%
Fe ₂ O ₃	7	-	8	wt%
CaO	0.2	-	0.8	wt%
$Al_2O_3 + TiO_2$	1.8	-	2.0	wt%

Serpentinites are rocks composed of serpentine $(Mg_3Si_2O_5(OH)_4)$. Serpentinite is derived from the hydrothermal alteration of previously existing minerals of ultramafic rocks, such as olivine and pyroxene. During the alteration of olivine to serpentine the silica content of the rock is enriched and consequently the sequestration capacity is reduced. Upon heating to 600°C serpentine breaks down again into forsterite and silica under release of its crystal water:

$$2Mg_3Si_2O_5(OH)_4 \rightarrow 3Mg_2SiO_4 + SiO_2 + 4H_2O$$
 (Deer et al., 1992) (9)

In many deposits part of the olivine is altered to serpentine.

Most olivine is used in the iron and steel industry as an additive during the sintering process, slag conditioner and foundry sand. The world's combined annual production of olivine, serpentine and dunite material for iron and steel applications is in excess of 8 million tonnes (Table 2). Of this, the pure olivine mineral part is 4 million tonnes of which 3.3 million tonnes are applied in Europe. The former USSR states are not included in these figures. Large amounts of olivine and serpentine are also used for construction purposes. For example, in Japan about 50% of the total production of some 5-6 million tonnes per year is used in construction. Norway is the largest producer of olivine. Other producers include Australia, Austria, Brazil, China, Greenland, Italy, and Japan.

Country	Production (* 10 ³ tonnes/year	Country	Production (* 10 ³ tonnes/year		
Norway	3500	Italy	250		
Japan	2000	Turkey	150		
Spain	700	Mexico	150		
South Korea	500	USA	100		
China	350-500	Australia	80		
Taiwan	400	Austria	20		
Brazil	350				
	Total production 870				

 Table 2
 World production of peridotite and serpentinite for iron and steel applications (2003, former USSR not included) Source: USGS industrial minerals.

One should keep in mind that the composition of so-called commercial grades of olivine or dunite may differ from the bulk composition of the body that it is mined from. This is illustrated by the comparison of commercial grade olivine derived from the Orhaneli dunite in Turkey and the compositional range of 18 dunite samples form the intrusive (Table 3). A full overview of olivine occurrences is given in Appendix B.

Commercial g	rade	Range 18 selected samples		
Component	wt %	Component	wt %	
MgO	48 - 50	MgO	45.1-49.1	
SiO ₂	41 - 43	SiO_2	36.4-45.6	
FeO	6.1 - 6.6	FeO	6.2 - 9.2	
CaO	0.2 - 0.8	CaO	0.02 - 0.41	
Al_2O_3	no data	Al_2O_3	0.22 - 1.4	

Table 3Comparison of 'commercial grade' olivine and the bulk composition of the Orhaneli dunite in
Turkey (Örgün et al., 2004)

4 Amount, costs, and practical considerations

4.1 Amount and cost of the olivine needed

If we would like to sequester enough CO_2 in order to fulfil the requirements that Kyoto sets for the European Union (counting 15 member states at the time), how much olivine would we need? Between 2008 and 2012, Europe needs to reduce its CO_2 emission by 8 % as compared to the emission in 1990. In 1990, the 15 EU-countries produced 3135 million tonnes of CO_2 , in 2005 the amount increased to 3163 million tonnes (European Environmental Agency). In order to reach the target, 279 million tonnes of CO_2 has to be sequestered per year; at 1.25 tonne of CO_2 per tonne of olivine, that implies that we need to mine 223 million tonnes of olivine. The current worldwide (without the former USSR) olivine production is 4 million tonnes per year, so a significant increase in the mining of olivine would have to take place. For the sequestration of the annual worldwide production of CO_2 (27 246 million tonnes), an amount of 21 796 million tonnes of olivine would be needed.

The price of olivine ranges from 15 to 75 Euro per tonne including delivery at a European location and depending on the grade and size. The energy used for grinding to $< 75 \mu m$ adds $\in 0.66$ to the price per tonne, grinding to $< 38 \mu m$ adds another $\in 4.20$. Besides the CO₂ sequestration efficiency is reduced by 0.77 % and 4.80 % when these respective grinding treatments are applied due to the CO₂ production during the process.

4.2 Practical considerations

An increase in the olivine production can be realised: large known occurrences are not yet in production. In order to avoid the costs and CO_2 production of transportation (1103 g of CO_2 /tonne·km for trucks and 0.038 g CO_2 /tonne·km for shipping) and considering the large difference in reaction rates between warmer and colder climates, it is important to develop olivine exploitation in the area around the equator. In Figure 2 the presence of peridotite and serpentinite ore bodies are depicted and comparison with worldwide seawater temperatures (Figure 3) shows that especially in the Caribbean, at the southern end of the Arabian Peninsula and in the area around Indonesia and the Philippines a good match occurs between the presence of olivine and a high seawater temperature. The amount and quality of olivine that is present in Africa is not well known, but if significant quantities of good quality are present, mining would be profitable on this continent as well.



Figure 2 Peridotite and serpentinite ore bodies (red dots)



Figure 3 Temperature of the ocean water at the surface as measured by NOAA (The National Oceanic and Atmospheric Administration) from August 6-11, 2001.

Mineral carbonation is usually applied under high pressure and temperature conditions, but this literature study shows that even under natural conditions carbonation of the mineral olivine is likely to happen relatively quickly. Olivine is one of the most common minerals in the world, and occurs on all continents. The current production of 4 million tonnes per year (data from 2003) is mainly mined in Norway and Japan and is used in the steel and iron industry.

Two different scenarios, the spreading of olivine on the coastline and the application of olivine as a replacement of lime in agriculture, have been evaluated based on a model developed by Olsen (2007). A small grain size, low pH value, high temperature, and the presence of organic acids are favourable for the dissolution of olivine. Under tropical conditions 80% of the olivine will dissolve in 6 years when applied in an agricultural setting and in 28 years when the olivine is spread along the coastline.

One factor that can not be accurately quantified is the difference between laboratory and field data: the model indicates that the dissolution of olivine occurs 100 to 1000 times slower in the field as compared to the rates that are determined in the lab. However, several of the factors that contribute to the difference between laboratory and field (reactivity of the surface, contact of the surface with solution, local saturation of the solution) are of limited or no relevance to these scenarios. For example, the olivine will be ground, so the decrease in dissolution rate that is caused by a decreased reactivity of the surface will be avoided. Overall we may have to increase the time estimate for an agricultural setting, but the reaction rate of olivine in seawater could actually be close to laboratory conditions, making spreading of olivine along the coastline under tropical conditions a viable option for CO_2 sequestration.

If the EU would like to meet its requirements under the Kyoto protocol by sequestration of CO_2 with olivine (at 1.25 tonnes of CO_2 per tonne of olivine), the worldwide production of olivine would need to increase by a factor 55. In order to capture the annual worldwide CO_2 production, the mining of olivine would need to increase another 100 times. Many large olivine occurrences are not yet in production, and several of those are located in tropical regions, so the costs of transportation would be limited if the olivine was applied in the vicinity of the mine. Currently the cost of olivine (mined in Greenland) ranges from 15 to 75 Euro per tonne, which means that the price per tonne of CO_2 avoided is between 12 and 60 Euro.

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A Additional scenario calculations

In this table some additional calculation of the time in years it takes to dissolve under several conditions. Do note that the difference in rates between experimental and field conditions has not yet been taken into account. The grain sizes are chosen because of the known price for grinding to these sizes, however, when the olivine is ground to < 75 µm the average particle size will actually be smaller. The calculated lifetimes of the particles are consequently too high. Seawater pH values vary between 7.9 and 8.5, hence the selected pH values.

	DOC	temperature	pН	grainsize	100%	80%
	(mg/L)	(°C)		(µm)	dissolved	dissolved
					(years)	(years)
Coastline	10	10	7.9	75	388	161
				38	197	82
			8.5	75	498	207
				38	252	105
		25	7.9	75	101	42
				38	51	21
			8.5	75	129	54
				38	66	27
		30	7.9	75	66	28
				38	34	14
			8.5	75	85	35
				38	43	18
Agriculture	40	8	4.0	75	12	5
				38	6	3
			5.5	75	64	27
				38	32	13
			7.0	75	212	88
				38	107	45
		25	4.0	75	3	1
				38	1.3	0.6
			5.5	75	14	6
				38	7	3
			7.0	75	45	19
<u></u>				38	23	10

B Overview of olivine occurrences

Continent	Country	Mineral	Reserves	Exploitation
Europe	Albania	harzburgite/dunite		
	Austria	dunite/serpentinite		0.1 Mt/yar
				by Magnalithe GmbH,
				Hartsteinwerke Preg,
				Brandner&Lipp
	Cyprus	dunite		
	Great Britain	dunite/harzburgite		by Trevassik Quarry,
		/serpentinite		Lizard Quarry
	Greece	dunite/harzburgite		by Grecian Magnesite S.A.,
				Magnomin S.A.
	Greenland	dunite	> 46 Mt	Minelco
	Italy	dunite	> 100 Mt	0.25 Mt/year
				by Nueva Cives
	Kosovo	harzburgite/dunite		
	Norway	dunite/serpentinite	> 1025 Mt	3.05 Mt/year
		/peridotite		by North Cape Minerals
	Russia	peridotite/dunite		
	(excluding	/olivine/harzburgite		
	Siberia)			
	Spain	dunite/serpentinite	> 150 Mt	0.95 Mt/year
		/peridotite		by Pasek España SA
	Sweden	serpentinite/dunite		
	Switzerland	serpentinite/dunite		
Asia	India			
	Japan	dunite/serpentinite		
	Malaysia	dunite		
	Oman	dunite		
	New	harzburgite/dunite		
	Caledonia			
	Papua New	dunite/harzburgite		
	Guinea			
	Philippines			
	Taiwan	dunite		
	Thailand	dunite		
	Tibet	dunite		
Australia	Australia	dunite/serpentinised		
N. Zeal.and		peridotite		
	New Zealand	dunite		
America	Argentina	dunite/websterite		
	Brazil	dunite/harzburgite		
	Canada	dunite/serpentinite		
	Central	ophiolite/dunite		

	America and Caribbean	harzburgite		
	Colombia	dunite		
	Mexico	ophiolite/dunite		
	Venezuela	serpentinised dunite		
	USA	dunite/serpentinite	> 1 900 Mt	by Unimin Corp
Africa	Gabon	dunite		
	Ghana	peridotite/dunite /pyroxenite		
	Guinea	ultramafite/olivine		
	Morocco	peridotite		
	Sierra Leone	peridotite		
	South Africa	dunite		
	Sudan	dunite/harzburgite		
	Tanzania	dunite		
	Zimbabwe	cumulus olivine		

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