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Statistical scaling of nanoscale mineral dissolution rates

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Dissolution reactions are key drivers of mineral weathering processes taking place in subsurface environments. Upon shaping morphology of mineral-water interfaces, dissolution reactions determine, e.g., changes of surface roughness at the pore level and formation of preferential flowpaths across geologic formations. Therefore, these reactions underpin a wide variety of natural and engineered processes such as, e.g., mobility of chemicals across groundwater bodies or long-term stability of carbon sequestration strategies. Experimental investigations document that dissolution rates exhibit several-fold variations even across the same mineral surface. This wide variability stems from the action of local mechanistic processes such as, e.g., etch pits. These form at natural crystal defects that are randomly distributed within mineral lattices. Starting from the nanoscale, the action of such local phenomena then propagates across various (spatial and temporal) scales and is ultimately responsible for heterogeneous distributions of dissolution rates typically required to effectively characterize reactive transport models. Hence, accurate assessment and modeling of dissolution rates must incorporate the effect of these processes. Given the impossibility of including these surface details in reactive models in a deterministic way, reliance on stochastic approaches is the only viable way to embed the richness of processes governing evolution of mineral-fluid interfaces and their inherently (stochastic) multiscale nature. In this context, understanding and quantifying how key statistical traits of rate spectra (i.e., sample probability densities of reaction rates) transition with scale are critical research aspects. We investigate scaling behavior of dissolution rates through analysis of sample structure functions associated with rate maps of a calcite sample subject to dissolution acquired via Atomic Force Microscopy. Sample structure functions correspond to absolute *q-th* order statistical moments of spatial increments (i.e., differences between values of observed dissolution rates taken between two locations separated by a given distance, spatial correlations being recovered as a particular case for $q = 2$). Our analysis documents the emergence of distinct scaling regimes that can be directly linked to nanoscale mechanistic processes driving mineral-water interface evolution. Finally, we provide an original stochastic framework that can capture under a unified theoretical lens key traits of sample rate spectra, along with observed scaling behavior.