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Atmospheric CO_2 removal rates using magnesium oxide powder

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Magnesium oxide (MgO) looping is a proposed negative emissions technology with the potential for gigatonne-scale CO_2 removal. Magnesite ($MgCO_3$) is calcined to MgO with high purity CO_2 being stored and the regenerated MgO being repeatedly used to capture CO_2 [1,2]. However, this technology's technical and economic feasibility depends on largely unknown carbonation rates. Therefore, the goal of this study was to determine carbonation rates in a laboratory-controlled environment. An MgO powder deposit (10 cm thick; 76 kg/m^2) was tilled daily and exposed to wetting and drying cycles to simulate weathering over 5 months. CO_2 removal rates were independently determined by measuring CO_2 fluxes and total inorganic carbon (TIC). The MgO powder hydrated (80 wt%) to form brucite [$Mg(OH_2)$]. CO_2 fluxes were mainly depended on water content and porosity: -3.9 (saturated/wet), -12.5 (optimal), and -2.2 (dry) kg CO_2/m^2/yr. Optimal conditions for CO_2 removal were when the deposit had a porosity of 44–61% and water content of 7–17 wt%. Dypingite [$Mg_5(CO_3)_4(OH)_2{\sim}5H_2O$] was the sink of atmospheric CO_2, and TIC increased from 0.2–3.8% CO_2. The $\delta^{13}C$ values of the solids (avg. -14.9‰ VPDB) were well below those expected for dypingite precipitated in equilibrium with atmospheric CO_2, demonstrating that CO_2 supply was rate-limiting, despite the daily mixing of the deposit. A linear extrapolation of the 5-month rates shows that only 3–18% of the MgO would react in 1 yr and require areas of 80,000–455,000 km^2 to sequester 1 Gt CO_2/yr. At these rates, 90% carbonation of MgO would require 5–27 yr. However, this linear extrapolation likely underestimates the time needed for complete carbonation as it does not account for the progressive depletion of reactants (MgO/brucite) that will cause rates to slow. Continued monitoring of the MgO deposit over 1 yr will help refine carbonation rates. Although MgO looping is an innovative approach to CO_2 removal, rates will need to be accelerated to make this approach a competitive alternative to existing direct air capture technologies.

[1] McQueen et al. 2020. Nat. Commun. 11, 3299. [2] Kelemen et al. 2020. Chem. Geol. 550, 119628.