Cesium Transport in Hanford Sediments: Application of an Experimentally Based Cation Exchange Model

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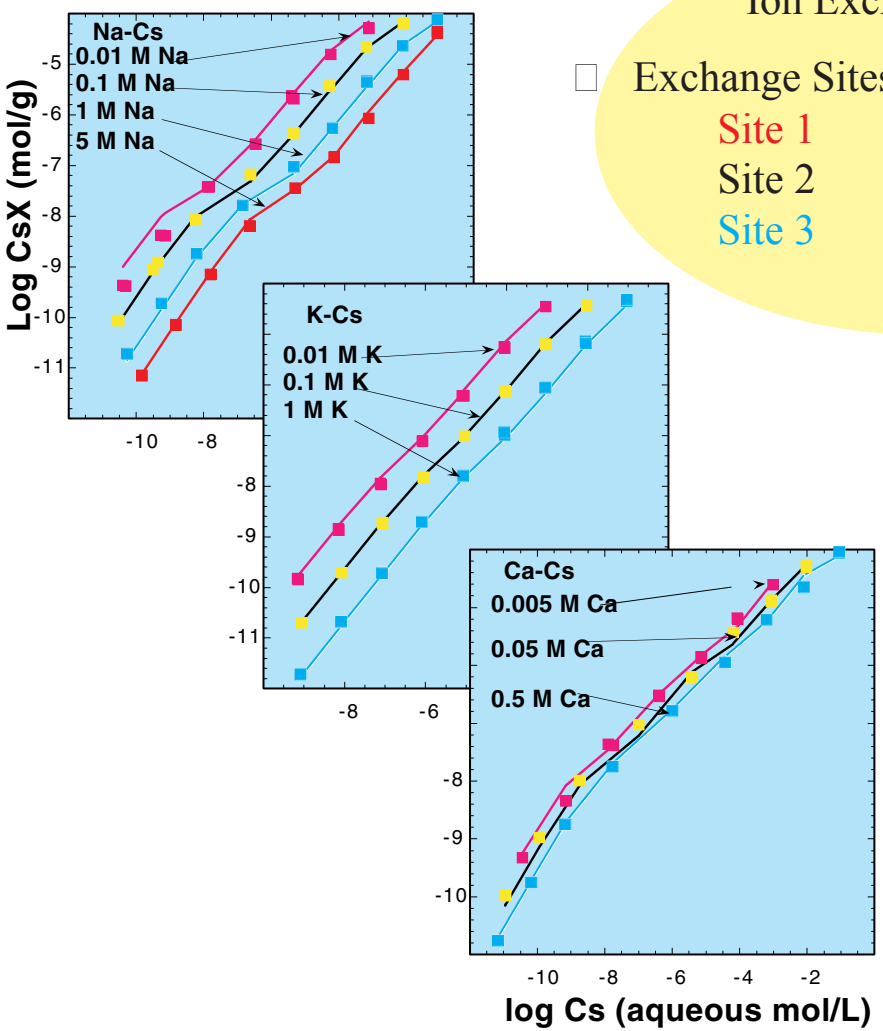
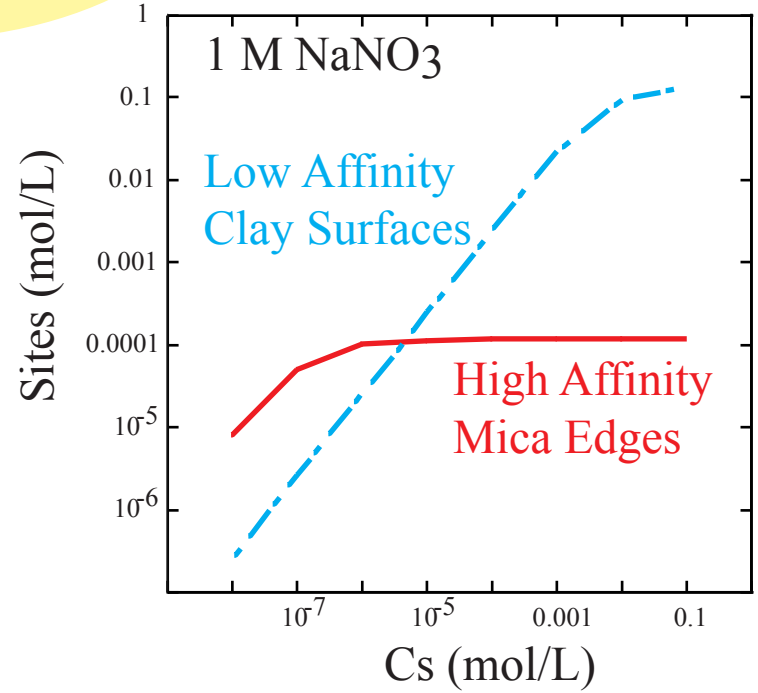
Global fit of 9 selectivity coefficients and 3 exchange sites from binary batch experiments (Data from Zachara et al, 2000)

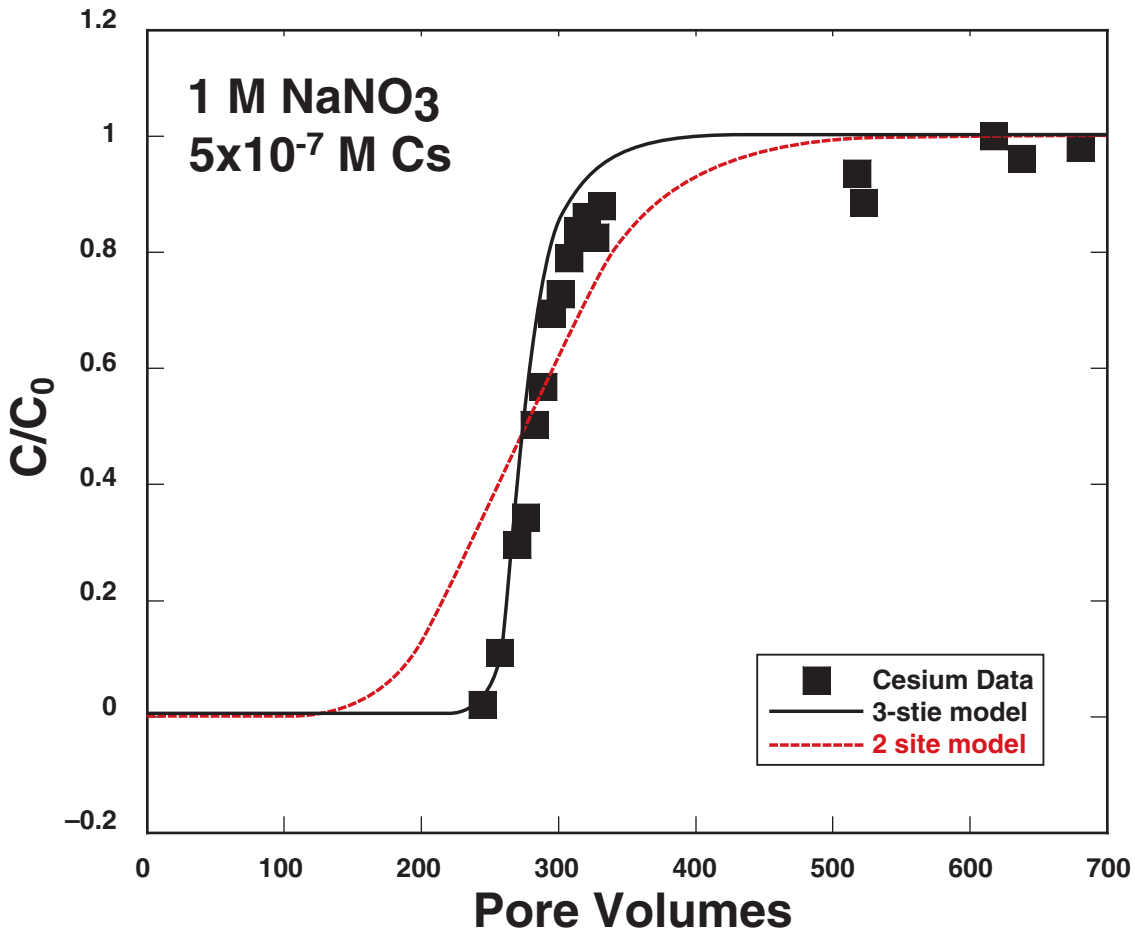


Ion Exchange Model

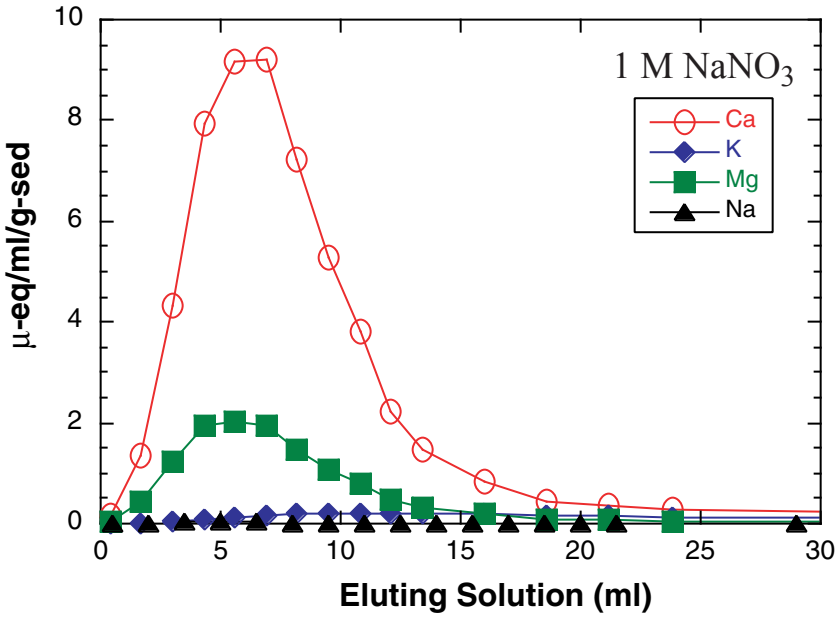
Exchange Sites	CEC (μeq g^{-1})
Site 1	0.03
Site 2	1.70
Site 3	120



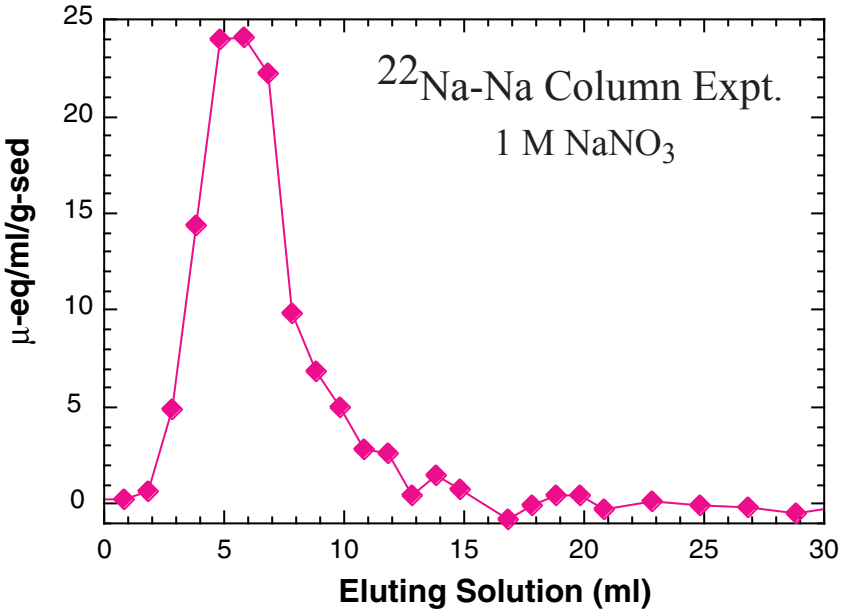
Comparison of column-weighted 2-site and 3-site exchange models



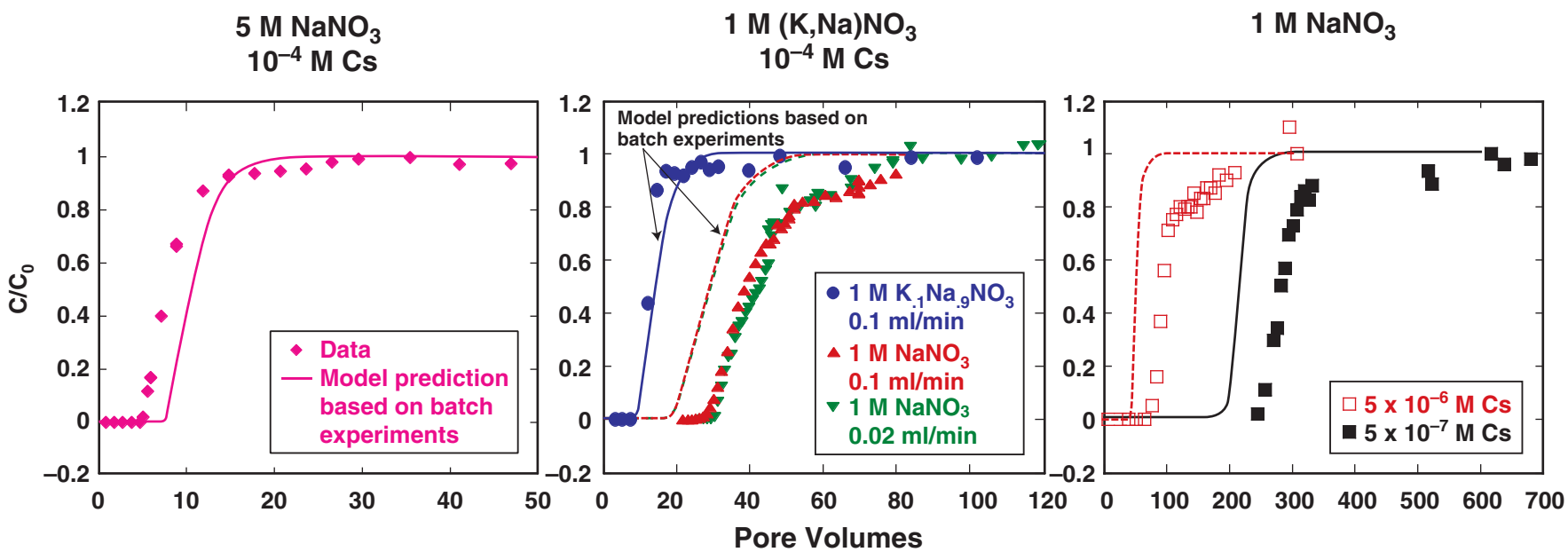
Hanford sediment CEC determined from ^{22}Na-Na column exchange experiments is about 3 times CEC determined in low ionic strength batch experiments.



	CEC (μeq/g)
Cation Elution Column Expt.	106
^{22}Na-Na Column Expt.	120
^{22}Na-Na Batch Expt.	46

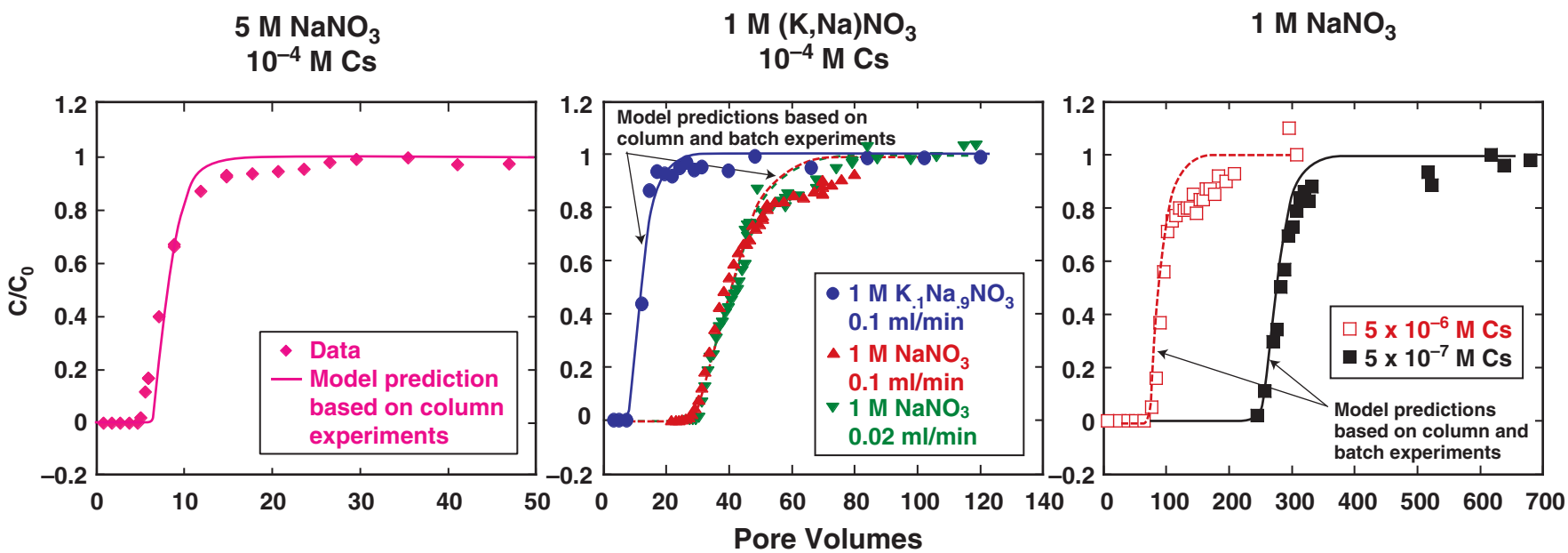


Batch-derived multi-cation exchange model: Comparison of predicted and experimental cesium reactive transport through Hanford sediments in (K,Na)NO_3 solutions



Clay Sites **Mica Sites**

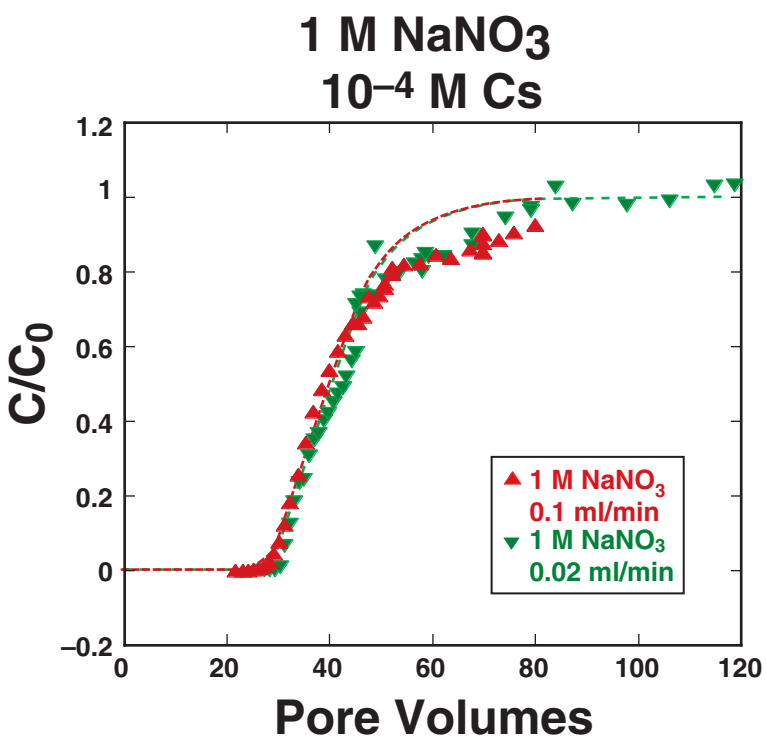
Column-weighted multi-cation exchange model: Comparison of predicted and experimental cesium reactive transport through Hanford sediments in (K,Na)NO_3 solutions

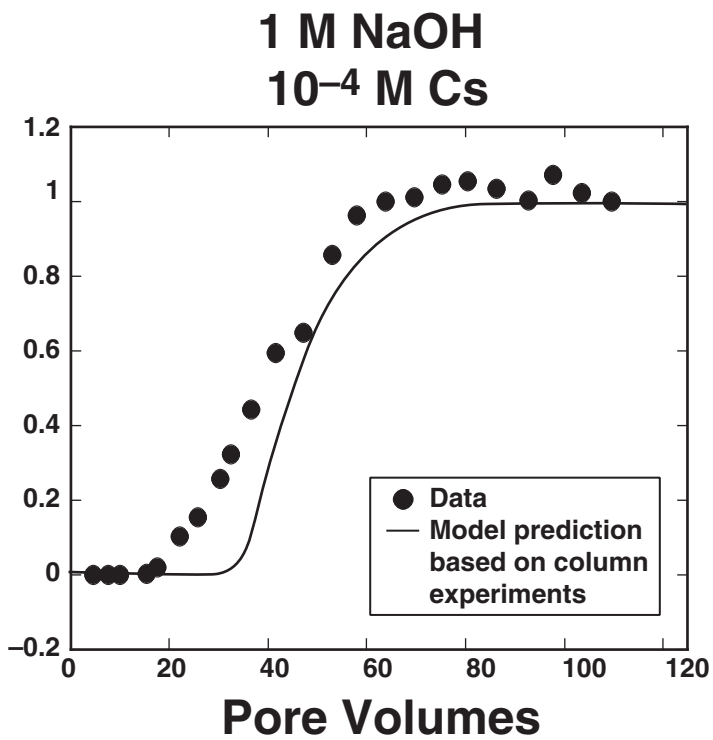


Clay Sites **Mica Sites**

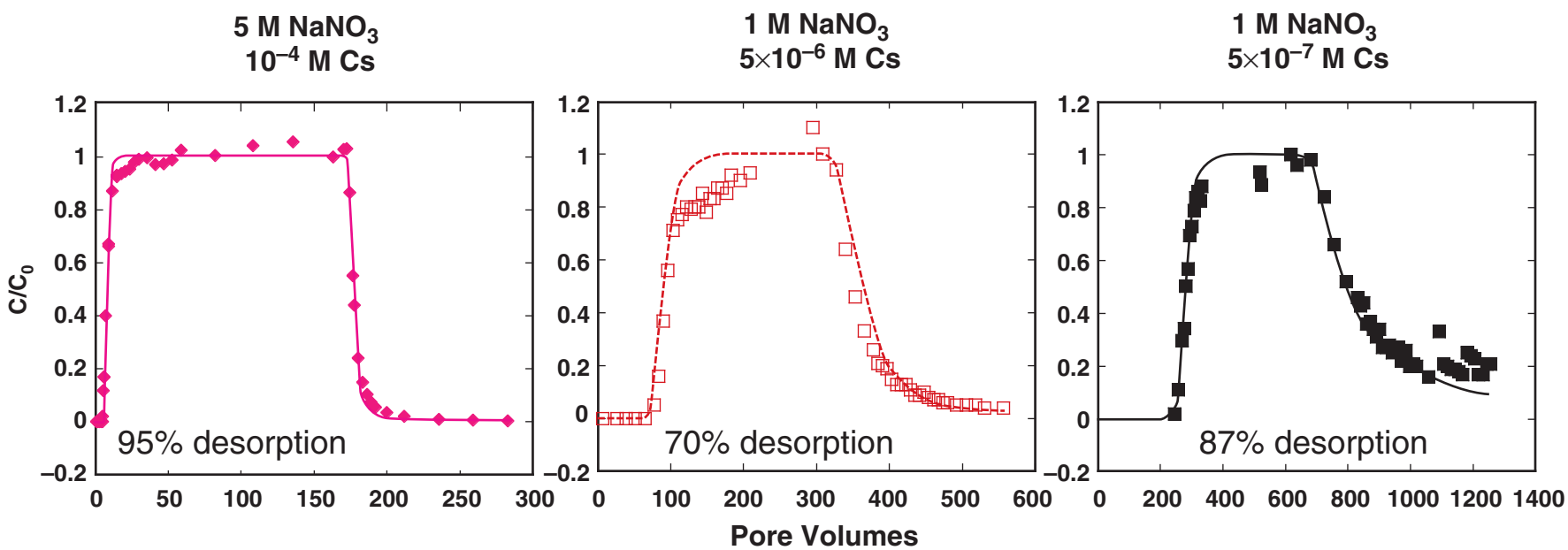
Multi-cation exchange model is independent of pH

(Model predictions using column parameters)



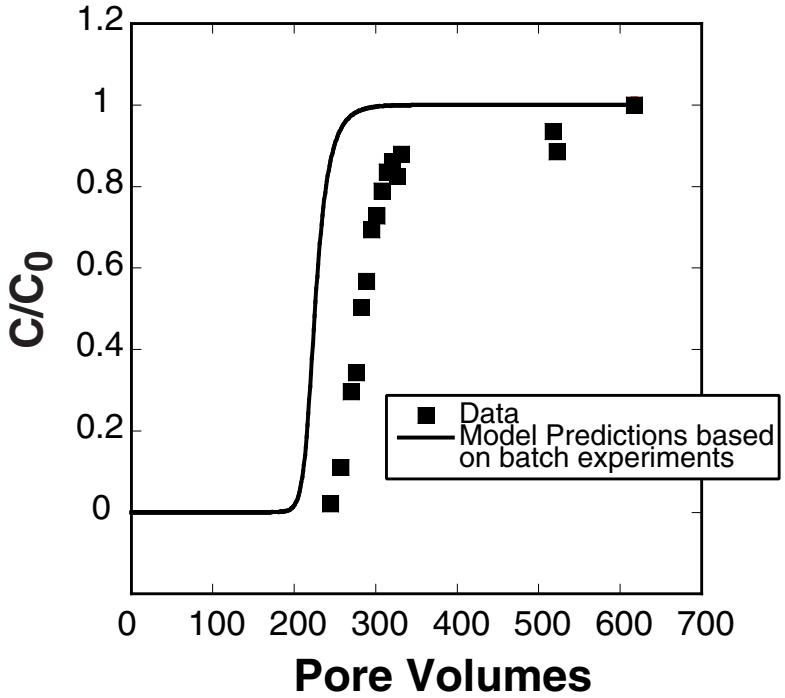


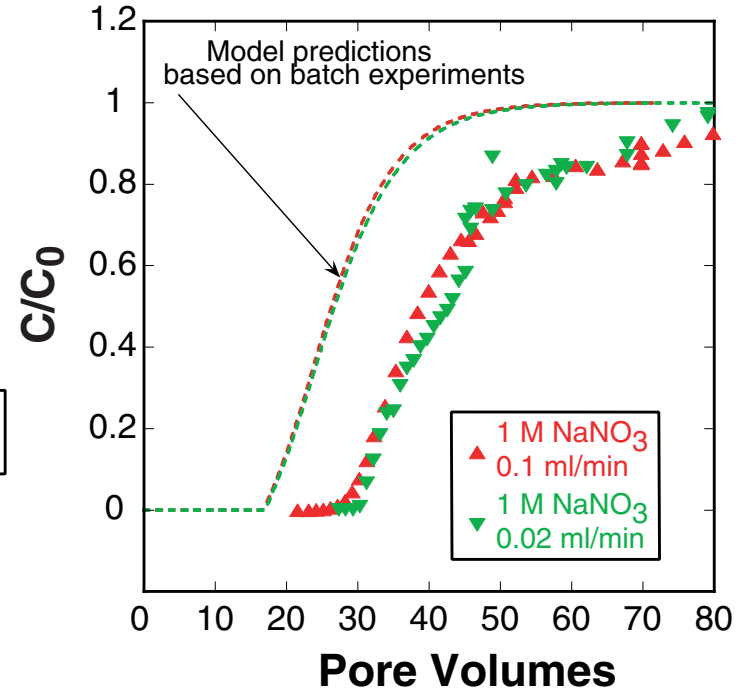
Evidence of exchange kinetics in at various Cs concentrations
(predictions calculated using column parameters)



Clay Sites **Clay and Mica Sites** **Mica Sites**

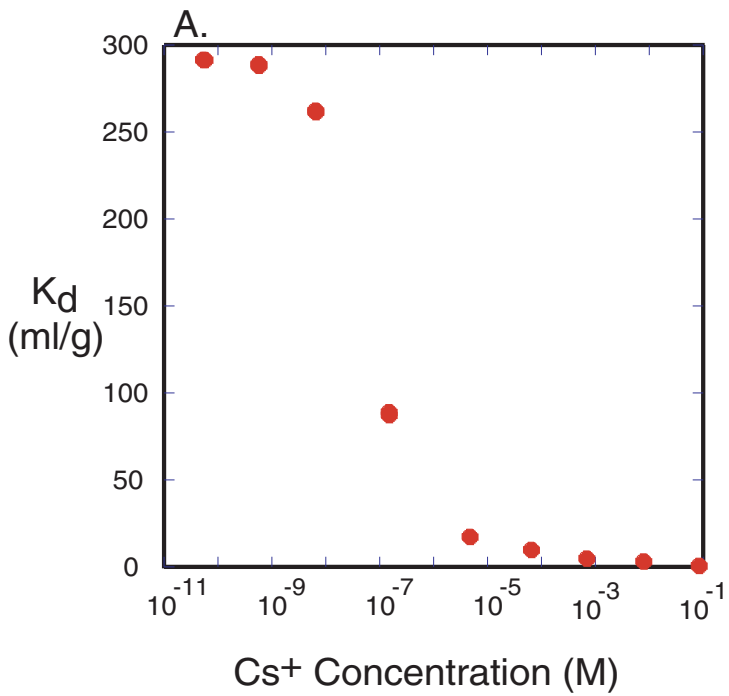
Inclusion of the "kinetic sites" helps resolve the discrepancy between the observed and predicted retardation based on the batch multi-cation exchange model

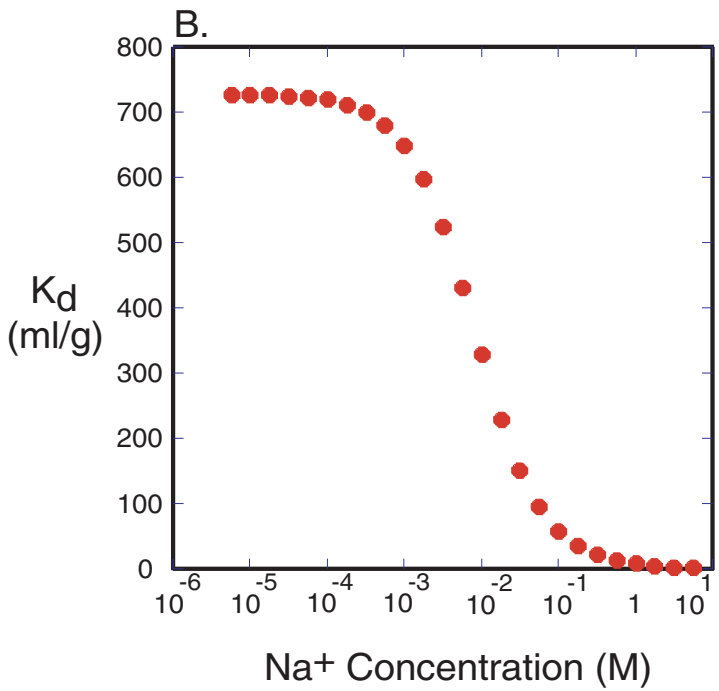




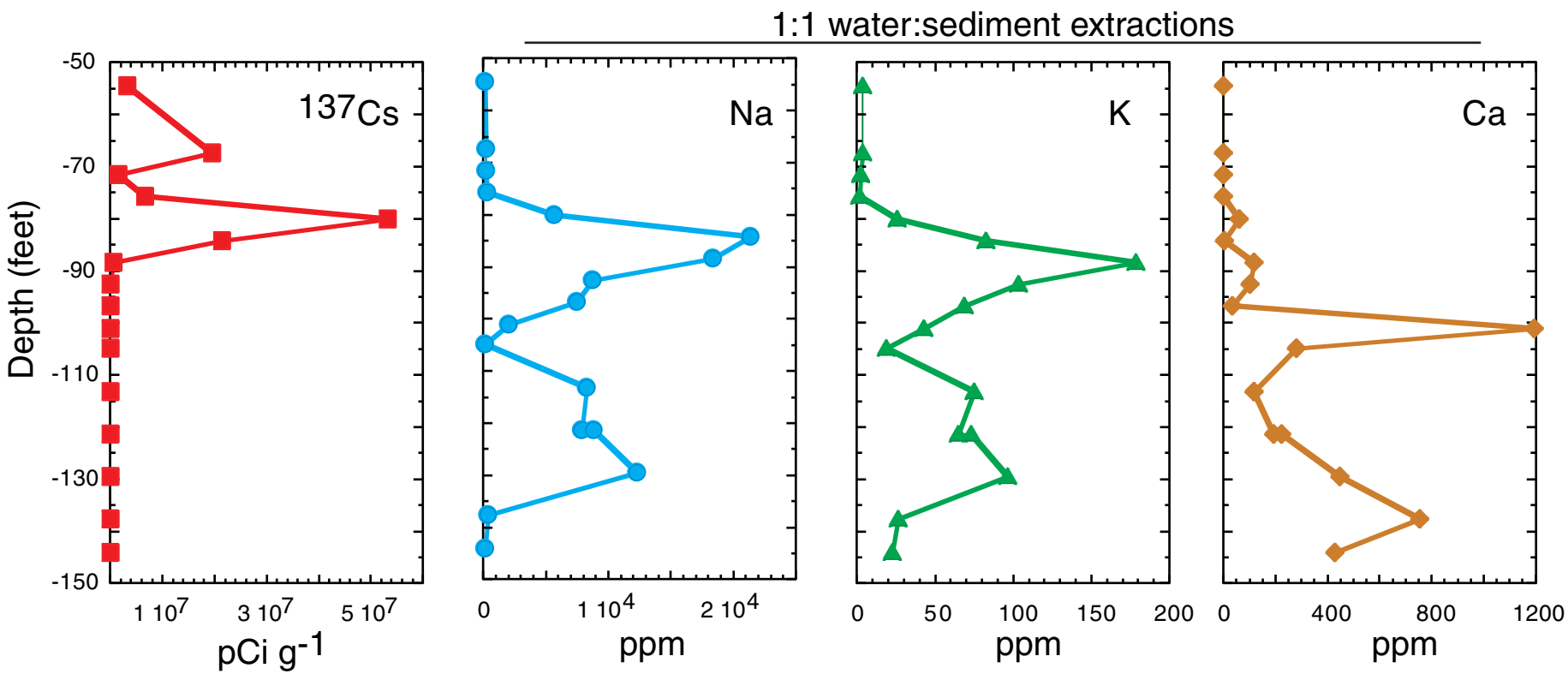
	1 M $NaNO_3$ and 5 x10^{-7} M Cs	1 M $NaNO_3$ and 10^{-4} M Cs
Observed Retardation	**282**	**40.3**
Model Retardation Batch Parameters	**230**	**35.5**
Model Retardation Batch Parameters Corrected for Kinetic Sites	**262**	**40.5**

The cesium K_d is dependent on dissolved cesium and sodium concentrations





Ion Exchange in the Hanford Sediments: ^{137}Cs and cation concentrations measured in the slant core taken beneath Tank SX-108



(Data from Serne, PNNL)

Conclusions

- The multi-cation exchange model captures much of the chemical effects responsible for the dependence of ^{137}Cs Kd on competing cation and cesium concentrations

  - ❑ This is a significant improvement over the use of a constant K_d to model ^{137}Cs form tank leaks to the vadose zone sediments.

  - ❑ The mismatch between the predictions derived from a global fit of batch data covering a wide range of cesium and salt concentrations and our 1-D column experiments may be related to the sorption of cesium at "kinetic" sites.

  - ❑ In the absence of these new experimental data, model parameters based on weighted column experiments can be used in field-scale analyses of transport to provide a more direct measure of retardation of cesium in Hanford sediments.