Methods for evaluating leaching and transport of redox-sensitive heavy metals at contaminated industrial sites

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Percolation water prognosis: Problem
The German Soil Protection Law (Bundesbodenschutzgesetz - BBodSchG, 1998) prescribes – in case of a supposed soil contamination – a prognosis of the quality and quantity of the water percolating through the unsaturated (veinose) zone into the groundwater (Fig. 1). In most cases the contaminated soil lies at the soil surface and is separated from the groundwater body by the unsaturated zone. The percolation water prognosis has to take into account the contaminant leaching from the source zone, the downward percolation through the unsaturated zone, and the interaction into the groundwater. The focus in this research project is on soils contaminated with redox-sensitive heavy metals, such as Cr and As.

Percolation water prognosis: Project structure
Our approach for the percolation prognosis consists of three parts (Fig. 2): 1. An analysis of the emission of metals from the contaminated source soil. This is done principally by using leaching, soil saturation extracts, and column experiments. 2. A prognosis of the throughflow of the unsaturated zone. This is performed with simulations of water flow and reaction solute transport. 3. A validation of the results gained by those first two steps. This is achieved by analysis of percolation water extracts in situ and undisturbed soils by leaching, which are installed at two different soil depths.

Study site
The main study site, Neumarkt/Oberpfalz, served as storage area for impregnated wooden mast and railway sleepers. Contamination consists of Cr, As, Cu, Hg and 
f favourable conditions for leaching. The main study site at the city of Neumarkt in Oberpfalz (Germany) is a meso-organic forest site (Fig. 3). Maximum total contents of As are 300–400 ppm, and Cr 1000 ppm.

Soil column experiments
For the column experiments, undisturbed columns were taken from different depths by pressing PVC tubes of diameter 12.5 cm and height 30 cm vertically into the soil. In order to account for spatial variability, columns were taken from different horizontal locations. The setup of the laboratory flow experiments is depicted in Fig. 4. The induced flow was saturated and driven only by gravity. The flow interruption yields information on non-equilibrium processes, such as kinetic sorption or intra-aggregate diffusion.

Measured leaching data
Measured concentrations of Cr in the leachates, (Fig. 5), columns taken from soil depth (30–65 cm), show an initial rapid decrease of concentrations. It should have been expected that Cr-concentrations in the bottom outflow remain at the same level for about one pore volume. The flow interruption is seen here as an increase of concentrations after about 25 cm cumulative bottom outflow.

Inverse Simulation of transport and sorption parameters – HYDRUS-1D
(Simunek et al., 1998)
One-dimensional nonequilibrium chemical transport of solutes
Total sorbed concentration:
Sum of equilibrium and kinetic sorbed concentration:
Temporal change of sorbed concentrations:
Inverse parameter optimization of chromium desorption data
Fig. 6: Inverse parameter optimization of chromium concentration in the column outflow, exemplarily for a column from 30–66 cm depth. Left: Optimization of the first part of the column experiment alone. Right: Optimization of the whole column experiment.

Direct simulation of chromium transport
Fig. 7: Direct simulation of chromium transport for a time of 10000 days (<2.7 years) using climate data for the period 1990—2001. Kd = 10 cm/g, α = 0.001 d−1, tmax = 0, N = 1; black line conc. at 1 m depth; blue conc. at 1.5 m depth; green conc. at the groundwater table (6 m depth).

Summary and Conclusions
- Estimation of sorption parameters from newly developed column experiments was performed with a two-site equilibrium-kinetic approach by inverse numerical modelling
- Parameter identification not always unique, other processes not accounted for
- Rapid decrease of desorbed concentrations could in the model only accounted for with exceedingly high dispersivities
- For practical engineering applications, the method has to be simplified
- Open questions: Spatial variability, Water repellency

References