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Hierarchical modelling of solute diffusion in clays: From the molecular to the continuum scale

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The diffusion properties of clays are most often investigated in laboratory small-scale experiments using uncharged, anionic, and cationic tracers. Typically, the flux of anions through clay samples is lower than that of water tracers like tritiated water. This is explained by anion exclusion effects, which push back anions from the charged clay surfaces towards the centre of the pores and thus reduce the anion accessible porosity to half or less of the water accessible porosity. The diffusion of cations is slowed down because of sorption to the negatively charged clay surfaces. In certain cases, however, the cation flux is larger than expected from the sorption capacity, which is often attributed to surface diffusion, that is, a diffusion of sorbed cations along the clay interlayers or along external surfaces. The consistent description of such phenomena like anion exclusion and surface diffusion with continuum-scale models is rather complicated and relies mostly on empirical parameters. A consistent prediction requires consideration of pore-scale processes, because the ion or tracer specific diffusion behaviour depends on the type and the precise arrangement of the clay phases within the sample. In this presentation, we report on our approach to upscale the specific transport behaviour of anions, cations, and water tracers from the molecular or pore scale to the continuum scale. We developed a random walk model that operates on a pore-scale description of the clay structure. Obtaining such a description for clays is itself a difficult task, because it is hardly possible to directly visualise the very small pores in the nanometre range. Instead, the description has to rely on a combination of mostly indirect methods. We then used the model to predict diffusion coefficients of water tracers, anions, and cations in a given clay. It is possible to obtain these coefficients in a consistent way for a single structural description, using input parameters derived from independent measurements or simulations. A very promising way of obtaining such parameters is molecular modelling, where the individual motions of molecules and atoms are obtained based on inter-atomic interaction potentials. Combining the results of atomistic simulations with the random walk approach, we were able to upscale local molecular diffusion of ions and water to the macroscopic scale. At present, the limitations of the description of the pore structure create large uncertainties of the upscaled results, but the method seems to be very promising for the future.