



# Offsetting of CO<sub>2</sub> emissions by air capture in mine tailings at the Mount Keith Nickel Mine, Western Australia: Rates, controls and prospects for carbon neutral mining



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## ABSTRACT

The hydrated Mg-carbonate mineral, hydromagnesite [ $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$ ], precipitates within mine tailings at the Mount Keith Nickel Mine, Western Australia as a direct result of mining operations. We have used quantitative mineralogical data and  $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$  and  $\text{F}^{14}\text{C}$  isotopic data to quantify the amount of CO<sub>2</sub> fixation and identify carbon sources. Our radiocarbon results indicate that at least 80% of carbon stored in hydromagnesite has been captured from the modern atmosphere. Stable isotopic results indicate that dissolution of atmospheric CO<sub>2</sub> into mine tailings water is kinetically limited, which suggests that the current rate of carbon mineralization could be accelerated. Reactive transport modeling is used to describe the observed variation in tailings mineralogy and to estimate rates of CO<sub>2</sub> fixation. Based on our assessment, approximately 39,800 t/yr of atmospheric CO<sub>2</sub> are being trapped and stored in tailings at Mount Keith. This represents an offsetting of approximately 11% of the mine's annual greenhouse gas emissions. Thus, passive sequestration via enhanced weathering of mineral waste can capture and store a significant amount of CO<sub>2</sub>. Recommendations are made for changes to tailings management and ore processing practices that have potential to accelerate carbonation of tailings and further reduce or completely offset the net greenhouse gas emissions at Mount Keith and many other mines.

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## 1. Introduction

Storage of CO<sub>2</sub> in carbonate minerals has been recognized as a safe and effective method for mitigating rising concentrations of atmospheric greenhouse gases (Seifritz, 1990; Lackner et al., 1995; Lackner, 2003). Unlike traditional Carbon Capture and Storage (CCS), which relies predominantly upon stratigraphic trapping of injected supercritical CO<sub>2</sub> within rock formations, carbon mineralization technologies utilize direct production of carbonate minerals as traps for CO<sub>2</sub>. Since carbon mineralization was first proposed as a method for storing CO<sub>2</sub> (Seifritz, 1990), most of the work

on this subject has focused on the development of rapid, large-scale ex situ methods for trapping and storing CO<sub>2</sub> at industrial point sources (reviewed in Huijgen and Comans, 2003, 2005; IPCC, 2005; Sipilä et al., 2008; Power et al., 2013a). Most processes developed to date are based on reaction of CO<sub>2</sub> with naturally occurring non-carbonate minerals such as silicates, hydroxides or oxides. Weathering of these minerals in nature is generally a slow process and high temperatures and pressures are needed to induce carbon mineralization reactions on the short timescales (i.e., hours) required for development and deployment of industrial carbonation reactors (Sipilä et al., 2008; Zevenhoven et al., 2011). Although rapid and technologically feasible, the financial viability of this approach to carbon mineralization is currently limited by low carbon prices (Power et al., 2013a,b).

Recent research has focused increasingly on two alternative approaches: (1) in situ carbon mineralization and (2) ex situ carbon

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mineralization at industrial sites. In situ carbon mineralization aims to promote subsurface carbonation by injection of CO<sub>2</sub>, or solutions bearing dissolved CO<sub>2</sub>, into mafic and ultramafic formations (e.g., Cipolli et al., 2004; Kelemen and Matter, 2008; Gislason et al., 2010). Ex situ carbon mineralization at industrial sites focuses on low-temperature and low-pressure carbonation of alkaline industrial wastes such as smelter slag, fly ash, alkaline and saline brine, construction waste, and mine tailings (e.g., Wilson et al., 2006, 2009a,b, 2010, 2011; Manning, 2008; Ferrini et al., 2009; Huntzinger et al., 2009; Power et al., 2009, 2010, 2011a; Renforth et al., 2009, 2011; Ballirano et al., 2010; Zhao et al., 2010; Pronost et al., 2011, 2012; Bobicki et al., 2012; Harrison et al., 2013; Manning and Renforth, 2013).

Carbonation of the mineral waste from ultramafic mines has been documented previously (Wilson et al., 2006, 2009a,b, 2011; Pronost et al., 2012; Beinlich and Austrheim, 2012; Oskierski et al., 2013); however, previous estimates of carbonation rates have generally relied on small sample sets and have not been explained within the context of geochemical modeling. Complementary laboratory experiments and reactive transport modeling have recently been developed to investigate and quantify controls on the rate of carbon mineralization in mine tailings (e.g., Wilson et al., 2010; Pronost et al., 2011; Bea et al., 2012; Harrison et al., 2013; Assima et al., 2013a,b). Thus, a detailed framework now exists for assessing, monitoring and modeling carbon mineralization in mine tailings. Here, for the first time, we apply isotopic and crystallographic carbon accounting methods and reactive transport modeling to quantitatively assess the rate of, and controls on, carbon mineralization on the scale of a large operating tailings facility at the Mount Keith Nickel Mine, Western Australia. This represents the first implementation of a detailed framework for carbon accounting during carbon mineralization via air capture in an active industrial setting and on a landscape scale.

At the Mount Keith Nickel Mine, the hydrated Mg-carbonate mineral, hydromagnesite [Mg<sub>5</sub>(CO<sub>3</sub>)<sub>4</sub>(OH)<sub>2</sub>·4H<sub>2</sub>O], develops as a weathering product within ultramafic mine tailings. Consequently, accounting of the amount of atmospheric CO<sub>2</sub> that is being trapped and stored within this mineral could be used to offset the mine's greenhouse gas emissions. The isotope system (i.e., δ<sup>13</sup>C, δ<sup>18</sup>O, and F<sup>14</sup>C) employed by Wilson et al. (2009a) has been used to assess capture and storage of atmospheric CO<sub>2</sub> within hydromagnesite in the tailings at Mount Keith. Further to this, quantitative phase analysis using the Rietveld method and powder X-ray diffraction (XRD) data are used to estimate the amount of hydromagnesite in the tailings. Quantification of hydromagnesite at Mount Keith is non-trivial, because the mineral content of mine tailings is generally heterogeneous due to frequent changes in ore mineralogy, processing procedures, and tailings management practices. The mineralogy of tailings piles also changes with time as tailings react with the atmosphere and meteoric and process waters to produce secondary mineral phases. As a result, geostatistical methods that rely on the presence of regular and predictable geological structures cannot be used to predict the mineralogy of a mine tailings pile. Thus, in order to estimate the total amount of CO<sub>2</sub> captured from the atmosphere and stored within secondary carbonate mineral phases at Mount Keith, it is necessary that the mineralogy of its tailings storage facilities be well constrained. We have constructed a database of quantitative mineralogical results for a large suite of tailings samples, taken from various depths below the surface of the tailings storage facility at Mount Keith.

Because the ages are known for many of the tailings flows at Mount Keith, we have been able to use our quantitative mineralogical results to build a time-dependent reactive transport model that describes the geochemical evolution of its tailings storage facilities (Bea et al., 2012; this study). From this model, we illuminate the mechanisms governing carbon mineralization and obtain an

empirical rate for hydromagnesite precipitation. Finally, the reactive transport model has been used to assess potential methods by which tailings management practices could be tailored to enhance carbon mineralization and maximize offsetting of greenhouse gas emissions at Mount Keith.

## 2. Locality and sampling strategy

### 2.1. The Mount Keith Nickel Mine

The MKD5 orebody at Mount Keith is located in the North Eastern Goldfields district of Western Australia (Fig. 1) and is the largest nickel producer in Australia (Grguric, 2003). The deposit at Mount Keith occurs in the NNW/SSE-trending Agnew–Wiluna greenstone belt in the Archean Yilgarn Craton (Hill et al., 1990). The MKD5 orebody is hosted by komatiitic peridotite (primarily dunite), which attained mid–upper greenschist facies as a result of regional metamorphism (Barrett et al., 1977). Retrograde serpentinization and carbonation of the host peridotites resulted from infiltration by H<sub>2</sub>O–CO<sub>2</sub>-rich fluids (Barrett et al., 1977; Grguric et al., 2006). Resulting metamorphic assemblages (from proximal to distal) are (1) talc–magnesite, (2) antigorite–magnesite, and (3) lizardite–brucite–hydrotalcite group (Grguric et al., 2006).

Conventional, staged-cutback, open pit mining methods are practiced at MKD5, yielding approximately 11 Mt of ore annually (Grguric, 2003). The mining operation at MKD5 produces approximately 370,000 t of greenhouse gases (cited as CO<sub>2</sub> equivalent) and approximately 11 Mt of ultramafic tailings each year (BHP Billiton, 2005). Ore from the MKD5 deposit is processed using froth flotation methods to concentrate sulfide minerals (Grguric et al., 2006). Additives used in processing include citric and sulfuric acids, guar gum, Na-dithionite, Na-ethyl xanthate, and (historically) soda ash. In 2004, ore reserves contained 0.52 wt.% nickel (Grguric et al., 2006), primarily in high-Ni pentlandite [(Fe,Ni)<sub>9</sub>S<sub>8</sub>], godlevskite [(Ni,Fe)<sub>9</sub>S<sub>8</sub>], heazlewoodite [Ni<sub>3</sub>S<sub>2</sub>], and millerite [NiS]. Recovery of these minerals from the flotation circuit is typically about 70% (Grguric et al., 2006). The material rejected from the processing plant is piped to one of two tailings storage facilities (TSF2, which was the only facility in operation at the time of sampling), and is suspended in the hypersaline process water used in the flotation circuit. Tailings are deposited from spigots located on risers at nine points in TSF2 (Fig. 1a).

The tailings storage facility at Mount Keith was constructed in two phases. The two cells of TSF1 (Fig. 1a) were the sole receptacles for tailings from July 1994 until the facility was decommissioned in January 1997 (Stolberg, 2005). A centralized discharge tailings storage facility (TSF2) was commissioned to replace TSF1 by January 1997 (Stolberg, 2005) and remains in operation today. Along the circumference of TSF2, the outer 100–400 m of the compound are dedicated to catching and storing tailings in the event that mineral waste from the interior of the facility should overflow. This design feature has been effective and provides snapshots of tailings compositions from several overflow events. Tailings in the various overflow cells of TSF2 had been permitted to weather, without addition of new tailings or process water, for periods of one, three, and between seven and eight years prior to sampling in 2006. At the time of sampling, tailings in the interior of TSF2 (along the radial access road, Fig. 1a) had last been deposited approximately zero or one half years prior to collection.

### 2.2. Strategy for sampling at Mount Keith

Limited sampling of the tailings storage facilities at Mount Keith was begun in April 2005. More extensive sampling was done in September and October of 2006. In excess of 800 samples were

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