



Fate of transition metals during passive carbonation of ultramafic mine tailings via air capture with potential for metal resource recovery



Jessica L. Hamilton^{a,b,*}, Siobhan A. Wilson^{a,c}, Bree Morgan^{a,d}, Connor C. Turvey^a, David J. Paterson^e, Simon M. Jowitt^{a,f}, Jenine McCutcheon^{b,g}, Gordon Southam^b

^a School of Earth, Atmosphere and Environment, Monash University, Clayton, Melbourne, VIC 3800, Australia

^b School of Earth and Environmental Sciences, The University of Queensland, St Lucia, QLD 4072, Australia

^c Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, AB T6G 2R3, Canada

^d School of Geosciences, The University of Sydney, Camperdown, NSW 2006, Australia

^e Australian Synchrotron, Clayton, Melbourne, VIC 3168, Australia

^f Department of Geoscience, University of Nevada, Las Vegas, 4505 S. Maryland Pkwy, Las Vegas, NV 89154, USA

^g School of Earth and Environment, Maths/Earth and Environment Building, University of Leeds, Leeds, LS2 9JT, United Kingdom

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ABSTRACT

Mineral carbonation in ultramafic mine tailings is generally accepted to be a safe and long term means of trapping and storing CO₂ within the structures of minerals, but it poses the risk of releasing potentially hazardous metal contaminants from mineral wastes into the environment. Stockpiles of reactive, finely pulverised ultramafic mine tailings are ideal natural laboratories for the observation and promotion of the carbonation of Mg-silicate and Mg-hydroxide waste minerals via reaction with atmospheric or industrial CO₂. However, ultramafic mine tailings commonly contain first-row transition metals (e.g., Cr, Co, Cu, Ni) in potentially toxic concentrations within the crystal structures of Mg-silicates, sulphides, and oxides. These transition metals are likely to be mobilised by mineral carbonation reactions, which require mineral dissolution to supply cations for reaction with carbon. At Woodsreef Chrysotile Mine, New South Wales, Australia, transition metals (i.e., Fe, Cr, Ni, Mn, Co, Cu) are most concentrated within minor oxides (magnetite and chromite) and trace alloys (awaruite, Ni₂₋₃Fe and wairuite, CoFe) in serpentine tailings, however, mobilisation of transition metals appears to occur predominantly during dissolution of serpentine and brucite, which are more abundant and reactive phases, respectively. Here, we present new synchrotron X-ray fluorescence mapping data that provide insights into the mobility of first-row transition metals (Fe, Cr, Ni, Mn, Co, Cu) during weathering and carbonation of ultramafic mine tailings collected from the Woodsreef Chrysotile Mine. These data indicate that the recently precipitated carbonate minerals, hydromagnesite [Mg₅(CO₃)₄(OH)₂·4H₂O] and pyroaurite [Mg₆Fe₂(CO₃)(OH)₁₆·4H₂O] sequester trace metals from the tailings at concentrations of 10 s–100 s of ppm, most likely via substitution for Mg or Fe within their crystal structures, or by the physical trapping of small (µm-scale) transition-metal-rich grains (i.e., magnetite, chromite, awaruite), which are stabilised within alkaline carbonate cements. Trace transition metals are present at relatively high concentrations in the bulk tailings (i.e., ~0.3 wt.% NiO and Cr₂O₃) and they are largely retained within the unaltered mineral assemblage. The weathering products that occur at the surface of the tailings and form a cement between grains of partially dissolved gangue minerals immobilise transition metals on spatial scales of micrometres and at comparable concentrations to those observed in the unaltered tailings. The end result is that trace metals are not present at detectable levels within mine pit waters. Our observations of metal mobility during passive carbonation suggest that mineral products of accelerated carbonation treatments are likely to sequester trace metals. Thus, accelerated carbonation is unlikely to pose an environmental risk in the form of metalliferous drainage so long as the neutralisation potential of the tailings is not exceeded.

Understanding both trace transition metal geochemistry and mineralogy within materials targeted for mineral carbonation could allow optimisation of treatment processes and design for recovery of valuable metals. In *ex situ* reactors employing acid pre-treatments, trace metals mobilised from reactive phases such as serpentine and brucite could potentially be recovered using pH-swing methods, while recalcitrant metal-rich accessory minerals, including magnetite, awaruite and chromite, could be recovered from treated residue material by conventional mineral separation processes. Recovery of valuable metals (i.e., Ni, Cr and Co) as by-products of

* Corresponding author at: School of Earth and Environmental Sciences, The University of Queensland, St Lucia, QLD 4072, Australia.
E-mail address: j.hamilton@uq.edu.au (J.L. Hamilton).

accelerated mineral carbonation technologies could also provide an important economic incentive to support broader adoption of this technology.

1. Introduction

The increasing concentration of anthropogenic CO₂ in the atmosphere is likely driving long-term changes to Earth's climate (Hallett, 2002; IPCC, 2005, 2013). As such, there is a growing need to develop mitigation technologies, both to reduce anthropogenic CO₂ emissions and to capture atmospheric CO₂ from the air for long-term storage in a stable form. Mineral carbonation, which was first proposed as a mitigation strategy for CO₂ emissions by Seifritz (1990), captures CO₂ within the structures of environmentally benign carbonate minerals by enhancing natural silicate weathering, promoting the safe, long-term storage of CO₂. Importantly, this is the only proposed CO₂ storage technology that is considered to be permanent over geological timescales, given that many carbonate minerals are known to persist at the Earth's surface for 1000 s of years or longer (Lackner et al., 1995; Oelkers et al., 2008; Olajire, 2013; Power et al., 2009; Seifritz, 1990).

Natural carbonation of mine tailings in Australia, Canada, the U.S.A., and Norway (99–6200 g CO₂/m²/y; Beinlich and Austrheim, 2012; Gras et al., 2017; Lechat et al., 2016; Levitan et al., 2009; Oskierski et al., 2013; Pronost et al., 2012; Turvey et al., 2017; Wilson et al., 2006, 2009a,b, 2011, 2014) occurs at rates that are typically orders of magnitude greater than average CO₂ uptake associated with silicate weathering (0.7–62.1 g CO₂/m²/y, Power et al., 2013a,b, units converted from Ludwig et al., 1998). The enhanced reactivity of mine tailings is largely due to the orders of magnitude increase in mineral surface area that results from crushing during ore processing (Wilson et al., 2009a). The greatest uptake of CO₂ by mine tailings yet reported occurs at the Mt Keith Nickel Mine in Western Australia, where nearly 40,000 t of atmospheric CO₂ are sequestered annually via natural reaction of the tailings with the atmosphere, representing an 11% offset of the mine's annual greenhouse gas emissions (Wilson et al., 2014). This enhanced reactivity of mineral wastes makes historical and operating mine tailings storage facilities provide ideal settings for observing transition metal mobility during passive mineral carbonation reactions on timescales of years to decades.

Ultramafic rocks are commonly mined for Cr, Ni-Cu-platinum group element sulphides, diamonds, talc, and (historically) chrysotile asbestos. They are also the preferred feedstock material for mineral carbonation because they contain high abundances of silicate minerals with high Mg contents {i.e., olivine [(Mg,Fe)₂SiO₄] and serpentine [Mg₃Si₂O₅(OH)₄]}, and minor amounts of highly reactive phases such as brucite [Mg(OH)₂] (Goff and Lackner, 1998; Lackner, 2003; Oelkers et al., 2008; Power et al., 2013a). Brucite forms via the hydration of olivine during serpentinisation and is usually present as a common but minor component of ultramafic tailings (1–15 wt.%; Harrison et al., 2012; O'Hanley and Offler, 1992). The lack of strong Si–O bonds within brucite means that it has a reactivity that is orders of magnitude greater than that of Mg-silicates, meaning that the abundance of brucite exerts strong control on mineral carbonation rates under ambient conditions (Harrison et al., 2012; Power et al., 2013a). In addition, the serpentine polymorphs, chrysotile and lizardite, consist of a sheet of silica tetrahedra bonded to an octahedral Mg–OH sheet that is commonly referred to as the 'brucite-like' layer (e.g., Wicks and O'Hanley, 1988). This 'brucite-like' layer is more reactive and dissolves faster than the accompanying Si-rich tetrahedral layer (Park and Fan, 2004; Rozalen and Huertas, 2013), increasing the reactivity of serpentine and enhancing release of Mg for CO₂ sequestration. Dissolution of these minerals in meteoric water (containing dissolved atmospheric CO₂ as carbonic acid, pH ~5.5) releases Mg²⁺ cations and increases alkalinity. Mg²⁺ cations in solution subsequently react with dissolved CO₃²⁻ to precipitate as

hydrated carbonate minerals such as hydromagnesite [Mg₅(CO₃)₄(OH)₂·4H₂O] and nesquehonite (MgCO₃·3H₂O), with Mg:CO₂ ratios and hydration states that are dependent on local environmental conditions (Ballirano et al., 2013; Davies and Bubela, 1973; Hänchen et al., 2008; Morgan et al., 2015; Wilson et al., 2009a). Since carbonic acid in meteoric water is a weak acid, acid leaching, or the addition of acid generating materials, has been widely employed to accelerate CO₂ uptake in ultramafic materials because it increases the rate of silicate mineral dissolution and the availability of Mg²⁺ cations for reaction (e.g., Maroto-Valer et al., 2005; Park and Fan, 2004; Power et al., 2010; Teir et al., 2007b). Following acid leaching, pH-swing methods can then be used to increase pH to alkaline conditions to allow precipitation of carbonate minerals (Park and Fan, 2004). In highly reactive mineral wastes, particularly where brucite is present, mineral dissolution under ambient conditions can provide sufficient alkalinity to induce carbonate precipitation (Harrison et al., 2012; Power et al., 2013a). However, ultramafic rocks commonly contain several wt.% Fe as well as low concentrations (< 1 wt.%) of other first row transition metals (e.g., Ni, Mn, Cr, Co, Cu). These are typically found within the crystal structures of Mg-silicate, Mg-hydroxide, sulphide, and oxide minerals as well as within alloy minerals such as awaruite (Ni₂₋₃Fe) and wairauite (CoFe) (Challis and Long, 1964; Goff and Lackner, 1998; Kmetoni, 1984; Margiotta et al., 2012; Natali et al., 2013; Schreier, 1987; Sciortino et al., 2015). First row transition metals may be mobilised by dissolution of their mineral hosts during either natural weathering reactions or the artificially accelerated reactions that can be used to enhance mineral carbonation rates (Margiotta et al., 2012; Oelkers et al., 2008; Olajire, 2013; Olsson et al., 2014a,b; Schreier, 1987; Teir et al., 2007a). Because first-row transition metals can be toxic to biota in elevated concentrations, concern has been raised that the release of metalliferous waters from mineral carbonation facilities or geoengineered landscapes could adversely affect natural systems (Olsson et al., 2014b). Consequently, the potential mobility of trace metals during carbonation reactions is an essential consideration for implementation of *ex situ* mineral carbonation technologies, *in situ* enhanced weathering of ultramafic landscapes or mineral wastes, and *in situ* CO₂ injection into rock formations (Hamilton et al., 2016; Kirsch et al., 2014; Marcon and Kaszuba, 2013, 2015; Oelkers et al., 2008; Olajire, 2013; Olsson et al., 2014a; Olsson et al., 2014b; Seal et al., 2010; Thomas et al., 2013; Thomas et al., 2016).

Previous studies have reported elevated concentrations of trace metals (particularly Ni, Cr³⁺ and carcinogenic Cr⁶⁺) in soils and waters associated with serpentinites (Margiotta et al., 2012; McClain and Maher, 2016; Morrison et al., 2015; Schreier, 1987; Schreier and Lavkulich, 2015). Uptake of these trace metals by Fe-hydroxide and clay minerals has been documented in serpentinites (Morrison et al., 2015), but the influence of hydrated Mg-carbonate minerals on trace metal mobility within ultramafic environments is relatively unexplored. This is a critical knowledge gap, given that hydrated Mg-carbonate minerals are commonly present in high abundances [up to 14.6 wt.% hydromagnesite; (Oskierski et al., 2013)] in passively carbonated mine tailings, and therefore may provide an important sink for potentially toxic trace metals.

Hamilton et al. (2016) demonstrated that Mg-carbonate minerals and Fe-oxyhydroxide phases sequester transition metals during their formation in laboratory experiments designed to emulate the Mg- and transition-metal-rich conditions produced during accelerated mineral carbonation. However, the mobility of potentially toxic trace metals during naturally occurring mineral carbonation in ultramafic landscapes has not yet been extensively explored. Here, we investigate (1)

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