PFLOTRAN Isotope Partitioning for Kinetic and Equilibrium Processes in GDSA Framework

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Problem

- Certain kinetic reactions (e.g., kinetic colloid partitioning) are not yet possible for isotopes in GDSA Framework [1].
- Multiple isotopes for single element presents challenge.
- Kinetic process model should couple with equilibrium partitioning model of GDSA Framework.
- How will GDSA Framework include kinetic partitioning reactions for isotopes?
  - Likely by operator splitting

Conceptual Model

- Kinetic processes simulated first at each time step
  - Decay releases daughters to aqueous phase
- Equilibrium partitioning simulated at the end of each time step using the Isotope Partitioning Model (IPM) of GDSA Framework

Algorithm

- Kinetic Processes
  - Calculate total bulk isotope concentrations in cell over all phases after transport, colloid partitioning, and decay/ingrowth but prior to equilibrium partitioning step for aq., ads., and ppt:
  \[ C_i(t) = C_i(aq) + C_i(ads) + C_i(ppt) + C_i(col) \]
- Partitioning among EQ Phases
  - Calculate bulk aqueous concentration of element (i) (C_i(aq)), allowing for equilibrium adsorption partitioning (K_i), assuming no precipitate phase:
  \[ C_i(aq) = \frac{C_i(aq)(t)}{1 + K_iC_i(aq)} \]
- Determine whether kinetic process occurs:
  - yes:
    - Calculate concentration of total isotopes of each redox-specific element (i) in EQ regime:
    \[ C_i(EQ) = C_i(EQ)(a) + C_i(EQ)(ads) + C_i(EQ)(ppt) \]
    - Calculate conc. of total elements in EQ regime:
    \[ C_i(EQ) = \sum C_i(EQ) \]
    - Calculate new isotopic mole fractions in EQ regime:
    \[ X_i(EQ) = \frac{C_i(EQ)}{C_i(EQ)} \]
    - Distribute isotopes in EQ regime (maximum entropy):
    \[ C_i(aq)(t) = C_i(aq)(E)X_i(EQ) \]
    \[ C_i(ads) = C_i(ads)(E)X_i(EQ) \]
    \[ C_i(ppt) = C_i(ppt)(E)X_i(EQ) \]

Figure 1. Conceptual model of radionuclide (RN) equilibrium partitioning processes (blue) and kinetic processes (lavender).

Figure 2. Proposed coupling of IPM and kinetic colloid partitioning model.

Test

- Batch reactor with initial concentrations of 237Np, 233U, 229Th, and 230Th equilibrated among aqueous (aq), adsorbed (ads), and precipitated (ppt) phases.
- Isotopes decay (233U and 229Th will also ingrow):
  - 237Np → 233U → 229Th and 230Th irreversibly adsorbed at initial bulk concentrations of 10^{-10} mol L^{-1}.
- Simulate over time, allowing decay and ingrowth to change the total isotope concentrations and equilibrium partitioning.

Results

- Constitutive relationships are maintained.
- Proposed algorithm for integrating kinetic partitioning reactions with the Isotope Partitioning Model (IPM) of GDSA Framework [1] is ready for implementation.

Conclusions


Reference