

Geochemical modelling of CO₂ sequestration in ultramafic mine wastes from Australia, Canada, and South Africa

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Passive carbonation in ultramafic mine wastes results from the spontaneous reaction of minerals with atmospheric CO₂ to form stable carbonates that sequester CO₂. Rates of passive carbonation have previously been determined using quantitative X-ray diffraction (QXRD) analysis of numerous samples of mine wastes, which yields valuable information about cation sources and carbon sinks [1,2]. However, these extensive mineralogical assessments are technically demanding and may be cost-prohibitive for routine monitoring of CO₂ sequestration. Here, we propose using mass-balance geochemical models (inverse modelling) as an alternative for determining passive carbonation rates. Water chemistry data, tailings mineralogy, and operational information are routinely collected by mines and were used as inputs for models. The predictive capabilities of these models were tested with data from the Mount Keith nickel mine (Australia) and Diavik diamond mine (Canada), where carbonation rates were previously determined using QXRD [1,2]. Inverse modelling of Mount Keith waters indicated that carbonation of brucite [Mg(OH)₂] to form hydromagnesite [Mg₅(CO₃)₄(OH)₂·4H₂O] is the dominant reaction in tailings [1]. A CO₂ removal rate of 3,800 g CO₂/m²/yr was due to 40% brucite carbonation in the tailings. These findings are consistent with those of Wilson et al. (2014), who reported 45% brucite carbonation and a rate of 2,400 g CO₂/m²/yr. Modelling of Diavik waters predicted that nesquehonite [MgCO₃·3H₂O] precipitation results in a CO₂ removal rate of ~300 g CO₂/m²/yr versus 400 g CO₂/m²/yr determined with QXRD [2]. Modelling of mine waters from the Venetia diamond mine (South Africa; 2015–18) demonstrates how passive carbonation is affected by mineral processing, seasonality, and mineral-water reactions. Moreover, the modelled passive carbonation rate of ~200 g CO₂/m²/yr corroborates recent field experiments. Our study establishes a foundation to develop further geochemical models for routine monitoring of passive carbonation at active mines, which can assist in decision-making on implementing enhanced CO₂ sequestration strategies. In addition, access to more complete data, including additional sampling locations (e.g., tailings porewaters) and parameters (e.g., Si) will improve our models.

[1] Wilson et al. (2014) *Int. J. Greenh. Gas Control*. 25, 121-140. [2] Wilson et al. (2011) *Environ. Sci. Technol.* 45, 7727-7736.