Ca and Sr sorption on Ca-illite experimental study and modelling

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Ca AND Sr SORPTION ON Ca-ILLITE: EXPERIMENTAL STUDY AND MODELLING

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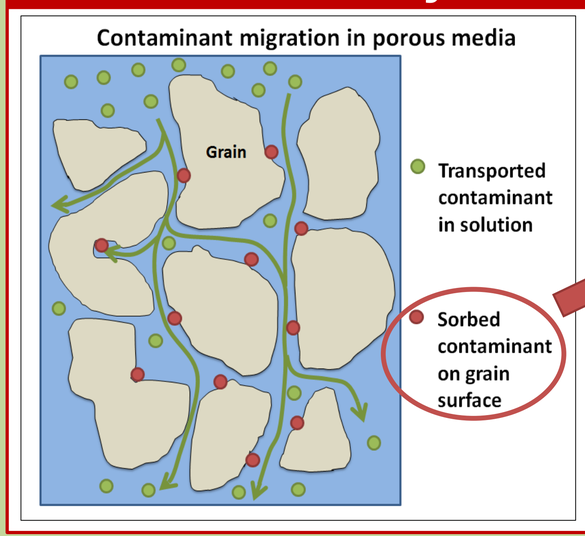
Context & objectives

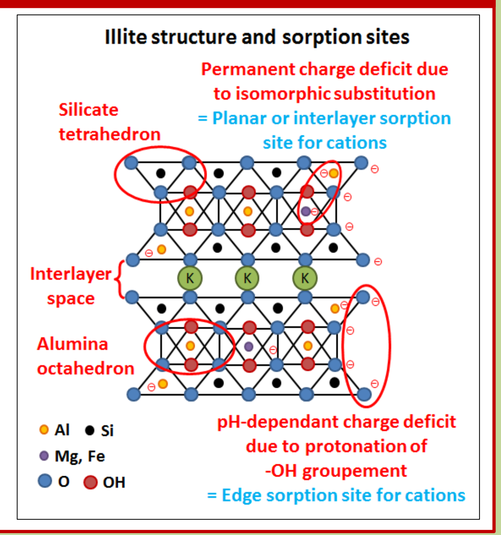


To ensure the environmental monitoring of nuclear sites, **sorption models** enabling the **prediction** of **contaminant migration** in **soils** and **aquifers** need to be developed.

Recently, a **multi-site ion exchanger model** based on the **Multi-site Ion Exchanger Theory**[1] has **successfully** been **applied** to evaluate the **sorption** of a **contaminant** in a **natural soil** under **static** and **dynamic conditions**[2,3].

*Need of a **database** containing **sorption properties** of **pure minerals** present in the studied environment*

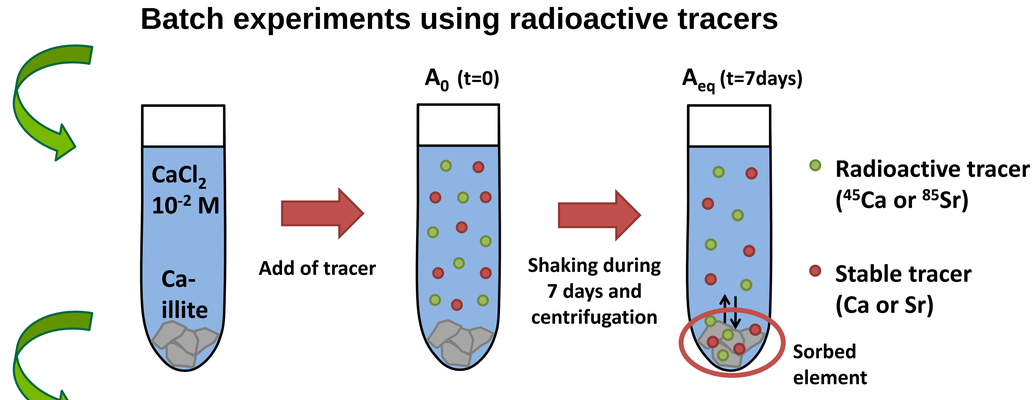
Lack of Ca and Sr sorption data onto illite



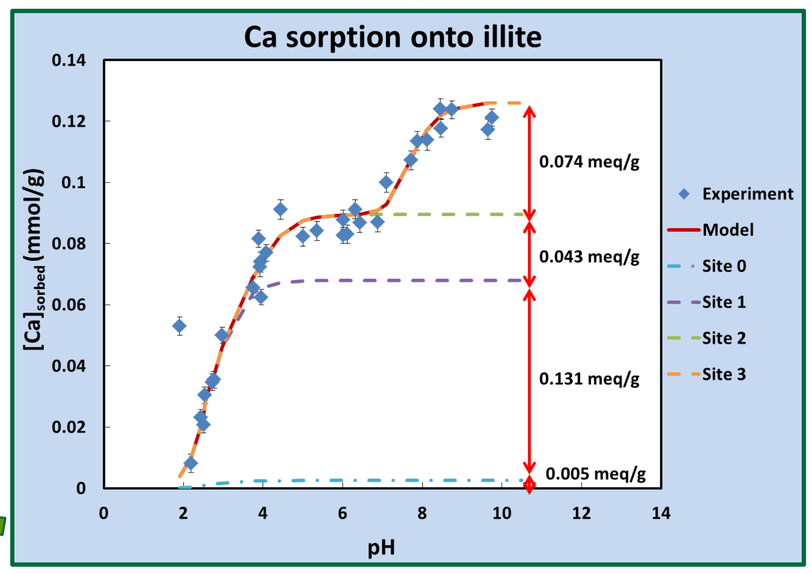
Methodology & results

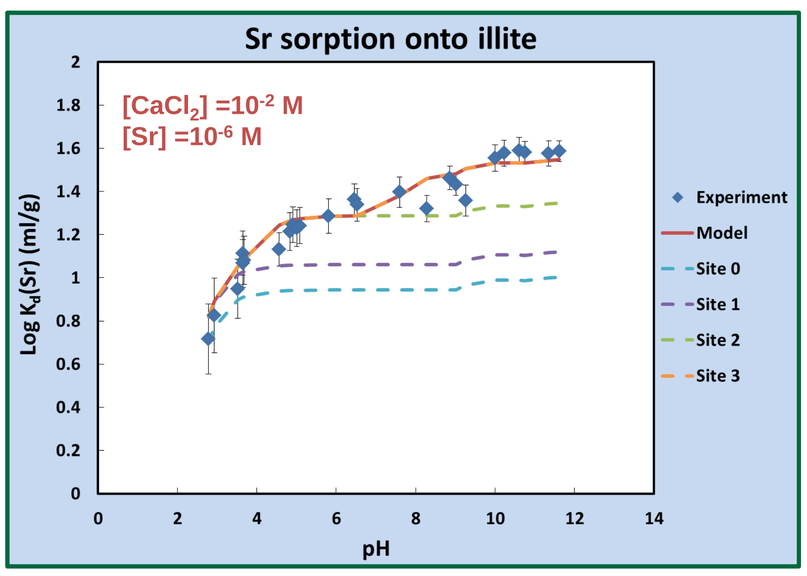
❑ Acquisition of sorption data on pure minerals

Batch experiments using radioactive tracers



$$K_d = \frac{[M]_{sorbed}}{[M]_{eq}} = \left(\frac{A_0}{A_{eq}} - 1\right).\frac{V_{Solution}}{m_{solid}}$$





❑ Determination of selectivity coefficients

Multi-site Ion Exchanger Theory [1]

Ion exchange equation on a X_i site between major cation M^{m+} and H^+:

$$m\{(X_i^-) - H^+\} + M^{m+} = \{(X_i^-)_m - M^{m+}\} + mH^+$$

Ion exchange equation on a X_i site between major cation M^{m+} and trace element N^{n+}:

$$m\{(X_i^-)_n - N^{n+}\} + nM^{m+} = n\{(X_i^-)_m - M^{m+}\} + mN^{n+}$$

Selectivity coefficients :

Components of the database

$$K^{*i}_{mH^+/M^{m+}} = \frac{[(X_i^-)_m - M^{m+}](H^+)^m}{[(X_i^-) - H^+]^m (M^{m+})} \qquad K^{*i}_{mN^{n+}/nM^{m+}} = \frac{[(X_i^-)_m - M^{m+}]^n (N^{n+})^m}{[(X_i^-)_n - N^{n+}]^m (M^{m+})^n}$$

$$K_d = \frac{\sum_i [(X_i^-)_m - M^{m+}]}{[M^{m+}]}$$

Site saturation over pH

Identification of 4 site concentrations

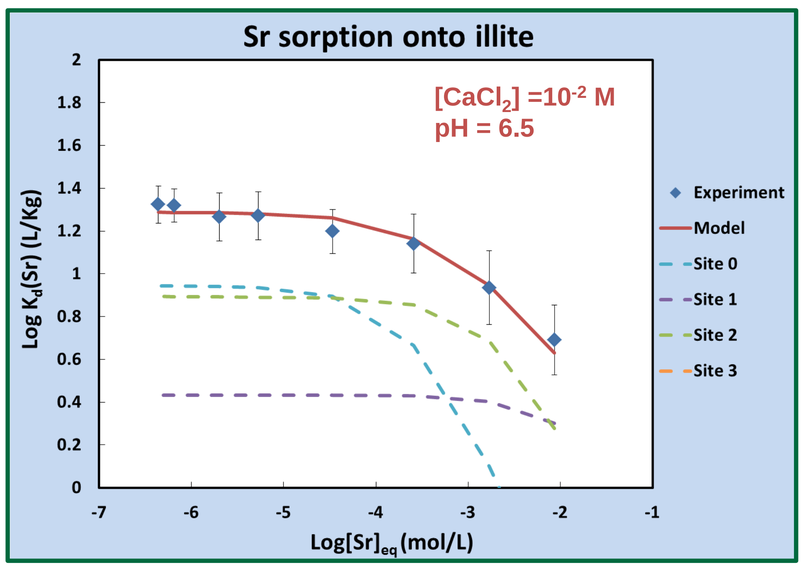
Acquisition of selectivity coefficients $K^{*i}_{H^+/Ca^{2+}}$

Sorption site	Concentration (mol/kg of dry solid) [4]	Log $K^{*i}_{H^+/Ca^{2+}}$	Log $K^{*i}_{H^+/Sr^{2+}}$
Site 0	**0.005**	-0.80 ± 0.19	0.74 ± 0.20
Site 1	**0.131**[4]	-2.21 ± 0.08	-2.60 ± 0.80
Site 2	**0.043**[4]	-4.54 ± 0.23	-3.97 ± 0.16
Site 3	**0.074**[4]	-12.01 ± 0.20	-11.53 ± 0.12

Kd ↗ over pH

Fitting with the 4 sites previously identified

Acquisition of selectivity coefficients $K^{*i}_{H^+/Sr^{2+}}$



Kd ↘ over Sr concentration

Higher sorption at low Sr concentration

Acquisition of selectivity coefficients $K^{*i}_{H^+/Sr^{2+}}$

Conclusions & perspectives

Conclusion :

- ✓ **Acquisition of Ca and Sr sorption data in a $CaCl_2$ background solution on a conditioned Ca-illite under static conditions**
- ✓ **Sorption data well reproduced by the Multi-site Ion Exchanger Theory[1] considering 4 types of sites and their concentrations**
- ✓ **Determination of selectivity coefficients between cations Ca^{2+}, Sr^{2+} and H^+**

Perspectives :

- ❑ **Selectivity coefficients obtained has been used under static and dynamic conditions to model Ca or Sr sorption on complex solids composed in part of illite**
- ❑ **Same methodology will be used to model Cs sorption on complex solids composed in part of illite under static and dynamic conditions**

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[4] Gorgeon L. Contribution à la modélisation physico-chimique de la rétention de radioéléments à vie longue par des matériaux argileux. Thèse de doctorat, Université Pierre et Marie Curie (Paris VI), Paris; 1994.

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