



# The dissolution of olivine added to soil: Implications for enhanced weathering



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## ARTICLE INFO

### Article history:

Available online 27 May 2015

Editorial handling by M. Kersten

## ABSTRACT

Chemical weathering of silicate minerals consumes atmospheric CO<sub>2</sub> and is a fundamental component of geochemical cycles and of the climate system on long timescales. Artificial acceleration of such weathering (“enhanced weathering”) has recently been proposed as a method of mitigating anthropogenic climate change, by adding fine-grained silicate materials to continental surfaces. The efficacy of such intervention in the carbon cycle strongly depends on the mineral dissolution rates that occur, but these rates remain uncertain. Dissolution rates determined from catchment scale investigations are generally several orders of magnitude slower than those predicted from kinetic information derived from laboratory studies. Here we present results from laboratory flow-through dissolution experiments which seek to bridge this observational discrepancy by using columns of soil returned to the laboratory from a field site. We constrain the dissolution rate of olivine added to the top of one of these columns, while maintaining much of the complexity inherent in the soil environment. Continual addition of water to the top of the soil columns, and analysis of elemental composition of waters exiting at the base was conducted for a period of five months, and the solid and leachable composition of the soils was also assessed before and after the experiments. Chemical results indicate clear release of Mg<sup>2+</sup> from the dissolution of olivine and, by comparison with a control case, allow the rate of olivine dissolution to be estimated between 10<sup>-16.4</sup> and 10<sup>-15.5</sup> moles(Mg) cm<sup>-2</sup> s<sup>-1</sup>. Measurements also allow secondary mineral formation in the soil to be assessed, and suggest that no significant secondary uptake of Mg<sup>2+</sup> has occurred. The olivine dissolution rates are intermediate between those of pure laboratory and field studies and provide a useful constraint on weathering processes in natural environments, such as during soil profile deepening or the addition of mineral dust or volcanic ash to soils surfaces. The dissolution rates also provide critical information for the assessment of enhanced weathering including the expected surface-area and energy requirements.

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## 1. Introduction

Weathering of silicate rocks at the Earth’s surface consumes CO<sub>2</sub> and releases nutrients to fuel the biological cycle. As such, weathering is one of the fundamental geochemical processes shaping the evolution and environment of the planet. On long timescales, silicate weathering provides the ultimate sink for CO<sub>2</sub> released by volcanic degassing and, because the rate of such weathering is temperature dependent, this sink is thought to respond to climate change to provide a strong negative feedback stabilising the Earth’s climate (e.g. Berner and Kothavala, 2001; Walker et al., 1981). As such, silicate weathering is likely to have been the fundamental process that maintained the Earth’s climate within the narrow

bands necessary for life for several billion years. An increase in global weathering rates is expected in response to anthropogenic warming and this increased weathering will ultimately (on the timescale of hundreds of thousands of years) serve to remove CO<sub>2</sub>.

Despite considerable work in recent decades, significant gaps in understanding natural weathering remain. Unpacking the mechanisms that control weathering at a catchment and global scale has proved difficult. Laboratory investigations have shown that dissolution rates ( $W_r$ ) are a function of temperature ( $T$ ; e.g. White et al., 1999), mineral saturation ( $\Omega$ ; e.g. Nagy et al., 1991), pH (e.g. Pokrovsky and Schott, 2000) and mineral surface area (SA e.g. Holdren and Speyer, 1985), and these relationships can be collected into a single, well tested, expression (e.g. Eq. (1)).

$$W_r = SA \cdot k \cdot e^{\frac{E}{RT}} a_{H^+} (1 - \Omega) \quad (1)$$

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where  $k$  is a dissolution rate constant (Palandri and Kharaka, 2004). However, applying this relationship to investigate weathering on a field scale has persistently produced dissolution rates that are several orders of magnitude slower than determined in laboratory experiments (see White and Brantley, 1995 and references therein). It is difficult to isolate the variables in Eq. (1) in the field, and what is measured (through analysis of drainage or soil pore waters) is their combined and attenuated effect. Assumptions regarding the density of the mineral and the depth of the weathering zone (e.g. Calmels et al., 2011) are necessary to convert a spatially explicit rate (e.g. in moles  $\text{km}^{-2} \text{year}^{-1}$ ) into one that is normalised to mineral surface area (e.g. in moles  $\text{cm}^{-2} \text{s}^{-1}$ ). These assumptions can result in large uncertainties in rates. Furthermore, the proportion of the surface area that is actively weathering may change over time as a result of encapsulation in secondary mineral precipitation or formation of a cation depleted/silica rich surface layer (White and Brantley, 2003). The hydraulics and biogeochemistry of soils are complex. Intermittent rainfall, and/or evapotranspiration, may result in soil pore water becoming saturated or oversaturated with minerals. Soil pH may fluctuate as a function of rapidly evolving carbon dioxide partial pressure (see Manning and Renforth (2012) for summary) or organic compound degradation and exudation (particularly low molecular mass organic acids; van Hees et al., 2000). Imprinted over these natural processes may be additional weathering from human activity (e.g. tilling and irrigation, Pačes, 1983). A major challenge for weathering research is the ability to derive meaning in the outputs of short-term highly controlled laboratory studies for complex natural environments.

The lack of understanding of the controls on weathering in the field, and the limited available dissolution kinetic data on minerals intentionally added to the environment, means that it is not possible to predict with any accuracy the rate of chemical weathering (or the fate of resulting solutes) that would result from any intentional manipulation of soil weathering. Such manipulation, sometimes termed, ‘enhanced weathering’ (Hartmann et al., 2013), has been considered as a method of removing  $\text{CO}_2$  from the atmosphere by applying crushed minerals to the land surface (Schuiling and Krijgsman, 2006; Manning, 2008; Renforth, 2012; Köhler et al., 2010; Moosdorf et al., 2014), to the ocean (Harvey, 2008; Köhler et al., 2013; Khesghi, 1995; Renforth et al., 2013), or to coastal zones (Hangx and Spiers, 2009; Schuiling and de Boer, 2010). It is anticipated that the weathering products and sequestered carbon from all of these proposals will be transported to the ocean, which already have a considerable residence time (e.g.  $10^5$  years, see Rau, 2011). The uncertainty in dissolution kinetics is one of the primary reasons why, in recent literature, it has been possible for various workers to suggest that accelerated weathering might be used to remove several billion tonnes (Pg) of  $\text{CO}_2$  from the atmosphere per year (Schuiling and Krijgsman, 2006), while others have suggested that enhanced weathering is limited by mineral saturation and may have restricted use for intentional  $\text{CO}_2$  removal (Hartmann et al., 2013; Köhler et al., 2010). The goal of this study was to test an experimental method that may be used to obtain robust dissolution rate data for minerals intentionally added to soils.

Only a limited number of studies have investigated the dissolution rate of minerals intentionally added to the environment. Peters et al. (2004) investigated changes in stream chemistry due to the addition of 50 tonnes of ground wollastonite ( $\text{CaSiO}_3$ ) onto the Hubbard Brooke catchment, US. The authors noted an increase in the concentration of Ca in the stream draining the catchment, and attributed this to the dissolution of the added mineral. Surface-area-normalised dissolution rates were between  $10^{-15}$  and  $10^{-20}$  moles  $\text{cm}^{-2} \text{s}^{-1}$ . Because the majority of the solution in the Hubbard Brooke watercourse is derived from shallow groundwater, (and lysimeters placed in the soil showed little contribution

of the distributed wollastonite to the groundwater), it is thought that the chemical response to the addition is a result of dissolution of the material that fell initially into the watercourse (about 1.5% of the total). Manning et al. (2013) were able to calculate a single minimum dissolution rate of  $10^{-16}$  moles  $\text{cm}^{-2} \text{s}^{-1}$  for crushed dolerite that was weathered in an artificial soil created using quarry fines mixed with organic materials (food industry waste and compost). They inferred a mass balance of calcium from mineral carbonate formation; the actual dissolution rate is therefore poorly constrained. In a growth experiment (closed soil system) ten Berge et al. (2012) measured the change in water soluble Mg to determine a dissolution rate of olivine between  $10^{-14}$  and  $10^{-16}$  moles  $\text{cm}^{-2} \text{s}^{-1}$ . While this approach provided a dissolution rate, it did not fully quantify the magnesium efflux from the soil, and potentially over estimates the contribution to carbon sequestration. There is a need to develop an experimental approach capable of constraining a budget for weathering products while maintaining the complexity of soils.

## 2. Overview of experimental design

In this study, the complexity of the natural environment was brought into the controlled laboratory by extracting soil cores from arable agricultural land and establishing them as lysimeters in the laboratory (Lundström, 1990; Sigfusson et al., 2006). Crushed and ground olivine was added to the top of one soil core, and a nutrient solution was drip-fed into the top and collected at the base. This approach allows complex soil processes to be replicated in the laboratory, while providing enough control to develop a closed budget for olivine dissolution without fully deconstructing the effect of complex soil biogeochemistry. By comparing the effluent solutions from the olivine-modified core with those from a control core, we account for the background signal and assess the dissolution rate of the added olivine.

## 3. Experimental methods

### 3.1. Site description and soil core extraction

In May 2012, 3 soil cores (contained in an acrylic plastic tube  $0.1 \times 1.0 \text{ m}$ ) were extracted from an agricultural field in North Oxfordshire using a premier Compact 110 percussion window sampler (Perdiswell Farm;  $1^\circ 19' 50.03'' \text{W}$ ,  $51^\circ 51' 27.55'' \text{N}$ , Fig. 1A). The location at the time was used for growing *Vicia faba* (broad bean), and had previously been used for a range of arable crops (e.g. *Triticum* sp., wheat). The soil overlies Jurassic limestone and mudstone from the Great Oolite Group, and has been in agricultural use for over 100 years. The soils were calcareous with a thin (10–15 cm) organic rich plough layer, underlain by weakly differentiated subsoil and parent material (Fig. 1B and C) to 1 m depth.

### 3.2. Soil chemical analysis

Soil was removed at 5 cm depth intervals from one of the cores, oven dried for 24 h (the moisture content assessed by mass loss during drying), and the fine fraction (<2 mm) was separated. XRF analysis was conducted using a PANalytical Axios Advanced XRF spectrometer for major elements (University of Leicester) calibrated using BCS375, BCS376, and BCS372/1 standards. To analyse the elemental composition of the exchanged cations, carbonate phases and bulk soil respectively, steps 1 (1 M Sodium Acetate), 2 (1 M Acetic Acid) and 5 (concentrated HF) of a sequential leach were performed using the method outlined in Tessier et al. (1979). For organic (TOC) and inorganic (TIC) carbon determination, 2 aliquots of each sample (one of which was ashed at  $450^\circ \text{C}$  overnight to remove organic carbon) were introduced into

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