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Modelling study on buffering pH and retaining U using a simplified uranium mill tailings pile example

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The hypothetical problem that is presented here considers the release and migration of uranium from a simplified uranium mill tailings pile towards a river. The modeling exercise with the coupled reactive transport model HP2 illustrates the effect of the geochemical conceptual model for sorption on (i) the buffering of the pH in the soil/aquifer system and (ii) the retention of U in the soil. The HP2 module, which couples the PHREEQC geochemical code with HYDRUS (2D/3D), is a two-dimensional equivalent of the one-dimensional HP1 program that was first released in 2005 (Jacques et al., 2008), and used successfully in many applications. Sorption of U is described using a multi-site cation exchange model (see Jacques et al., 2008). This sorption model also buffers the acid pH due to proton exchange. Two scenarios are considered: a soil with a relatively low (8.1 × 10-3 mol/kg) and relatively high (8.1 × 10-2 mol/kg) sorption capacity. In the third scenario, specific sorption of U and other cations and anions on Fe-oxides is described using a non-electrostatic surface complexation model with a very low capacity (8.1 × 10-4 mol/kg), in addition to low exchange complexation.

Proton exchange on the cation exchanger buffers the acidity by replacing calcium with protons on the exchanger; the spatial extent of the pH-perturbed region is smaller in the scenario with the higher exchange capacity. Specific sorption has only a small effect on the pH-perturbed zone, although it is important to note that its capacity is one order of magnitude lower than in the scenario with the low sorption capacity. U reaches the river system within 1000 d in scenarios with low and high exchange capacities. Only in the scenario with specific sorption, U migration within the ground water system is retarded, compared to the other two cases. The results of the three scenarios do not seem to be intuitive, especially the equally fast movement of U in the scenario with a high exchange capacity compared to one with a low exchange capacity. This is the result of a complex interplay between different aqueous components (U, H, C, Ca), sorption sites, and minerals and processes such as aqueous complexation and acidity buffering. Especially, higher buffering capacity in the scenario with a high sorption capacity produces alkaline pHs for longer times compared to scenarios with low sorption capacities, and consequently, a higher mobility of U-C complexes and lower Kd values, at least for some time.

This example shows possible effects of interacting geochemical processes such as aqueous complexation, proton exchange (and related acidity buffering), and U sorption. It highlights the importance of both pH (and the pH buffering mechanism) and specific sorption sites on the mobility of U in a ground water system subject to acidification in a uranium mill tailings pile.

Jacques, D., Šimůnek, J., Mallants, D., van Genuchten, M.T., 2008. Modeling Coupled Hydrologic and Chemical Processes: Long-Term Uranium Transport following Phosphorus Fertilization. Vadose Zone Journal 7, 698–711.