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Chloride accessible porosity fractions across the Jurassic sedimentary rocks of northern Switzerland

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Clay-rich sedimentary rocks in northern Switzerland are investigated as a potential site for nuclear waste disposal. Chloride tracer profiles are key to assess the long-term transport regime in the hostrock and bounding formations as well as boundary conditions. Anions are depleted close to the negatively charged surface of clay minerals compared to the free porosity further away, that contains a charge-balanced electrolyte solution. The ratio between the free and total porosity is termed chloride accessible porosity fraction (f_{Cl}). This parameter is crucial to scale bulk porewater Cl concentration from aqueous extraction to free porewater Cl concentrations and hence produce chloride tracer profiles as a basis for solute transport modelling.

To date f_{Cl} values of ~0.5 have been applied to many of the investigated clay-rich rocks, however, there is a lack of data for clay-poor rocks (clay mineral content <30%). A vast set of f_{Cl} data from advective displacement (AD), squeezing (SQ) and through-diffusion (DI) experiments gives insight into differences between the three methods as well as variability of f_{Cl} at low clay mineral content. We observe a strong dependence of the f_{Cl} on lithology (e.g., clay content) and on the ionic strength. At high ionic strength, all three methods give consistent results but at low ionic strength the f_{Cl} from DI are substantially lower than those from AD and SQ. The variation of the f_{Cl} from DI with the ionic strength is in agreement with theoretical diffuse layer models. Two empirical approaches are tested to extrapolate the experimentally derived f_{Cl} values across the whole sedimentary sequence: 1) a model based on the clay mineral content 2) a model based on formation-specific averages of experimentally derived f_{Cl} values. This results in a coherent f_{Cl} dataset for all Jurassic formations sampled in this extended study, and this can be used to compile profiles of Cl in the free porewater and explain the processes leading to their shapes. The field-based models remove most apparent outliers seen in bulk-Cl profiles, and compare well with Br/Cl bulk profiles that do not depend on assumptions regarding anion exclusion.