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# Enhanced weathering of olivine in seawater: The efficiency as revealed by thermodynamic scenario analysis

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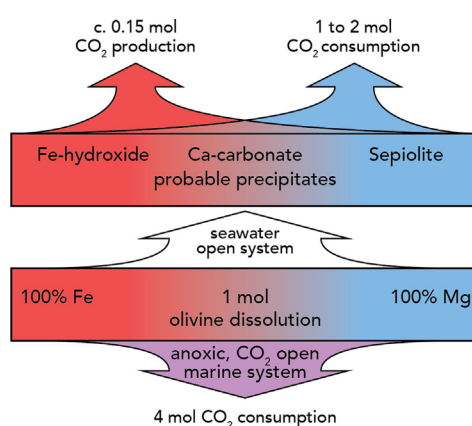
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## HIGHLIGHTS

- Measures may be needed to lower the atmospheric CO<sub>2</sub> level and combat climate change as well as ocean acidification
- Enhanced weathering of olivine in marine environment is such a measure
- The efficiency of olivine weathering depends on the Mg/Fe fractions of olivine and precipitation of secondary minerals or not
- For Fe-rich olivine, CO<sub>2</sub> may be released to the atmosphere and ocean pH might decrease during oxic olivine weathering

## GRAPHICAL ABSTRACT



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## ABSTRACT

Enhanced weathering of olivine has been suggested as a measure to lower the atmospheric CO<sub>2</sub> level and it might also mitigate ocean acidification. This study aimed to characterise how olivine can weather in seawater, to elucidate the role of secondary precipitation and to ascertain the efficiency in terms of molar CO<sub>2</sub> removal per mole of olivine dissolution. Geochemical thermodynamic equilibrium modelling was used, which considered both the variable mineralogical composition of olivine and the kinds of secondary precipitates that may be formed. The advantage is that such an approach is independent from local or regional factors as temperature, related kinetics, mineralogy, etc. The results show that the efficiency falls when secondary precipitates are formed. When Fe-bearing olivine undergoes weathering in an oxic environment, Fe(III) hydroxides will inevitably be formed, and as a result of this acidifying process, CO<sub>2</sub> could be released to the atmosphere. This might also enhance ocean acidification when Fe-rich olivine becomes used. Ocean alkalisation only happens when more than 1 mol/kg<sub>H<sub>2</sub>O</sub> Mg-rich olivine weathers. Maintenance of supersaturation for calcite or aragonite as holds in seawater reduces the efficiency by about a factor of two compared to the efficiency without secondary precipitation. Precipitation of sepiolite as Mg silicate reduces the efficiency even more. Magnesite precipitation has a similar effect to Ca carbonate precipitation, but according to the literature magnesite precipitation is improbable at ambient conditions and relatively low supersaturation. When less than 0.05 mmol olivine/kg(seawater) weathers the efficiency is slightly different than at higher intensities, due to strong buffering by seawater alkalinity.

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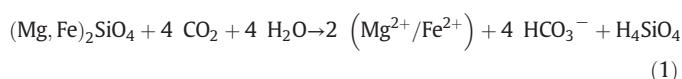
## 1. Introduction

One mitigating measure that has attracted attention to halt rising atmospheric concentrations of CO<sub>2</sub> is enhanced weathering: anthropogenically induced weathering resulting from introducing relatively rapidly weathering minerals, such as olivine, into the environment (Seifritz, 1990; Lackner, 2003; Lenton and Britton, 2006; Schuiling and Krijgsman, 2006; Köhler et al., 2010; Hartmann et al., 2013; Williamson, 2016). In principle, enhanced weathering can take place in terrestrial and marine environments (Hartmann et al., 2013). A similar measure is 'mineral carbonation', which is the accelerated weathering of easily weatherable rocks or minerals under high temperature and pressure; this can take place in geological formations or aboveground (e.g. Béarat et al., 2006; Alfredsson et al., 2008; Kelemen and Matter, 2008). The socio-scientific interest in these topics has stimulated research on the temperature dependence of weathering rates and the related formation of secondary products.

An idea proposed in the case of the marine environment is to scatter olivine grains on the open ocean, which will have largely dissolved before they disappear out of the surface mixed layer which is averagely 64 m thick (Köhler et al., 2013); it has also been proposed to scatter crushed olivine in shallow seawater (Schuiling and De Boer, 2011). Articles in the Dutch press led Hangx and Spiers (2009) to question whether the second idea is feasible geochemically and in terms of environmental economics. The rationale behind the idea of spreading olivine in shallow seawater is that in this water, and especially in coastal seawater, the grains undergo frequent agitation, which results in abrasion of the grains and in any coatings on the grains being rubbed off. These coatings are undesirable because they can retard or even prevent weathering, as a result of the limited exchange between CO<sub>2</sub> in the air and interface with the olivine crystal (e.g. Olsson et al., 2012; Wang and Giammar, 2012).

Weathering reactions are often alkalinising. Enhanced weathering of olivine in marine environment may then not only mitigate rising atmospheric concentrations of CO<sub>2</sub> but also mitigate the associated CO<sub>2</sub> problem of ocean acidification. Ocean acidification causes the risk that biologically controlled calcium carbonate precipitation will no longer be able to take place, and this is potentially damaging for marine ecosystems such as coral reefs (e.g. Atkinson and Cuet, 2008; Artioli et al., 2012; Feely et al., 2012). Addition of limestone or quicklime has been proposed to raise the ocean alkalinity initially in order to capture atmospheric CO<sub>2</sub> in the ocean and later on to combat ocean acidification as well (Kheshgi, 1995; Rau and Caldeira, 1999; Caldeira and Rau, 2000; Harvey, 2008; Ilyina et al., 2013): more anthropogenic carbon would be adsorbed by the ocean while buffering the acidifying effect of CO<sub>2</sub> on ocean pH. A substantial CO<sub>2</sub> release happens in producing quicklime from limestone which must be dealt with as well (Paquay and Zeebe, 2013; Renforth et al., 2013). Ocean liming and enhanced weathering of olivine in marine environment are thus comparable mitigating measures compared to alternatives as geological disposal of CO<sub>2</sub>.

Mg-rich and Fe-rich olivine occur often in nature, in a range of mixtures between the end-members forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) and fayalite (Fe<sub>2</sub>SiO<sub>4</sub>) (e.g. Deer et al., 2013). Huang (1989) noted that the most frequently occurring olivine appears to contain 30–50% Fe, which is the typical Fe fraction of olivine in gabbros (Deer et al., 2013). Ultramafic rocks such as dunite and peridotite contain much Mg and correspondingly contain the highest content of Mg-rich olivine (Krauskopf, 1982; Deer et al., 2013). The percentage of Mg in dunite-olivine varies: in regular dunites the range is 88–93% (Su et al., 2016) and in Fe-rich dunites it is 80–88% (Rehfeldt et al., 2007; Ackerman et al., 2009). Enhanced weathering of olivine is routinely described as a dissociation reaction of olivine (Schuiling and Krijgsman, 2006; Olsson et al., 2012; Schuiling and De Boer, 2011; Köhler et al., 2013; Hauck et al., 2016):



Moosdorf et al. (2014) assumed pure forsterite. Such reaction equations pay no attention to which secondary products might be formed. For example, dissolved Fe<sup>2+</sup> is not stable in an oxic environment and so will be oxidised to Fe oxyhydroxide, which is an acid-producing reaction. Béarat et al. (2006) and Hartmann et al. (2013) also presented the subsequent reaction for pure forsterite with MgCO<sub>3</sub> precipitation:



Note that the molar ratio of olivine to CO<sub>2</sub> differs between these two reaction equations by a factor of 2. Mineral carbonation of olivine is mainly written as a weathering reaction of forsterite to MgCO<sub>3</sub> (Béarat et al., 2006; Mani et al., 2008; Wang and Giammar, 2012; Johnson et al., 2014; Lafay et al., 2014). Köhler et al. (2010) noted that enhanced weathering of olivine in the marine environment will have an impact on the carbon cycle and the alkalinity, but did not elaborate further. It should be noted that in contrast to rainwater, seawater has a strong weathering signature, with high concentrations of Mg, SiO<sub>2</sub>, and alkalinity and also a fairly high pH. The degree of undersaturation will therefore be more limited than in slightly acid rainwater, and this can limit olivine weathering in this marine environment.

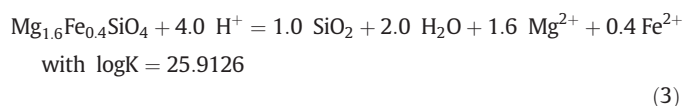
The research described here had three aims: to characterise how olivine can weather in seawater, to elucidate the role of secondary precipitation and to ascertain the related efficiency in terms of molar CO<sub>2</sub> removal per mole of olivine dissolution. The key questions addressed were:

- What is the difference in the weathering of forsterite, fayalite and a compound crystal that is a mixture of Mg<sub>x</sub>Fe<sub>1-x</sub> olivine?
- What is the influence of equilibrium or non-equilibrium with atmospheric CO<sub>2</sub>?
- What is the influence of maintaining or not maintaining equilibrium with Ca carbonate and/or Mg carbonate and, more specifically, with or without precipitation of magnesite, MgCO<sub>3</sub>?
- What is the influence of precipitation or non-precipitation of sepiolite (Mg<sub>4</sub>Si<sub>6</sub>O<sub>15</sub>(OH)<sub>2</sub>·6H<sub>2</sub>O) as a weathering product?

The first question is relevant because different olivine minerals occur in different rock types. The second and third questions are relevant because marine environments vary in the extent to which they may be closed to CO<sub>2</sub> exchange, with the beach zone having relatively good exchange with the air; also, marine environments do not have a thermodynamic equilibrium with pure carbonate materials such as calcite, dolomite, magnesite and aragonite. A multicomponent geochemical modelling approach based on thermodynamic equilibrium was followed. The study therefore focused on achieving an end stage in the weathering and not on how rapidly this was achieved or the factors also determining this. This implies that the outcome has a general, global meaning and is not limited by local or regional boundary conditions such as temperature, reaction kinetics, mineralogical composition of the seabed or suspended matter, etc.

## 2. Model set-up

The calculations were performed with the multicomponent geochemical model code PHREEQC (Parkhurst and Appelo, 2013), which is standardly equipped with several thermodynamic databases that can be used. In this research the LLNL database was used because it includes the solution reactions of fayalite and forsterite. Two olivine solid solutions were added:



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