Multiscale Assessment of Prediction Uncertainty in Coupled Reactive Transport Models

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Reactive transport simulations provide a systematic framework for integrating hydrologic and biogeochemical conceptual process models into a quantitative description of subsurface behaviors, which can then be used to project future concentrations. Assessing the uncertainty associated with these projections is difficult, because subsurface environments are open and complex, and subject to multiple interpretations and conceptualizations. This challenge is addressed using the method of multimodel analysis, in which model predictions are based on a weighted average of predictions of multiple plausible models, each of which is associated with a model averaging weight. For the multimodel analysis, several theoretical issues on quantification of model and parametric uncertainty are resolved in this study. First, we developed a statistical method to evaluate effects of the correlation of temporal residuals on model averaging weights for both synthetic and column experimental data. Disregarding the temporal correlation of the residuals may result in unrealistic weights for individual models that cannot be realistically justified. This problem is resolved by estimating the residual correlation using time series theories; the method is general and can be applied to other reactive transport models. For quantification of parametric uncertainty, we found that parametric uncertainty quantified using simpler regression methods is similar to that based on computationally expensive Bayesian methods. Finally, a Bayesian method of data-worth analysis was developed for selecting the potential dataset that can maximize the reduction of predictive uncertainty. This method is unique in that it reduces both parametric and model uncertainty whereas conventional methods of are limited to considering parametric uncertainty.

Parametric and conceptual model uncertainty for the stimulated bioreduction of U(VI) by microbial processes at the Rifle IFRC site was evaluated using a 1-D reactive transport model abstracted from a realistic 3-D model of one of the field experiments. The sensitivities of key biogeochemical concentrations vary in space and time and model nonlinearities have a significant impact on calculated sensitivities. However, sensitivities computed at the optimal parameter estimates provided similar information to a global sensitivity screening method. Observed data constrained parameter values much more tightly than indicated by likely parameter ranges used in the global sensitivity analysis. This suggests that model nonlinearities may not invalidate estimates of uncertainty based on linear methods. Synthetic observations obtained with the 3-D model were that were evaluated using 1-D modeling framework showed systematic errors in the 1-D model parameter estimates. Without considering the impact of non-uniform flux, the predictive uncertainty of the 1-D model was significantly underestimated.

Parameter and model uncertainty were also evaluated for a tracer tests that had increased or decreased alkalinity that were conducted at the Naturita, CO, UMTRA site. Significant uncertainty results from imperfect knowledge of the adsorption reactions that immobilize uranium at the mineral surfaces. A calibration study of ion exchange reactions and alternative adsorption reactions to observations at four wells demonstrated that the most probable adsorption models all contained dicarbonato surface complexes. Cross validation studies using the most probable models showed that the model parameters were robust and gave good predictions of the experiments excluded in the cross validation study. In addition, predictions using the most probable calibrated models bracketed the observations at a fifth well that was not included in the calibration study.