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Integration of parametric uncertainty and natural variability in the modeling of radionuclides speciation in rivers – Comparison with field data and predictions under global change scenarios

PHILIPPE CIFFROY

EDF R&D

For simulating the fate of metals and radionuclides in rivers, geochemical speciation models can be used. They generally combine many processes such as complexation with inorganic ligands, sorption to oxides and/or clay, complexation with dissolved and particulate organic carbon (DOC, POC) and sorption and/or co-precipitation to carbonates. It remains however difficult to estimate the confidence level one can place in model results since many parameters (*e.g.* thermodynamic constants, site densities on particles, etc.) and environmental physico-chemical variables (e.g. pH, DOC, POC, etc.) used in speciation modeling show large uncertainty or natural variability.

To overcome these limits, a geochemical speciation model (including all the processes mentioned above) was developed and designed to conduct uncertainty and sensitivity analysis. Sensitivity analysis follows a stepwise structured approach, starting from computationally 'inexpensive' Morris method to most costly variance-based EFAST method. These methods enable the identification of the variables that are primarily responsible for the overall uncertainty.

Furthermore, there is a need for discriminating *natural variability* and *knowledge uncertainty* in global uncertainty assessment. *Natural variability* is indeed an inherent property of the system (i.e. 'incompressible uncertainty') while *knowledge uncertainty* is dependent of the current state of knowledge regarding the system (i.e., uncertainty that can be reduced with more research). For discriminating *natural variability* and *knowledge uncertainty*, a nested Monte Carlo approach was implemented.

As case studies, speciation was estimated for Co, Mn, Ag and Cs radioisotopes in seven French rivers. Uncertainty analysis allowed to quantify partition coefficients (K_d*s*) ranges that can be expected when considering all the sensitive parameters together. Predicted K_d*s* were compared to monitoring values for the investigated rivers. It was observed that the range of predicted values are in good agreement with those from monitoring campaigns. It demonstrated the relevance of a geochemical modeling approach for predicting consistent K_d*s* confidence intervals.

Speciation of radionuclides was also predicted under global change scenarios, considering physicochemical modifications like ocean acidification, changes in the quantity and quality of organic matter and/or modifications in sediment redox conditions.