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Characterization of solid/liquid fractionation dynamics of radionuclides on riverine suspended particulate matter for integration into numerical models

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The transfer and fate of radionuclides discharged from nuclear facilities into rivers are conditioned by the distribution of their activity between the dissolved and particulate phases. This solid/liquid fractionation is commonly defined by an empirical partition coefficient noted K_d and its use in numerical models assumes an instantaneous equilibrium and reversible solid/liquid partition. However, these assumptions may not be verified when residence times are short (as in river), when the physicochemical properties of the environment are modified, or according to the age and origin of the contamination.

The objective of this study is to evaluate whether transfer models integrating one or more kinetics to describe this fractionation allow better estimations than an equilibrium approach.

An experimental protocol involving sorption and desorption of radionuclides on suspended particulate matter (SPM) from rivers of the Rhône watershed (France) was developed to characterize the short and long-term dynamics of this solid/liquid fractionation and therefore parametrize and compare different modelling approaches. SPM and water were sampled *in situ* and systematically characterized (grain-size, major and trace elements contents...). Batches containing the suspension were spiked with ^{137}Cs, ^{60}Co, ^{54}Mn and ^{110m}Ag, radionuclides released by nuclear power plants into rivers. Sorption was followed up from 0.5 h to 63 days, and SPM contaminated during the sorption stage were resuspended in non-contaminated filtered water for 1 h to 32 days. Isotherms were also performed to evaluate SPM sorption capacity.

The transfer of the four radionuclides between the solid and liquid phases showed at least two kinetic steps, and steady state was not reached after two weeks of sorption, for three different SPM samples collected along the Rhône River. Three types of models have been parametrized using the experimental data obtained: the usual equilibrium "K_d" for reference, a coupling between equilibrium and kinetic "E-K", and a two-kinetics approach "K_1-K_2". The added value of their integration into global transfer models developed at IRSN (CASTEAUR-X) and EDF (MERLIN-Expo) will be investigated with test scenarios and sensitivity analyses.