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Closed-Flow Column Experiments - A Numerical Study on Breakthrough Oscillations Reveals a Decreased Uncertainty in the Inverse Determination of Transport Parameters

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The identification of transport parameters by inverse modeling often suffers from large uncertainties due to equifinality or parameter correlation when models are fitted to observations of the solute breakthrough in column outflow experiments. This issue can be approached by increasing the information potential of the observation, e.g. by running multiple experiments at different boundary conditions. A promising complementary approach of designing soil column experiments in order to further increase the obtained information is the closed-flow mode that is characterized by the recirculation of the effluent solution into the solution supply vessel. Depending on the experimental conditions, the solute concentration in the mixing vessel and the effluent follows a damped sinusoidal oscillation. As a result, the closed-flow experiment provides additional observables in the breakthrough curve, which are the initial exponential decrease in concentration in mixing vessel concentration, the oscillation wavelength and the extent of damping, each indicative for corresponding transport parameters. Furthermore, the concentration in the porous medium and the mixing vessel converges to equilibrium due to the closed loop. The evaluation of these emergent features allows intrinsic control over boundary conditions and impacts the uncertainty of parameters in inverse modeling.

We present a comprehensive numerical sensitivity analysis to illustrate the potential application of closed-flow experiments. We can show that the sensitivity with respect to the apparent dispersion can be controlled by the experimenter leading to a decrease in parameter uncertainty as compared to classical experiments by an order of magnitude for optimal settings. With these finding we are also able to show a reduction of equifinality found for situations, where rate-limited interactions impede a proper determination of the apparent dispersion and rate coefficients. This renders the closed-flow mode a useful complementary approach to classical column experiments in situations with severe parameter uncertainty.