

## When Do Mineral Spatial Distributions Control Dissolution Rates?

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We use column experiments and reactive transport modelling to understand and quantify the effects of magnesite spatial distribution on its dissolution rate under different conditions of flow velocity and permeability contrast [1,2]. Columns of 10 cm length were packed with magnesite and quartz sand distributed in different spatial patterns but with the same magnesite weight percentage (10%). The “Mixed” column contained a uniformly-distributed mixture of magnesite and quartz. In the “zoned” columns, magnesite grains were distributed in 3, 2, or 1 cylindrical zones within the sand matrix (“Three-zone”, “Two-zone”, and “One-zone” columns, respectively). Zones were oriented in the direction parallel to (flow-parallel) or transverse to (flow-transverse) the main flow direction. A total of 86 flow-through experiments were conducted.

The rates varied by more than 4 orders of magnitude under the tested conditions even though the amount of magnesite in each column was the same. Under sufficiently low flow velocities (0.015 to 0.4 m/d), the columns reach equilibrium and the spatial patterns have no effect on dissolution rates. Under higher flow velocities (0.4 to 18 m/d), the Mixed column has the highest dissolution rate among columns. The rates from the flow-transverse columns are lower but close to those from the Mixed column, with the largest difference being 15%. The rates from the flow-parallel columns can be lower than those from the Mixed column by a factor of 2 to 6. The largest difference occurred under the highest flow velocity of 18 m/d when the permeability of the magnesite zone is more than one order of magnitude lower than that of the sand zone. Under this condition, the mass transport between the magnesite and sand zone is limited by the significant difference in permeability. The rates do not relate well to the total magnesite surface area because they are the same in all columns. Instead, the rates correlate linearly with the effective surface area  $A_e$ , defined here as the surface area of magnesite grains surrounded by fluids with  $IAP/K_{eq}$  values lower than 0.1.

[1] Salehikhoo *et al* (2013) *Geochim Cosmochim Acta*, **108**, 91 – 106. [2] Li *et al* (2014) *Geochim. Cosmochim Acta*, **126**, 555-573.