



# The efficient long-term inhibition of forsterite dissolution by common soil bacteria and fungi at Earth surface conditions

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## Abstract

San Carlos forsterite was dissolved in initially pure H<sub>2</sub>O in a batch reactor in contact with the atmosphere for 5 years. The reactive fluid aqueous pH remained relatively stable at pH 6.7 throughout the experiment. Aqueous Mg concentration maximized after approximately 2 years time at  $3 \times 10^{-5}$  mol/kg, whereas aqueous Si concentrations increased continuously with time, reaching  $2 \times 10^{-5}$  mol/kg after 5 years. Element release rates closely matched those determined on this same forsterite sample during short-term abiotic open system experiments for the first 10 days, then slowed substantially such that the Mg and Si release rates are approximately an order of magnitude slower than that calculated from the short-term abiotic experiments. Post-experiment analysis reveals that secondary hematite, a substantial biotic community, and minor amorphous silica formed on the dissolving forsterite during the experiment. The biotic community included bacteria, dominated by *Rhizobiales* (*Alphaproteobacteria*), and fungi, dominated by *Trichocomaceae*, that grew in a carbon and nutrient-limited media on the dissolving forsterite. The Mg isotope composition of the reactive fluid was near constant after 2 years but 0.25‰ heavier in  $\delta^{26}\text{Mg}$  than the dissolving forsterite. Together these results suggest long-term forsterite dissolution in natural Earth surface systems maybe substantially slower than that estimated from short-term abiotic experiments due to the growth of biotic communities on their surfaces.

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## 1. INTRODUCTION

The goal of this study is to improve our understanding of forsterite reactivity in natural Earth surface systems. Towards this goal, forsteritic olivine was dissolved in initially pure H<sub>2</sub>O in a batch reactor for 5 years. Although no carbon or nutrients were added to the reactor, a community of microbes developed on the dissolving forsterite

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during the experiment. The temporal evolution of the reactive fluid chemistry and post-experiment analysis of the solid phases, including DNA sequencing, were used to provide insight into microbial-mineral interaction and to assess the degree to which this microbial community affected forsterite reactivity. Magnesium isotopic analyses were performed to determine how microbially mediated forsterite dissolution fractionates this element between the solid and fluid phase. The purpose of this communication is to report the results of this experimental study, providing insight into both the ability to harvest divalent metal cations from forsterite for mineral carbonation, and how microbial communities may affect weathering rates over extended time frames at Earth surface conditions.

The reactivity of forsterite and its silicate alteration products has received increasing attention as a potential source material for the divalent cations required to carbonate CO<sub>2</sub> during carbon storage efforts (e.g. Giammar et al., 2005; Oelkers and Schott, 2005; Bearat et al., 2006; Matter et al., 2007; Oelkers et al., 2008; Dufaud et al., 2009; Matter and Kelemen, 2009; Prigiobbe et al., 2009; King et al., 2010; Daval et al., 2011; Guyot et al., 2011; Beinlich et al., 2012; Broecker, 2012; Kohler et al., 2013; Gislason and Oelkers, 2014; Sissmann et al., 2014). This interest has led to a large number of studies aimed at characterizing forsterite dissolution behavior and rates at various fluid compositions and temperatures (Luce et al., 1972; Sanemasa et al., 1972; Grandstaff, 1978, 1986; Murphy and Helgeson, 1987, 1989; Blum and Lasaga, 1988; Banfield et al., 1990; Wogelius and Walther, 1991, 1992; Casey and Westrich, 1992; Awad et al., 2000; Chen and Brantley, 2000; Rosso and Rimstidt, 2000; Pokrovsky and Schott, 2000a,b; Oelkers, 2001b; Giammar et al., 2005; Hänchen et al., 2006; Olsen and Rimstidt, 2008; Davis et al., 2009; Beinlich and Austrheim, 2012; Rimstidt et al., 2012; Olsson et al., 2012; Plümper et al., 2012; Wang and Giammar, 2012; Garcia et al., 2013; Saldi et al., 2013; van Noort et al., 2013; Bundeleva et al., 2014; Declercq et al., 2013; Johnson et al., 2014; King et al., 2011, 2014; Martinez et al., 2014; Torres et al., 2014). A large fraction of these forsterite dissolution experiments were performed at acidic conditions, either due to the presence of aqueous HCl (e.g. Casey and Westrich, 1992; Oelkers, 2001b) or at elevated CO<sub>2</sub> pressures (e.g. Saldi et al., 2013; Johnson et al., 2014). More significantly, the bulk of these experiments were performed over short time periods; for example Olsen and Rimstidt (2008) reported forsterite dissolution rates based on experiments performed for a total of only two hours each.

A number of studies, however, suggest that the long-term dissolution behavior of dissolving olivine may be significantly different from its initial short-term behavior. Mg was observed to be preferentially released relative to Si from the forsterite surface during the initial stages of its dissolution at acidic pH (Seyama et al., 1996; Pokrovsky and Schott, 2000a,b; Zakaznova-Herzog et al., 2008; Oelkers et al., 2009; King et al., 2011). This behavior can result in the formation of a Si-enriched surface layer that may polymerize and influence dissolution rates; Daval et al. (2011) reported that forsterite dissolution rates decrease as acidic CO<sub>2</sub>-rich fluids become saturated with

respect to amorphous silica, similar to the behavior previously observed for multi-component mineral and glass dissolution (e.g. Grambow, 1985; Grambow and Muller, 2001; Oelkers, 2001a). Alternatively, the surface topography of the mineral itself can evolve due to dissolution altering its average reactivity (e.g. Bandstra and Brantley, 2008; Luttge et al., 2013). In addition, the dissolving olivine surface can become covered by secondary minerals and/or bacteria (c.f. Giammar et al., 2005; Zakaznova-Herzog et al., 2008; Olsson et al., 2012; Wang and Giammar, 2012; Hövelmann et al., 2012; Shirokova et al., 2012; Saldi et al., 2013). The degree to which secondary surface precipitates affect the dissolution rates of the primary mineral appears to depend on the structural match between the two minerals and the presence of interconnected porous pathways in the secondary phases (Hodson, 2003; Cubillas et al., 2005; Putnis, 2009; Stockmann et al., 2011, 2013; Saldi et al., 2013). Finally, long-term olivine dissolution rates can also be limited by the slow precipitation rates of secondary minerals, which could lead the fluid to approach equilibrium with the dissolving mineral (e.g. Zhu and Lu, 2009; Zhu et al., 2010; Saldi et al., 2012). This study was motivated in part to assess how these various processes might influence forsterite dissolution rates over time-scales larger than those typically considered in laboratory studies.

A number of studies demonstrated that processes such as secondary mineral formation, adsorption and/or uptake by higher plants occurring during weathering may alter the Mg isotopic composition of residual fluids (e.g. Black et al., 2008; Pogge von Strandmann et al., 2008; Bolou-Bi et al., 2010; Li et al., 2010; Teng et al., 2010; Tipper et al., 2010; Wimpenny et al., 2010; Opfergelt et al., 2012, 2014; Shirokova et al., 2013; Huang et al., 2012; Ilina et al., 2013; Mavromatis et al., 2012, 2014). Such processes could lead to global riverine fluxes that are isotopically lighter in Mg compared to the homogenous chondritic composition of the Earth's mantle (Tipper et al., 2006). Conversely, the formation of secondary carbonate minerals is known to preferentially remove isotopically light Mg isotopes from solution, causing the residual liquid to have a heavier composition (Mavromatis et al., 2014; Beinlich et al., 2014). Consequently isotopic analysis of the reactive fluids during sampled during our experiments may help to quantify whether Mg isotope fractionation occurs during forsterite dissolution, and what impact this may have on the composition of fluids at the Earth's surface.

## 2. MATERIALS AND METHODS

### 2.1. Batch reactor experiment

This study reports on the result of a single 5-year forsterite dissolution experiment performed in a batch reactor system. The San Carlos forsterite sample used in this study was originally prepared by Pokrovsky and Schott (2000a,b), who reported that its composition is consistent with Mg<sub>1.82</sub>Fe<sub>0.18</sub>SiO<sub>4</sub> (F<sub>0.91</sub>). San Carlos Olivine has been reported to contain numerous trace elements including ~40 ppm P (De Hoog et al., 2010). Transparent crystals

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