Upscaling of dissolution rate contributors from the atomic scale to the macroscopic scale

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The concept of rate spectra provides quantitative and mechanistic insight into the kinetics of fluid-solid reactions. Instead of a single mean rate it provides information about all rate components that combine to yield the overall rate. Critical modes of this spectrum reveal the dominant rate contributors [1,2]. In this study we combine dissolution reaction data based on kinetic Monte Carlo (KMC) simulations with experimental data from AFM, phase shift and white light interferometry, and µCT. This synthesis covers an analytical length scale from the sub-nm up to the cm. The temporal scale ranges from fast processes (KMC, AFM) to the evolution of surfaces within several hours to weeks (**Fig. 1**). We show the quantitative impact of parameters such as crystal defect type and distribution, of superimposing stepwaves, and of grain boundaries on the scale-dependent rate data. The key results are calculated rate spectra, which are constrained by rate-controlling mechanisms on diverse scales. Such mechanistic rate spectra are applicable to reactive transport investigations on the scale of pores within rocks and technical materials.

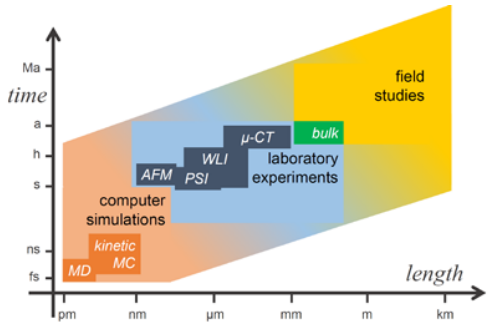

Fig. 1: Upscaling of rate data based on temporal and length scale-depending computational and analytical techniques

[1] Fischer & Luttge (2017) *EPSL* **457**, 100-105.
[2] Michaelis et al. (2017) *ES&T*, 10.1021/acs.est.6b05732