

A GENERIC REACTIVE TRANSPORT MODEL TO SIMULATE THE FATE OF HEAVY METALS IN SOILS: MERCURY AS A CASE STUDY

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Introduction - HPx

Soils are a common receptor of heavy metals, often acting as a pool due to strong adsorbing properties of many compounds. Recent developments in reactive transport codes hold promising prospects for the modelling the fate of heavy metals in the subsurface. In this abstract, we present the simulator HPx, which couples the HYDRUS codes with PHREEQC. HPx is specialized in variably-saturated soil systems and explicitly accounts for atmospheric boundary conditions and root water uptake. It combines most of the advanced features of the two individual codes (Šimůnek et al., 2006; Jacques et al., 2008). As a case study, we present a numerical model simulating mercury (Hg) fate in soils after a hypothetical anthropogenic contamination.

Case Study Mercury Fate - Methods

Figure 1 (a) shows the conceptual model developed for the fate and transport of Hg contamination from anthropogenic origin (Leterme et al., 2014). Flow (Richards equation, root water uptake) and transport (advection-dispersion in the aqueous phase, diffusion in gaseous phase) processes are defined for a uniform porous medium.

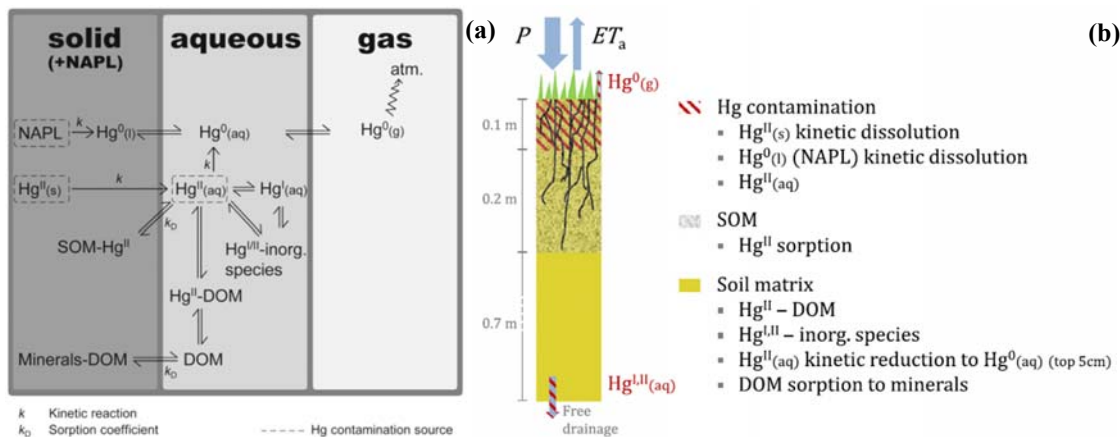


Figure 1. (a) Conceptual model of Hg speciation and reactions in the solid, aqueous and gas phases. Initial Hg release can be in the form of NAPL, solid or aqueous phase (source Leterme et al., 2014); **(b)** test case geometry for the simulation of Hg fate and transport in contaminated soil systems. P is the precipitation throughfall and ET_a the actual evapotranspiration (50-year time series from northern Belgium). Source: Leterme and Jacques, 2015.

Aqueous complexation reactions between inorganic and organic primary species are calculated using a thermodynamic approach (mass action laws). We use a version of the Thermoddem database (Blanc et al. 2012) updated for inorganic mercury species combined with surface complexation based on Skyllberg (2012). Complexation of Hg^{2+} with dissolved organic matter (DOM) is represented by complexation with

four reactants symbolizing humic and fulvic acids and thiols. An approach based on multiple proton/ion exchangers is used to describe the interactions between Hg and immobile SOM considering the same four types of reactive surface sites are considered. The reduction of Hg^{2+} to Hg^0 is described by a first-order kinetic reaction. DOM sorption to mineral surfaces is modelled using a Langmuir isotherm. The test case geometry is presented in Figure 1 (b). Initial contamination in the top 10 cm is assumed to be cinnabar (HgS), released to the soil system by a DOM-dependent rate equation (see details in Leterme et al., 2014).

Case Study Mercury Fate - Results

Figure 2 shows (a) the depth distribution of HgS and Hg sorbed to SOM over time, and (b) Hg^{2+} leaching at the bottom of the 1-m soil profile. In this example, Hg leaching starts only after ~32 years due to the strong sorbing behaviour of Hg. A detailed sensitivity analysis of the model is provided in Leterme and Jacques (2015).

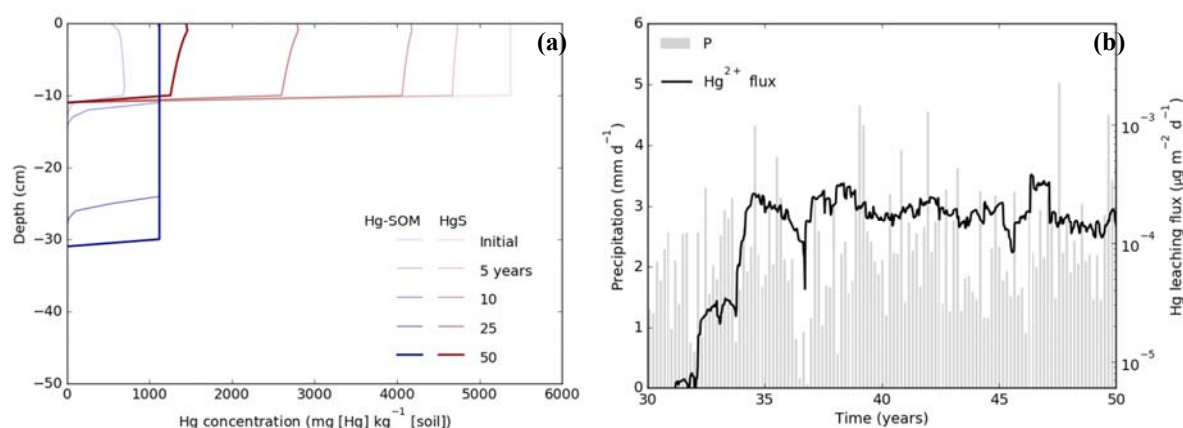


Figure 2. (a) Depth distribution of cinnabar (HgS ; red lines) and Hg sorbed to SOM (blue lines) at $t=0, 5, 10, 25$ and 50 years and (b) Hg^{2+} leaching flux vs time (starting at 30 years for clarity); with cinnabar as initial contamination.

Conclusion

Hg geochemistry is relatively complex and includes various speciation reactions and kinetic processes. Therefore, the test case presented here supports the versatility of HPx for modelling the fate of other heavy metals in soils. Potential applications include for example the protection of ecosystems, the reconstruction of historical contamination events or the evaluation of remediation strategies.

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