MODELLING THE EFFECT OF SORPTION MODEL ON pH BUFEERING AND U RETENTION

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Introduction

Geochemical speciation, interaction between different solutes, and interaction with the solid surfaces are critical processes determining the fate and transport of heavy metals and radionuclides in the vadose zone. Approaches based on linear or nonlinear equilibrium sorption neglect many of these aspects. On the other hand, so-called reactive transport models coupling flow, transport and geochemical processes in a single numerical tool allow for simulating a multicomponent system involving multiple interacting components which may result in an enhanced understanding of the possible critical processes (Steefel et al., 2015). This is demonstrated with a hypothetical example based on a simplified numerical model of leaching in simplified uranium mill tailing pile. The main objective is to evaluate the effect of the conceptual model of sorption on pH buffering in the soil and both will also influence the retention of U.

Methods

The example was inspired by a problem reported by Yeh and Tripathi (1991). We model the release and transport of U and acidified water from a U mill tailing pile towards a river in a 2D flow and transport domain (Figure 1).
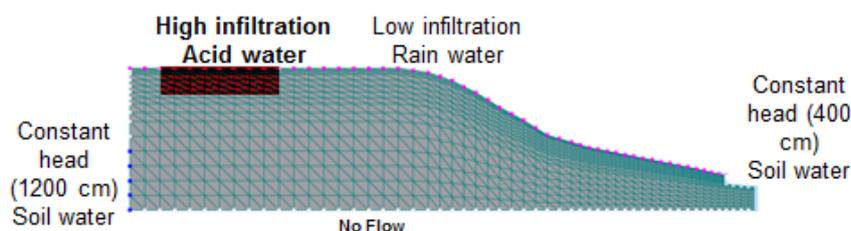

Figure 1. Simulation domain with mill tailing indicated in red. Schematic boundary conditions are also indicated.

The problem consists of eight components: Total H, Total O, Ca, C, uranium, sulfate, phosphate, and Fe. Sorption of U is described with a multi-site cation exchange model which also buffers the acid pH due to proton exchange – a variant with a low and with a high exchange capacity are simulated. In addition, the low exchange capacity model is combined with a specific U-sorption model described with a non-electrostatic surface complexation model.

The three models are implemented in the two-dimensional version of the HPx model (Šimůnek et al., 2006; Jacques et al., 2008). HPx couples the flow and transport model HYDRUS1/2D with the geochemical solver PHREECQ resulting in a versatile tool to tackle a large variety of problems related to the fate of heavy metals in soil systems.

Results

Figure 2 shows that a high exchange capacity buffers the pH more significantly. On the other hand, the specific sorption model retains U more than the other two models. However, a large difference between case 1 and case 2 is that U is more sorbed in the mill area, but once U is in the soil system it moves faster (not shown in Figure 2 – this is revealed by time series at specific points). U sorption increase in the pH range 2-5, but decrease again at pH > 5 due to U aqueous complexation (especially C-complexes).
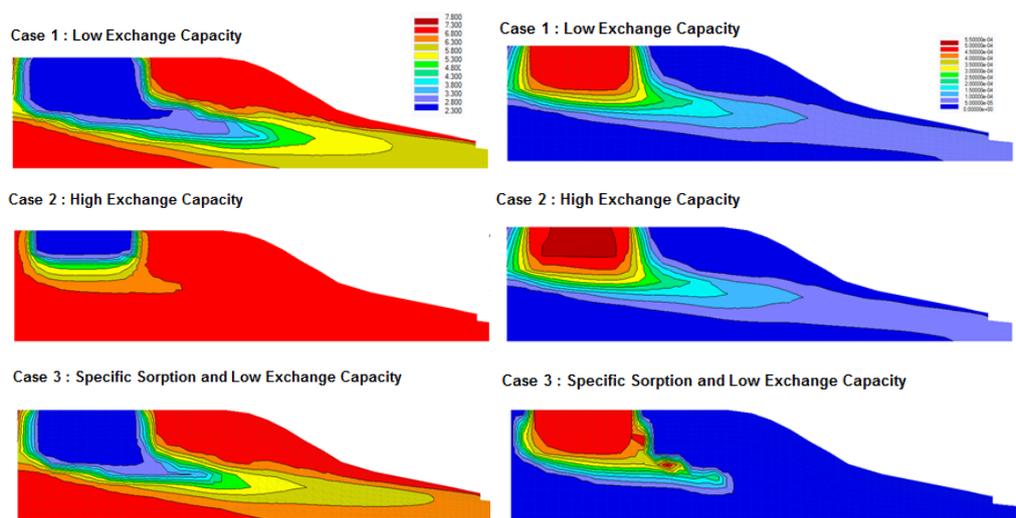
Figure 2. pH (left) and U distribution after 1000 d for the 3 cases.

In case 2, the similar pH in and near the mill tailing results in a higher sorption in case 2 (larger exchange capacity). However, further from the mill tailings, the higher buffer capacity results in higher pH values in case 2. Although a higher exchange capacity was assumed in case 2, the sorption is lower than in case 1 in this zone.

Conclusion

Interaction between pH dependency of aqueous speciation (e.g. U-C), conceptual sorption model, pH and buffering capacity results in a complex U transport process. It was illustrated that reactive transport codes could help to understand unintuitive behavior as e.g. a similar U pattern in cases with a low and high exchange capacity.

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