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Rolling stones; fast weathering of olivine in shallow seas for cost-effective CO_2 capture and mitigation of global warming and ocean acidification

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Abstract

Human CO_2 emissions may drive the Earth into a next greenhouse state. They can be mitigated by accelerating weathering of natural rock under the uptake of CO_2. We disprove the paradigm that olivine weathering in nature would be a slow process, and show that it is not needed to mill olivine to very fine, 10 µm-size grains in order to arrive at a complete dissolution within 1–2 year. In high-energy shallow marine environments olivine grains and reaction products on the grain surfaces, that otherwise would greatly retard the reaction, are abraded so that the chemical reaction is much accelerated. When kept in motion even large olivine grains rubbing and bumping against each other quickly produce fine clay- and silt-sized olivine particles that show a fast chemical reaction. Spreading of olivine in the world's 2 % most energetic shelf seas can compensate a year's global CO_2 emissions and counteract ocean acidification against a price well below that of carbon credits.

1 Introduction

The burning of fossil fuels, up to a volume (in oil equivalents) of 10 km^3 annually, has greatly accelerated half of the global carbon cycle and may greatly affect the future evolution of our planet (Hay, 2011). Instead of looking for solutions at the same scale by also accelerating the other half of the cycle by enhanced burial of organic matter and silicate weathering, the focus is on small-scale technical solutions. The intended policy is to capture CO_2 at refineries and power plants where it is produced and to store it in the subsurface, CCS (a Faustian bargain?, Spreng et al., 2007), at high expenses (Haszeldine, 2006; Bachu, 2008; McKinsey and Company, 2008), with the risk of leakage (Gherardi et al., 2007), and considering the energy penalty of ~25 % (McKinsey and Company, 2008) also at considerable economical and political risks; CCS and other techniques with a high energy penalty reinforce the incumbents with the risk of a lock-in of fossil-fuel-based technologies and a lock-out of other scenarios;

it increases the international transfers of resource rent cash flows towards politically less stable regimes with the risk of political instability (Jansen and Seebregts, 2010).

Since the birth of our planet, rock weathering has captured CO_2 and since the development of photosynthesis fossilizing organic matter also contributes to its removal (Berner and Kothavala, 2001). Volcanoes release between 0.13 and 0.44 Gt CO_2 annually (Gerlach, 2011), but this does not include the contribution by diffuse CO_2 emissions, which are for a large part not related to volcanism, but to the dissociation of subducted limestones, as evidenced by their $\delta^{13}C$ signature of around 0 ‰ (Schuiling, 2004). Although less spectacular, these may be more voluminous. We estimate that present-day CO_2 emissions are around 30 times larger than the natural emissions from volcanoes and limestone dissociation combined.

Similarly as mankind has accelerated the release of CO_2, it can accelerate natural rock weathering thus mitigating the CO_2 produced by fossil fuel burning.

In the reaction

$$(Mg, Fe)_2SiO_4 \text{ (olivine}^1) + 4\,H_2O \longrightarrow, 2\,(Mg, Fe)^{2+} + 4\,OH^- + H_4SiO_4$$

followed by

$$4\,OH^- + 4\,CO_2 \longrightarrow 4\,HCO_3^-.$$

CO_2 is consumed, and Mg^{2+}, Fe^{2+}, H_4SiO_4 and HCO_3^- are produced. The reaction of serpentine is similar:

$$Mg_3Si_2O_5(OH)_4 \longrightarrow 3\,Mg^{2+} + 6\,OH^- + 2\,H_4SiO_4$$

followed by

$$6\,OH^- + 6\,CO_2 \longrightarrow 6\,HCO_3^-.$$

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[1] Olivine = minable olivine consists, with minor variations, of 0.92 Mg_2SiO_4 (forsterite) and 0.08 Fe_2SiO_4 (fayalite).

Weathering of olivine in nature

“Olivine weathers more rapidly than any other major rock-forming silicate mineral ... so olivine does not usually survive to be part of the heavy-mineral fraction of most soils, sediments, or sedimentary rocks” (Velbel, 2009). Areas with groundwater passing through zones with a high proportion of reactive minerals show lower tendencies to acidification than others, and vegetation also has a positive effect on silicate weathering (Moulton et al., 2000), which is obvious from the fact that olivine grains from nearby sources are not commonly found in soils (Wilson, 2004). Biological activity can greatly accelerate the weathering of mafic silicates under the uptake of CO_2 (cf. Wilson, 2004; Needham et al., 2006; Taylor et al., 2009; Schuiling and de Boer, 2010; Brantley et al., 2011). Carbon sequestration in ultramafic mining waste may be considerable (Pronost et al., 2011; Wilson et al., 2011). “Experimental studies of ambient temperature olivine dissolution kinetics suggest that olivine dissolution is interface-limited, with the rate-determining step being the breakdown of a surface activated complex” (Velbel, 2009). The physical removal of surface complexes thus is expected to accelerate olivine weathering.

2 Experiment

Three Erlenmeyer flasks were filled with 250 ml tap water and 30 g olivine from Norway with a grain size of 0.71–1.4 mm and three others with 250 ml tap water and 30 g olivine grit (2–5 mm) from Turkey. A third experiment was carried out with a 50/50 mixture of both. The Erlenmeyer flasks were put on a table top rotary shaker, so that the grains kept rotating along the bottom. The Erlenmeyers were open to the air, permitting CO_2 exchange. In all cases the tap water had an initial pH of 8.22, and the olivine grains had been washed to remove any attached dust.

3 Results

Grains of both sizes became visibly rounded (Fig. 1). The abraded fine material caused the water in the bottles with fine-grained olivine to be cloudy. In the bottles with coarse-grained olivine the water was a milky white, opaque suspension (Fig. 2). Already after 6 h the pH had risen to 8.82 in one bottle with coarse-grained olivine. After 1, 4 and 10 days the pH in one of each of the two series of bottles was measured. In a bottle with fine-grained olivine the pH had risen to 8.91 after 24 h, and in a bottle with coarse grains to 9.02 (Table 1). This implies that the coarse grains had produced more slivers than the finer grains, likely due to their greater mass and consequently heavier mutual impacts. In this experiment the olivine reacted with a limited, fixed amount of water. Thus, logically the accumulation of reaction products slowed down and eventually stopped the reaction (Table 1), contrary to shallow marine settings where water refreshment is continuous and the tiny slivers, together with the dissolved magnesium and silica, are carried away.

Figure 3 shows the grain size distribution of the material that was scraped off the grains in the test with 50/50 coarse and fine grains. The amount of scraped off material was 9 g (30 %), considerably more than the amounts in the single grain size experiments which were slightly over 1 g in the separate fine and coarse fractions. The pH in the 50/50 mixture rose to 9.42 in 12 days. About 50 wt % of the abraded particles is 5 µm or less (Figs. 3 and 4). Such fine grains of olivine weather fast, as is evident from the rapid rise of pH during the experiments. The rate of solution may have been further increased by the deformation that grains undergo when they are strained. The phenomenon of enhanced dissolution of fine particles by repeated particle impact and rubbing is well known in the field of mineral processing, where it is called mechanical activation (Tromans and Meech, 2001).

4 Discussion

Contrary to the common opinion that olivine weathering in nature is a slow process (Lal, 2008; Hangx and Spiers, 2009; Köhler et al., 2010; Pronost et al., 2011), our experiments show that olivine grains when kept in motion weather fast because continuous mutual impacts remove reaction-inhibiting silica from the surface and tiny µm-size slivers are produced allowing a fast chemical reaction. The application of olivine and other (ultra)mafic minerals like serpentine in high-energy shallow marine environments can make a significant contribution in the fight against climate change. The counteracting effect on ocean acidification is immediate.

4.1 Application

For the removal of CO_2 olivine should be applied to environments where a fast reaction takes place. Wetlands and agricultural land in the humid tropics, where the Mg released will have the additional favour to act as fertilizer (Hartmann and Kempe, 2008), is an option (Hangx and Spiers, 2009), though the saturation of silicic acid is claimed to be limiting (Béarat et al., 2006; Köhler et al., 2010). The option proposed here, applying olivine grains with sizes up to that of the sediment that is transported locally, to high-energetic marine environments requires a substantial surface area. Many shelf seas experience continuous tidal currents and incidentally heavy wave action making them a suitable ball mill for the breakdown of olivine. For example part of the continental shelf between the Shetland Isles and France, that is the Southern Bight of the North Sea, the English Channel and the Irish Sea, is covered with sand waves (Kenyon and Stride, 1970), and in an around the English Channel an area of the order of 35 000 km^2 experiences bed shear stresses capable of transporting gravel (Mitchell et al., 2010). A volume of 0.35 km^3 olivine grains, one cm thick, when applied to the area where gravel can be transported, thus would compensate 5 % of a year's global CO_2 emissions. This 5 % exceeds the combined annual CO_2 emissions of the adjacent countries, The UK, France, The Netherlands, Belgium and Ireland, which together are responsible for about 4 % of the world's CO_2 emissions. This would be 1.2 Gt or 3000 megacarrier loads of 400 000 metric tons of olivine, to be transported to suitable locations where it can be put overboard. If the chemical breakdown would take longer, the effect is delayed, but still much larger than that of pumping CO_2 into many abandoned oil and gas reservoirs.

Costs of mining and milling are of the order of 3–4 € per ton olivine (O'Connor et al., 2000) (price level 2000). Thus, together with transport and spreading, the price per ton captured CO_2 will be less than 10.- €, that is well below the current price of CO_2 emission rights. Such low cost can be achieved when the weathering process of olivine and serpentine grains is left entirely to nature, without using technologies like thermal activation, addition of chemicals, mechanical activation, the use of autoclaves, subsurface injection of CO_2 in peridotites and other (ultra)mafic rocks, and milling to very fine grain sizes (cf. Lackner, 2002; Kelemen and Matter, 2008; Lal, 2008; Köhler et al., 2010; Haug et al., 2011; Krevor and Lackner, 2011; Zevenhoven et al., 2011). Large-scale experiments on the spreading of olivine grains along beaches and in shallow seas, where grain collisions make these the cheapest ball mills available, are more relevant than theoretical models and high-tech experiments which ignore important natural factors.

4.2 Feasibility

The option to stimulate natural rock weathering by applying olivine (Schuiling and Krijgsman, 2006) has been proposed not to be feasible because dissolution kinetics and passivating silica-rich surface-layer formation would make the reaction far too slow (Béarat et al., 2006; Lal, 2008; Köhler et al., 2010) and also because the quantity needed, $\sim$7 km^3 olivine per year (that is $\sim$1 m^3/human) would be too great to be possibly mined and distributed (Hangx and Spiers, 2009). However, as demonstrated above, dissolution kinetics as defined on the basis of theoretical calculations are much accelerated in nature by mechanical and biological processes. The volume of $\sim$7 km^3 (23 Gt; billion tons) olivine is great, but less than the volume of hydrocarbons – expressed in oil equivalent ~10 km^3 – that is annually retrieved, often from great depths and at distant locations. Olivine and serpentine are readily available at the Earth' surface on all continents (Kelemen and Matter, 2008; Köhler et al., 2010; Zevenhoven et al., 2011), and past mining efforts show that such volumes are exceeded by existing mines. E.g. the Bingham Canyon open pit mine in Utah has an excavated volume of 25 km^3, and worldwide 6 Gt ores and industrial minerals and more than 20 Gt construction minerals were mined in 2005 (Krausmann et al., 2009).

The above volume is large indeed. E.g. in Norway 1.3–3.6 million ton olivine was mined annually in the past 10 years (Geological-Survey-Norway, 2010). Expansion of olivine mining activities, possible on all continents, is a, though large-scale, simple technical enterprise, much easier and much cheaper than CCS and other expensive techniques for CO_2 sequestration. Extrapolation of the 3.4 million tons of olivine that were mined with 225 employees in Norway in 2004 (Geological-Survey-Norway, 2005) to the case in which all fossil fuel CO_2 produced by mankind will be neutralized by the application of olivine, shows that then 1 to 1.5 million people (~0.2 ‰ of mankind) will be employed in open-pit olivine mining world-wide, considerably less than in coal mining and oil industry. With 6–8 % of the Earth surface consisting of shelf seas, only about 2 % of these shelf seas would be needed to accomplish such goal.

4.3 Ocean chemistry

Mg is the third most abundant element in the ocean. The Mg/Ca ratio has varied in geological history from <2:1 to 5:1 today (Sandberg, 1975) and in an absolute sense Mg has varied more than a factor two (Lowenstein et al., 2001). After 100 years of addition of 7 km^3 ground olivine to the marine domain annually, the Mg in the ocean will have increased from 1296 ppm to 1296.6 ppm. Si is a limiting element for diatoms (Dugdale and Wilkerson, 1998), so olivine weathering will result in additional CO_2 uptake and in an increase of diatoms relative to other phytoplankton. Minable olivine consists, with minor variations, of 0.92 Mg_2SiO_4 (forsterite) and 0.08 Fe_2SiO_4 (fayalite). Thus Fe, a prime limiting element for ocean productivity (Morel and Price, 2003) released during olivine weathering will additionally favour the uptake of CO_2 and stimulate the carbon pump. Serpentine, $Mg_3Si_2O_5(OH)_4$, also abundantly available in parts of the world, releases Mg and Si under the uptake of CO_2 (see formulae on p.3). The increased bicarbonate content of sea water will reduce its acidity and favour corals and other carbonate-precipitating organisms. Upon the extra production of carbonate, half of the bicarbonate will be returned as CO_2 to the atmosphere in the long run (Berger, 1982), by which time CO_2-neutral energy supply may have become well established.

5 Conclusions

Olivine and other (ultra)mafic minerals in high-energy shallow-marine environments weather fast. Their application can make a significant contribution in the fight against climate change. The counteracting effect on ocean acidification is immediate. Large-scale spreading of olivine in shelf seas with adequate tidal currents and wave action can be started any moment. The scale on which olivine has to be spread, asks for a major expansion of olivine mining, a quintupling of the ores and industrial minerals mining industry, that is an amount more or less equal to the amount of construction minerals mined annually. This requires less effort and investments than those for the recovery of fossil fuels and CCS. The energy penalty, i.e. the extra fuel to be consumed for CCS is of the order of 25 % (McKinsey and Company, 2008); for the stimulated natural weathering of olivine this is less than 5 %.

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Table 1. Analytical data of samples after 1, 4 and 10 days, and 12 days for the 50/50 experiment. Fine olivine sand is 710–1400 µm, coarse olivine 2–5 mm. Samples were shaken continuously unless indicated otherwise. 50/50 refers to the experiment with a mixture of 15 g of the fine, and 15 g of the coarse fraction. The difference in Mg/Si ratios must be due to the precipitation of Mg- and Si-containing minerals. Indeed, XRD analysis showed a saponite-like ($Mg_3Si_4O_{10}(OH)_2$). $4\,H_2O$) phase and others that could not be readily identified. This is irrelevant in an open marine environment, where the sea water is continuously refreshed and such saturation is never reached. The decrease of C_{inorg} in the water must be due to the precipitation of mineral phases insufficiently developed to be recognized with XRD.

	pH	C_{inorg} ($mg\,l^{-1}$)	Mg-conc. ($mg\,l^{-1}$)	Si-conc. ($mg\,l^{-1}$)
Tap water	8.22	21.5	5.2	7.6
Day 1 fine	8.91	33.0	21.4	13.2
Day 4 fine	8.71 (no shaking for 3 days)	28.4	15.1	10.8
Day 4 (50/50)	8.93			
Day 10 fine	9.41	100.3	114.0	38.9
Day 1 coarse	9.02	38.7	34.7	24.9
Day 4 coarse	8.73 (no shaking for 3 days)	28.4	20.2	13.7
Day 10 coarse	9.22	63.1	74.5	34.9
Day 12 (50/50)	9.42	133.1	153.6	32.4

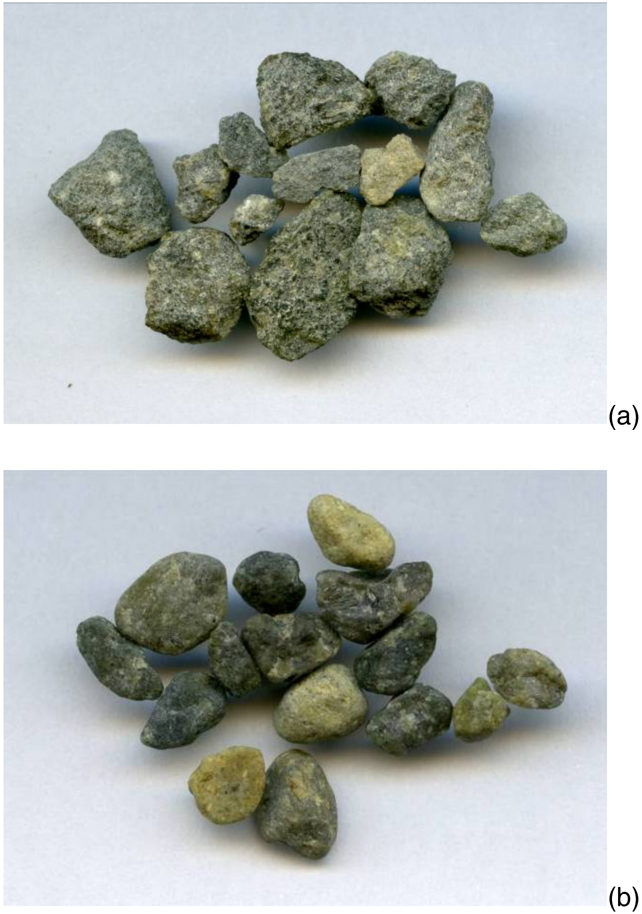

Fig. 1. Simulation of current action. **(a)** Olivine grit (2–5 mm) at start. **(b)** Olivine grit, rounded and polished after 3 days motion on a table top rotary shaker.

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Fig. 2. Coarse (2–5 mm) olivine grains in clear water (left panel) give a white milky opaque suspension (right panel) after continuous motion on the table top rotary shaker during 10 days.

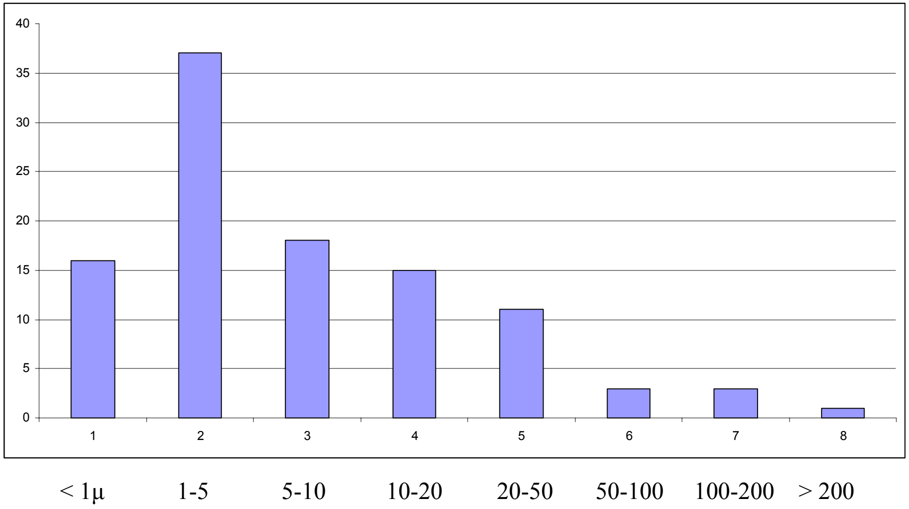

Fig. 3. Size distribution of the olivine scraps after 12 days of continuous motion on the table top rotary shaker. Scale in µm. Total weight 9 g. Original sample 30 g.

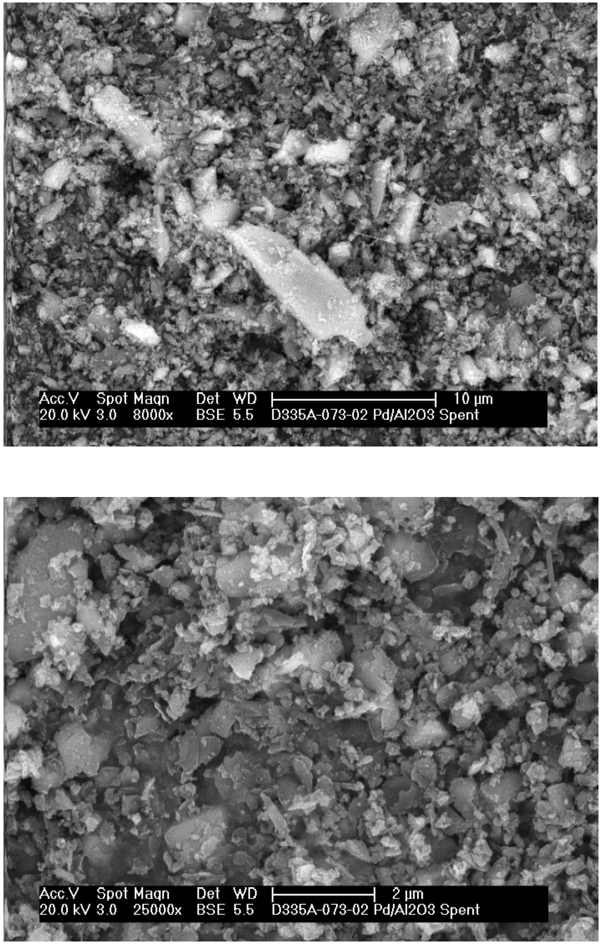

Fig. 4. SEM pictures of the clay- and silt-sized olivine dust that resulted from 10 days motion of the grains shown in Fig. 1.