

Supporting Information for
Additive Surface Complexation Modeling of Uranium(VI) Adsorption onto Quartz-Sand
Dominated Sediments

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Environmental Science and Technology

Number of pages (including this one): 21

Number of tables: 5

Number of figures: 5

Table SI-1. U(VI) and the relevant aqueous reactions and constants

Reactions	log K (I = 0)	References
$\text{UO}_2^{2+} + \text{H}_2\text{O} = \text{UO}_2(\text{OH})^+ + \text{H}^+$	-5.25	1
$\text{UO}_2^{2+} + 2\text{H}_2\text{O} = \text{UO}_2(\text{OH})_2(\text{aq}) + 2\text{H}^+$	-12.15	1
$\text{UO}_2^{2+} + 3\text{H}_2\text{O} = \text{UO}_2(\text{OH})_3^- + 3\text{H}^+$	-20.25	1
$\text{UO}_2^{2+} + 4\text{H}_2\text{O} = \text{UO}_2(\text{OH})_4^{2-} + 4\text{H}^+$	-32.40	1
$2\text{UO}_2^{2+} + \text{H}_2\text{O} = (\text{UO}_2)_2(\text{OH})_2^{3+} + \text{H}^+$	-2.70	1
$2\text{UO}_2^{2+} + 2\text{H}_2\text{O} = (\text{UO}_2)_2(\text{OH})_2^{2+} + 2\text{H}^+$	-5.62	1
$3\text{UO}_2^{2+} + 4\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_4^{2+} + 4\text{H}^+$	-11.90	1
$3\text{UO}_2^{2+} + 5\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_5^+ + 5\text{H}^+$	-15.550	1
$3\text{UO}_2^{2+} + 7\text{H}_2\text{O} = (\text{UO}_2)_3(\text{OH})_7^- + 7\text{H}^+$	-32.20	1
$4\text{UO}_2^{2+} + 7\text{H}_2\text{O} = (\text{UO}_2)_4(\text{OH})_7^+ + 7\text{H}^+$	-21.90	1
$\text{UO}_2^{2+} + \text{CO}_3^{2-} = \text{UO}_2\text{CO}_3(\text{aq})$	9.94	1
$\text{UO}_2^{2+} + 2\text{CO}_3^{2-} = \text{UO}_2(\text{CO}_3)_2^{2-}$	16.61	1
$\text{UO}_2^{2+} + 3\text{CO}_3^{2-} = \text{UO}_2(\text{CO}_3)_3^{4-}$	21.84	1
$3\text{UO}_2^{2+} + 6\text{CO}_3^{2-} = (\text{UO}_2)_3(\text{CO}_3)_6^{6-}$	54.00	1
$2\text{UO}_2^{2+} + \text{CO}_3^{2-} + 3\text{H}_2\text{O} = (\text{UO}_2)_2\text{CO}_3(\text{OH})_3^- + 3\text{H}^+$	-0.855	1
$3\text{UO}_2^{2+} + \text{CO}_3^{2-} + 3\text{H}_2\text{O} = (\text{UO}_2)_3\text{O}(\text{OH})_2\text{HCO}_3^+ + 3\text{H}^+$	0.649	1
$\text{UO}_2^{2+} + 3\text{CO}_3^{2-} + \text{Ca}^{2+} = \text{CaUO}_2(\text{CO}_3)_3^{2-}$	27.18	2
$\text{UO}_2^{2+} + 3\text{CO}_3^{2-} + 2\text{Ca}^{2+} = \text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq})$	30.70	2
$\text{UO}_2^{2+} + 3\text{CO}_3^{2-} + \text{Mg}^{2+} = \text{MgUO}_2(\text{CO}_3)_3^{2-}$	25.80	3
$\text{UO}_2^{2+} + \text{H}_4\text{SiO}_4 = \text{UO}_2\text{SiO}(\text{OH})_3^- + \text{H}^+$	-1.67	4
$\text{UO}_2^{2+} + \text{NO}_3^- = \text{UO}_2\text{NO}_3^+$	0.3	1
$\text{H}_2\text{O} = \text{H}^+ + \text{OH}^-$	-14.0	5
$\text{CO}_2(\text{g}) = \text{CO}_2(\text{aq})$	-1.472	1
$\text{CO}_2 + \text{H}_2\text{O} = 2\text{H}^+ + \text{CO}_3^{2-}$	-16.683	1
$\text{HCO}_3^- = \text{H}^+ + \text{CO}_3^{2-}$	-10.329	1
$\text{Na}^+ + \text{HCO}_3^- = \text{NaHCO}_3$	1.27	5
$\text{Na}^+ + \text{CO}_3^{2-} = \text{NaCO}_3^-$	-0.25	5
$\text{Na}^+ + \text{H}_2\text{O} = \text{NaOH} + \text{H}^+$	-14.18	5
$\text{K}^+ + \text{H}_2\text{O} = \text{KOH} + \text{H}^+$	-14.46	5
$\text{Al}^{3+} + \text{H}_2\text{O} = \text{AlOH}^{2+} + \text{H}^+$	-5.00	5
$\text{Al}^{3+} + 2\text{H}_2\text{O} = \text{Al}(\text{OH})_2^+ + 2\text{H}^+$	-10.1	5
$\text{Al}^{3+} + 3\text{H}_2\text{O} = \text{Al}(\text{OH})_3^0 + 3\text{H}^+$	-16.9	5
$\text{Al}^{3+} + 4\text{H}_2\text{O} = \text{Al}(\text{OH})_4^- + 4\text{H}^+$	-22.7	5
$\text{H}_4\text{SiO}_4(\text{aq}) = \text{H}_3\text{SiO}_4^- + \text{H}^+$	-9.810	1
$\text{H}_4\text{SiO}_4(\text{aq}) = \text{H}_2\text{SiO}_4^{2-} + 2\text{H}^+$	-23.140	1
$\text{Ca}^{2+} + \text{H}_2\text{O} = \text{CaOH}^+ + \text{H}^+$	-12.78	5
$\text{Ca}^{2+} + \text{CO}_3^{2-} + \text{H}^+ = \text{CaHCO}_3^+$	11.435	5
$\text{Ca}^{2+} + \text{CO}_3^{2-} = \text{CaCO}_3^0$	3.224	5
$\text{Mg}^{2+} + \text{H}_2\text{O} = \text{MgOH}^+ + \text{H}^+$	-11.44	5
$\text{Mg}^{2+} + \text{CO}_3^{2-} + \text{H}^+ = \text{MgHCO}_3^+$	11.399	5
$\text{Mg}^{2+} + \text{CO}_3^{2-} = \text{MgCO}_3^0$	2.98	5

$\text{Fe}^{3+} + \text{H}_2\text{O} = \text{FeOH}^{2+} + \text{H}^+$	-2.19	5
$\text{Fe}^{3+} + 2\text{H}_2\text{O} = \text{Fe}(\text{OH})_2^+ + 2\text{H}^+$	-5.67	5
$\text{Fe}^{3+} + 3\text{H}_2\text{O} = \text{Fe}(\text{OH})_3^0 + 3\text{H}^+$	-12.56	5
$\text{Fe}^{3+} + 4\text{H}_2\text{O} = \text{Fe}(\text{OH})_4^- + 4\text{H}^+$	-21.6	5
$2\text{Fe}^{3+} + 2\text{H}_2\text{O} = \text{Fe}_2(\text{OH})_2^{4+} + 2\text{H}^+$	-2.96	5
$3\text{Fe}^{3+} + 4\text{H}_2\text{O} = \text{Fe}_3(\text{OH})_4^{5+} + 4\text{H}^+$	-6.3	5

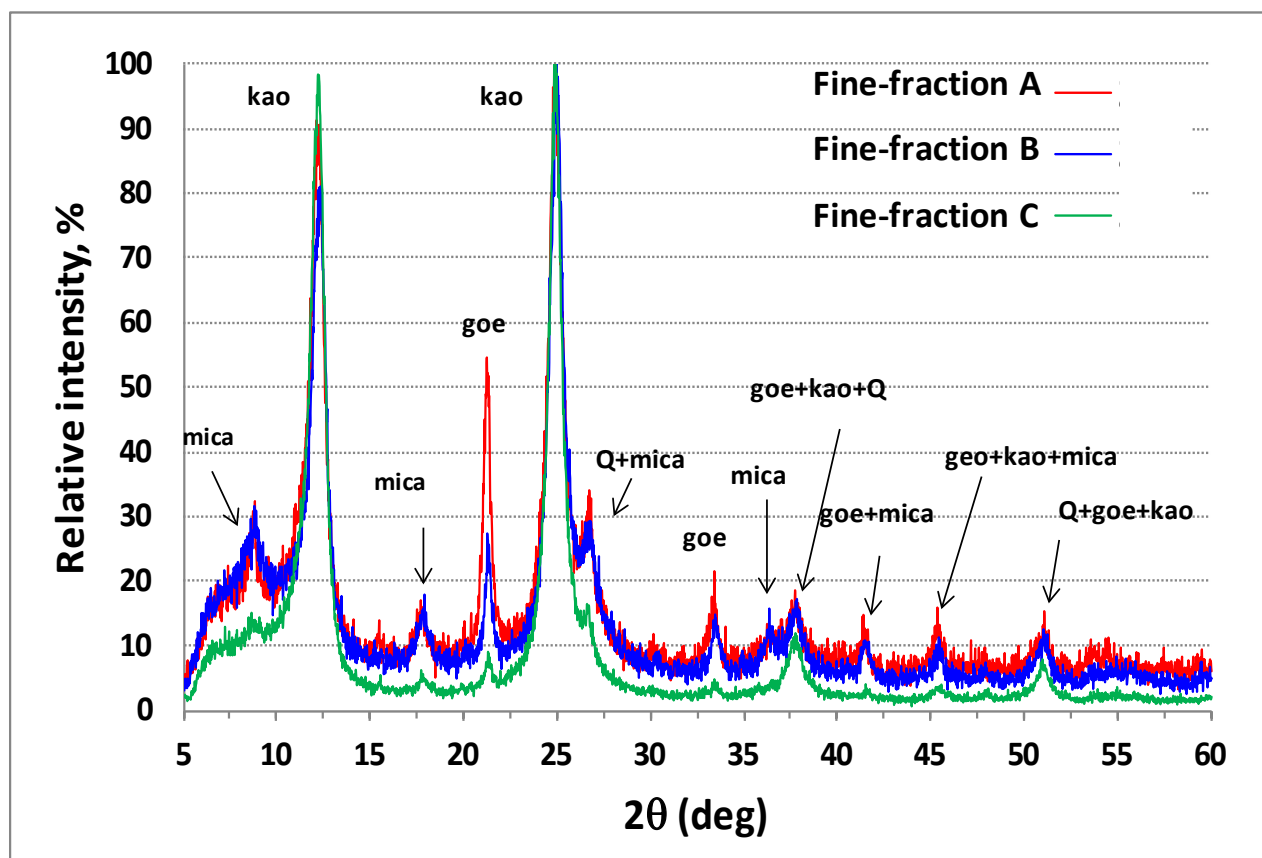


Fig.SI-1. Minerals were identified in the fine-fractions of plume sediments A, B and C using powder X-ray diffraction (XRD) (Rigaku SmartLab X-ray Diffractometer). It indicates that kaolinite and goethite are the major minerals with small amounts of mica and quartz.

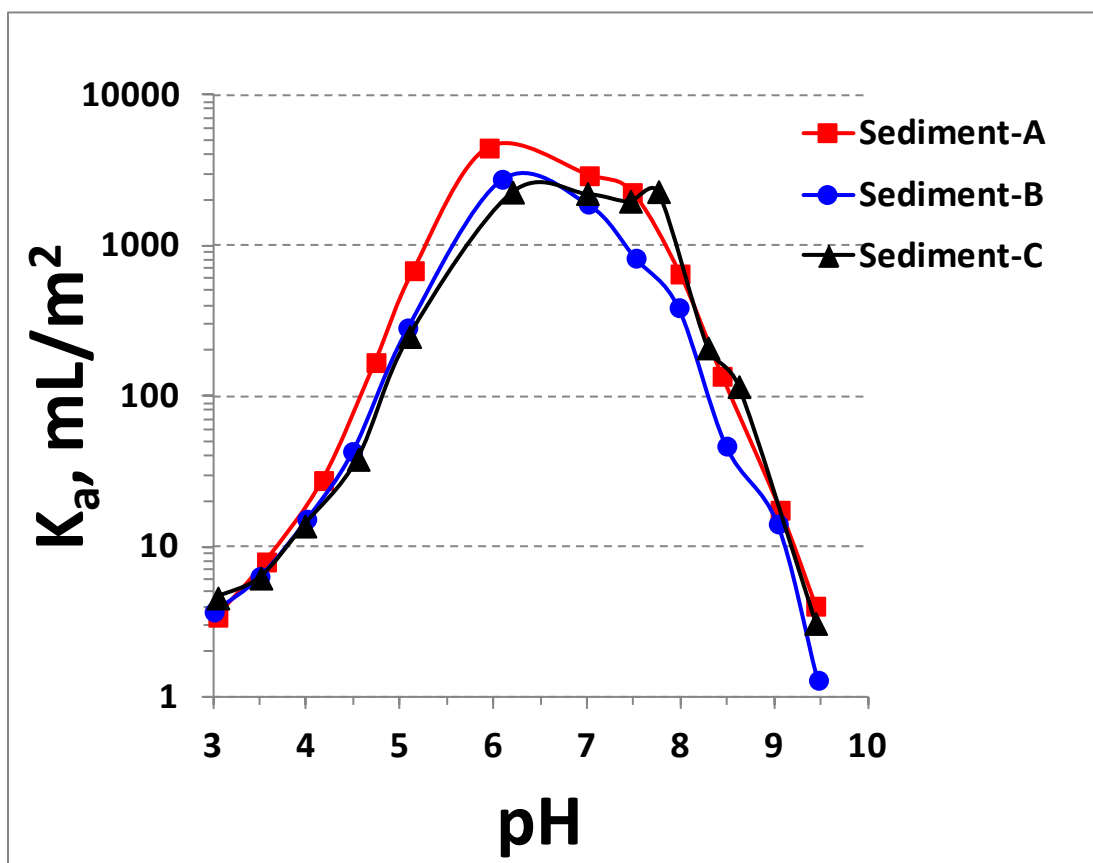


Figure SI-2. Comparison of U(VI) adsorption onto the quartz-sand dominated sediments (A, B and C) as a function of pH under conditions of 20g solid/L, 0.01 M NaNO₃, atmospheric CO₂ ($1 \times 10^{-3.5}$ atm) and ambient temperature (22.5 °C).

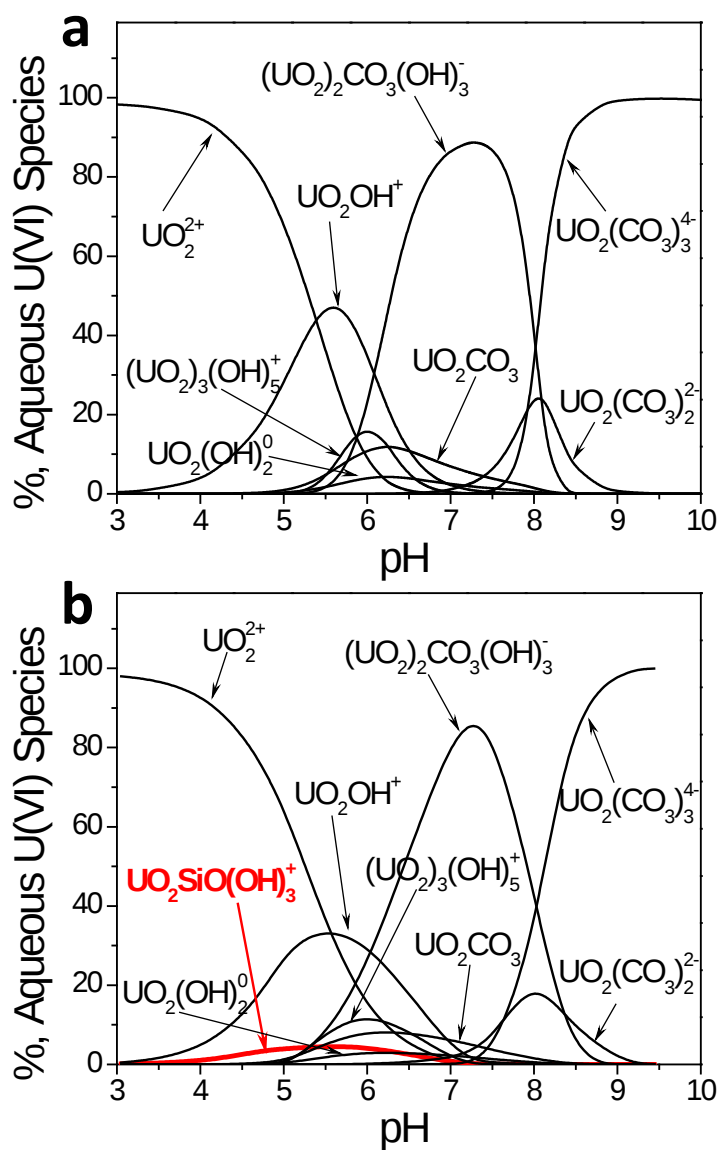


Figure SI-3. Aqueous U(VI) species distribution modeling in the absence of dissolved Si (**a**), and presence of dissolved Si in Table SI-3 sediment-A (**b**) under conditions of 1.0 μM U(VI), 0.01M NaNO_3 , atmospheric CO_2 ($10^{-3.5}$ atm) and 25 $^\circ\text{C}$.

Table SI-2. Measured equilibrium concentrations of coexisting ions (Al, Si, Na, Fe, Ca, and K) in the solutions of U(VI) adsorption experiments with bulk plume sediments A, B and C. The experiments were conducted as a function of pH (3.0 – 9.5) under conditions of a total added U(VI) = 1.0 μ M, an initially added [NaNO₃] = 0.01M, atmospheric CO₂(g) (P_{CO2} = 10^{-3.5} atm) and ambient temperature (22.5 °C).

Sediment A	pH	Al, mM	Si, mM	Na, mM	Fe, mM	Ca, mM	K, mM
	3.05	0.063	0.077	8.95	<0.0001	<0.001	0.097
	3.57	0.036	0.065	8.64	<0.0001	<0.001	0.026
	4.18	0.021	0.057	8.87	<0.0001	<0.001	0.014
	4.74	<0.001	0.053	9.01	<0.0001	<0.001	0.019
	5.15	<0.001	0.033	8.90	<0.0001	<0.001	0.025
	5.95	<0.001	0.038	9.33	<0.0001	<0.001	0.072
	7.02	<0.001	0.036	9.11	<0.0001	<0.001	0.073
	7.48	<0.001	0.027	9.38	<0.0001	<0.001	0.062
	7.99	<0.001	0.030	10.11	<0.0001	0.304	0.039
	8.44	<0.001	0.029	11.73	<0.0001	<0.001	0.051
	9.06	<0.001	0.027	17.56	<0.0001	<0.001	0.043
	9.44	0.001	0.027	37.84	<0.0001	<0.001	0.013
Sediment B	pH	Al, mM	Si, mM	Na, mM	Fe, mM	Ca, mM	K, mM
	3.01	0.054	0.057	9.83	<0.0001	<0.001	0.015
	3.50	0.041	0.048	9.84	<0.0001	<0.001	0.023
	4.00	0.017	0.029	9.68	<0.0001	<0.001	0.025
	4.49	0.004	0.024	9.56	<0.0001	<0.001	0.011
	5.08	<0.001	0.025	9.64	<0.0001	<0.001	0.026
	6.09	<0.001	0.018	9.82	<0.0001	<0.001	0.162
	7.01	<0.001	0.016	9.70	<0.0001	<0.001	0.029
	7.52	<0.001	0.015	10.32	<0.0001	<0.001	0.013
	7.98	<0.001	0.016	10.81	<0.0001	<0.001	0.033
	8.49	<0.001	0.014	12.48	<0.0001	<0.001	0.009
	9.04	0.004	0.014	17.28	<0.0001	<0.001	0.013
	9.47	0.007	0.018	40.10	<0.0001	<0.001	0.019

Table SI-2 (Continued)

Sediment C	pH	Al, mM	Si, mM	Na, mM	Fe, mM	Ca, mM	K, mM
	3.05	0.038	0.059	10.02	<0.0001	<0.001	0.009
	3.51	0.028	0.050	10.04	<0.0001	<0.001	0.046
	3.98	0.018	0.046	10.09	<0.0001	<0.001	0.021
	4.55	0.004	0.040	9.79	<0.0001	<0.001	0.028
	5.10	<0.001	0.027	10.00	<0.0001	<0.001	<0.001
	6.20	<0.001	0.021	9.55	<0.0001	<0.001	0.037
	7.00	<0.001	0.022	10.08	<0.0001	<0.001	0.016
	7.46	<0.001	0.026	10.20	<0.0001	0.001	<0.001
	7.76	<0.001	0.018	11.37	<0.0001	<0.001	<0.001
	8.29	<0.001	0.020	13.31	<0.0001	0.180	<0.001
	8.62	<0.001	0.020	16.45	<0.0001	<0.001	<0.001
	9.44	<0.001	0.024	39.44	<0.0001	<0.001	<0.001

Table SI-3. Solubility constants of selected minerals

Mineral	Reaction	log K (I = 0)	Ref.
Kaolinite	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4(\text{s}) + 6\text{H}^+ = \text{Al}^{3+} + \text{H}_4\text{SiO}_4(\text{aq}) + \text{H}_2\text{O}$	7.435	5
Quartz	$\text{SiO}_2(\text{s}) + 2\text{H}_2\text{O} = \text{Si}(\text{OH})_4(\text{aq})$	-3.98	5
Goethite	$\text{FeOOH}(\text{s}) + 3\text{H}^+ = \text{Fe}^{3+} + 2\text{H}_2\text{O}$	-1.0	5
Ferrihydrite (Amorphous to microcrystalline)	$\text{Fe}(\text{OH})_3(\text{s}) + 3\text{H}^+ = \text{Fe}^{3+} + 3\text{H}_2\text{O}$	3.0-5.0	5

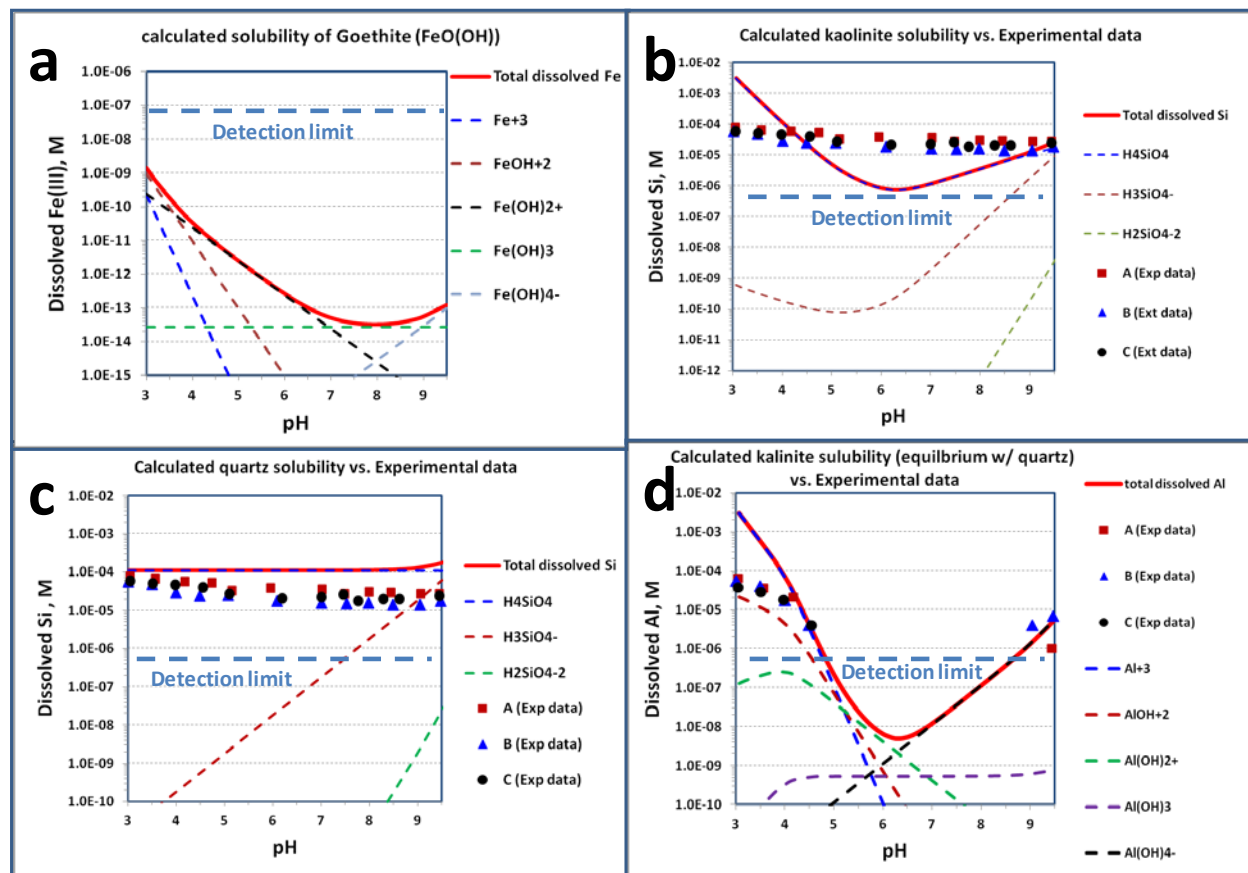


Fig.SI-4. Calculated solubility of goethite (a), kaolinite (b and d) and quartz (c) using the relevant reactions/constants in Table SI-1 and Table SI-4 under conditions of 0.01M NaNO_3 , 25 °C, and atmospheric CO_2 ($1 \times 10^{-3.5}$ atm).

Fig.SI-4a. The calculated solubility of goethite is less than 10^{-9} M Fe(III) under our experimental conditions of pH 3.0–9.5. It is consistent with our ICP-MS data, where the dissolved Fe was non-detectable using the calibration method defined effective detection limit ($\sim 10^{-7}$ M). It also suggests that no ferrihydrite phase exists in the sediments, otherwise the detected Fe should be significant high at acidic pH conditions.

Fig.SI-4b. Calculated solubility of Si for kaolinite is compared with the experimentally determined Si concentrations in batch sediments experiments. The shape of calculated total dissolved Si is inconsistent with that of experimentally measured data. It suggests that kaolinite is not the controlling phase for the dissolved Si.

Fig.SI-4c. Calculated solubility of Si for quartz is compared with the experimentally determined Si in batch sediments experiments. The shape of calculated dissolved Si concentrations is consistent with that of experimentally measured data, but all the experimentally determined data are consistently lower than the equilibrium solubility of quartz. The results indicate the dissolved Si

was mainly contributed by quartz, and solubility equilibrium was not achieved over the time frame of the experiments.

Fig.SI-4d. Calculated solubility of Al(III) for kaolinite in equilibrium with quartz is compared with the experimentally determined Al concentrations in batch sediments experiments. The experimentally determined concentrations are consistent with the calculated equilibrium concentrations at pH > 4.0, lower than the calculated equilibrium concentrations at pH < 4.0. In the pH range of 5.0 to 8.5, dissolved Al was non-detectable (effective detection limit $6 \times 10^{-7} \text{M}$), which is in good agreement with the calculated Al solubility for kaolinite. The lower determined Al data at pH < 4.0 suggest that dissolution has not reached equilibrium with kaolinite.

Surface Complexation Modeling (SCM) of Al(III) Adsorption onto Goethite and Silica

Modeling of Al(III) Adsorption onto Goethite. Lovgren et al.⁶ studied Al(III) adsorption onto goethite using the acid-base potentiometric titration method. Their experimental conditions are: [Al(III)] = 0.32 mM, goethite concentration = 11g/L, 0.100M NaNO₃, Ar atmosphere and 25°C. The specific surface area of goethite = 39.9 m²/g, and the total goethite surface sites concentration = 0.8mM. The experimental adsorption data were evaluated with a constant capacitance model (CCM). The surface reactions/constants proposed by Lovgren et al.⁶ are presented in Table SI-4a.

Lovgren et al.⁶ used the two pK model for surface protonation-deprotonation processes⁷ and applied the constant capacitance model (CCM) for surface complexation modeling (SCM) of Al(III) adsorption onto goethite. In this work, we use one pK model for surface protonation-deprotonation reaction⁷ and apply the diffusion layer model (DLM) for SCM. In order to incorporate the reactions/constants proposed by Lovgren et al.⁶ into our one pK and DLM models, we modified their reactions/constants and the details can be found in Table SI-4b.

The SCM of Al(III) adsorption onto goethite (Figure SI-5a) was completed using PHREEQC (v2) with the modified surface reactions (5) – (7) in Table SI-4b, the relevant aqueous reactions/constants in Table SI-1, and the DLM described in the modeling approach in the main text under the relevant experimental conditions used by Lovgren et al.⁶

It should be noted that the modified surface reactions/constants are not rigorously consistent with those of original ones. However, the modeling result indicates that the modified surface reactions/constants can describe the experimental data very well (Figure SI-5a), and can be used for modeling Al(III) adsorption onto the goethite at pH 3.0-7.0 under the relevant experimental conditions.

Modeling of Al(III) Adsorption onto Silica. Charlet et al.⁸ investigated Al(III) adsorption onto amorphous silica using acid-base potentiometric titration method. Their experimental conditions are: [Al(III)] = 0.0923 mM, silica concentration = 1.0g/L, 0.60M NaCl, Ar atmosphere and 25°C. The specific surface area of silica = 400 m²/g, the total silica surface sites concentration = 1.50mM. The experimental adsorption data were described with the constant capacitance model (CCM). The surface reactions/constants proposed by Charlet et al. are presented in Table SI-5a.

Charlet et al. used one pK model for silica surface deprotonation process and used the constant capacitance model (CCM) for SCM. We also use one pK model for silica surface deprotonation, but we use the diffusion layer model (DLM) for SCM. In order to incorporate the reactions/constants proposed by Charlet et al.⁸ into our one pK and DLM models, we modified their reactions/constants and the details can be found in Table SI-5b.

The SCM of Al(III) adsorption onto silica (Figure SI-5b) was completed using PHREEQC (v2) with the modified surface reactions (3) and (4) in Table SI-5b, the relevant aqueous

reactions/constants in Table SI-1, and the DLM described in the modeling approach in the main text under the relevant experimental conditions used by Charlet et al.⁸

It should be noted that the modified surface reactions/constants are not rigorously consistent with those of original ones. The modeling result indicates that the modified surface reactions/constants can be used for describing Al(III) adsorption onto the silica at pH 3.0-6.0 (Figure SI-5b) under the relevant experimental conditions.

Table SI-4. Surface reactions/constants proposed by Lovgren et al.⁶ are modified in this work for modeling Al(III) adsorption onto goethite (Figure SI-5a) under the relevant experimental conditions.

Modeling of Al(III) adsorption onto goethite	
A. Surface reactions/constants used by Lovgren et al.	Log K
(1) $>\text{FeOH} + \text{H}^+ = >\text{FeOH}_2^+$	7.47±0.02 ^a
(2) $>\text{FeOH} = >\text{FeO}^- + \text{H}^+$	-9.51±0.04 ^a
(3) $>\text{FeOH} + \text{Al}^{3+} + \text{H}_2\text{O} = (>\text{FeO})\text{Al}(\text{OH})^+ + 2\text{H}^+$	-1.49±0.04 ^a
(4) $>\text{FeOH} + \text{Al}^{3+} + 2\text{H}_2\text{O} = (>\text{FeO})\text{Al}(\text{OH})_2 + 3\text{H}^+$	-9.10±0.04 ^a
B. Modified surface reactions/constants used in this work^b (Figure SI-5a)	
(5) $>\text{FeOH}^{-0.5} + \text{H}^+ = >\text{FeOH}_2^{+0.5}$	9.18 ^b
(6) $>\text{FeOH}^{-0.5} + \text{Al}^{3+} + \text{H}_2\text{O} = (>\text{FeOH})\text{Al}(\text{OH})^{+1.5} + \text{H}^+$	7.00 ^c
(7) $>\text{FeOH}^{-0.5} + \text{Al}^{3+} + 2\text{H}_2\text{O} = (>\text{FeOH})\text{Al}(\text{OH})_2^{+0.5} + 2\text{H}^+$	-0.61 ^c
^a Lovgren et al. ⁶ ; ^b Obtained from Sherman et al. ⁹ for maintaining consistency with Table 3 in main text; ^c Estimated from Lovgren et al. by converting their two-pK (protonation-deprotonation) model to one-pK model ⁷ , i.e., $\text{Log K}(6) = \text{Log K}(3) + (\text{Log K}(1) - \text{Log K}(2))/2 = 7.00$, $\text{Log K}(7) = \text{Log K}(4) + (\text{Log K}(1) - \text{Log K}(2))/2 = -0.61$.	

Table SI-5. Surface reactions/constants proposed by Charlet et al.⁸ are modified in this work for modeling Al(III) adsorption onto silica (Figure SI-5b) under the relevant experimental conditions.

Modeling of Al(III) adsorption onto silica	
A. Surface reactions/constants used by Charlet et al.	Log K
(1) $>\text{QOH} = >\text{QO}^- + \text{H}^+$	6.97±0.09 ^a
(2) $>\text{QOH} + \text{Al}^{3+} + 3 \text{H}_2\text{O} = (>\text{QOH})\text{Al}(\text{OH})_3 + 3\text{H}^+$	-10.57 to -8.76 ^a
B. Modified surface reactions used in this work (Figure SI-4b)	
(3) $>\text{QOH} = >\text{QO}^- + \text{H}^+$	7.20 ^b
(4) $>\text{QOH} + \text{Al}^{3+} + 3 \text{H}_2\text{O} = (>\text{QOH})\text{Al}(\text{OH})_3 + 3\text{H}^+$	-9.00 ^c
^a Charlet et al.; ^b Obtained from Pabalan et al. ¹⁰ for maintaining consistency with Table 3; ^c Selected from Charlet et al. ⁸	

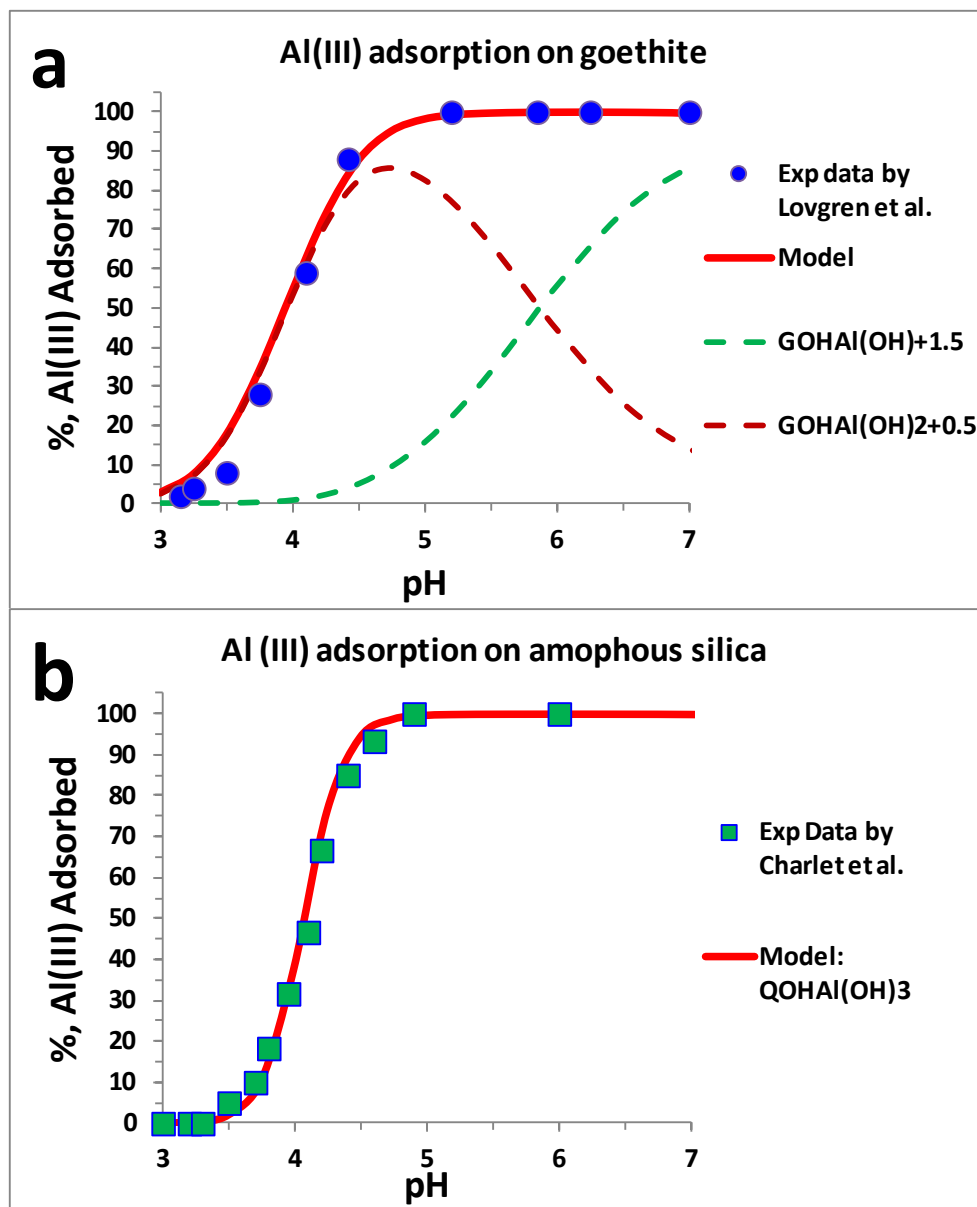


Figure SI-5. Surface complexation modeling of Al(III) adsorption onto (a) goethite and (b) silica (an analogue of quartz), with the modified surface reactions in Tables SI-4b and SI-5b, the relevant aqueous reactions/constants in Table SI-1, and the DLM described in the modeling approach in the main text under the relevant experimental conditions used by Lovgren et al.⁶ and by Charlet et al.,⁸ respectively. The experimental data were estimated from Lovgren et al.⁶ for Al(III) adsorption with goethite,⁶ and from Charlet et al. for Al(III) adsorption with silica.⁸

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PHREEQC input file for modeling U(VI) adsorption to sediment A

DATABASE (include all relevant aqueous and phases reactions and constants in a large PHREEQC database)

TITLE: Modeling of uranium(VI) adsorption to quartz-sand dominated sediment

SURFACE_MASTER_SPECIES

G	GOH-0.5	# GOH = goethite site
Ks	KsOH-0.5	# KsOH = kaolinite edge site
Q	QOH	# QOH = quartz site

SURFACE_SPECIES

Goethite

GOH-0.5 = GOH-0.5

log_k 0.0

GOH-0.5 + H+ = GOH2+0.5

log_k 9.18

2 GOH-0.5 + UO2+2 = (GOH)2UO2+

log_k 14.11

2 GOH-0.5 + UO2+2 + CO2 + H2O = (GOH)2UO2CO3- + 2 H+

log_k 4.35

2 GOH-0.5 + CO2 = (GO)2CO- + H2O

log_k 5.93

GOH-0.5 + CO2 + Na+ = GOCO2Na-0.5 + H+

log_k -3.02

GOH-0.5 + Al+3 + H2O = GOHAl(OH)+1.5 + H+

log_k 7.00

GOH-0.5 + Al+3 + 2 H2O = GOHAl(OH)2+0.5 + 2 H+

log_k -0.61

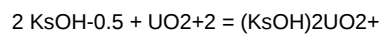
Kaolinite edge site

KsOH-0.5 = KsOH-0.5

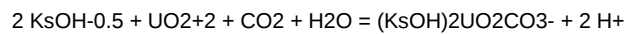
log_k 0.0

KsOH-0.5 + H+ = KsOH2+0.5

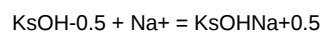
log_k 4.9



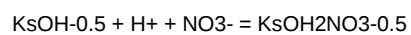
log_k 5.3



log_k -0.1

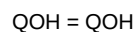


log_k -2.1

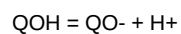


log_k 4.9

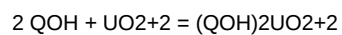
Quartz



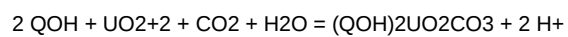
log_k 0.0



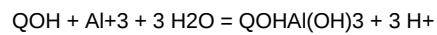
log_k -7.20



log_k 5.12



log_k 0.26



log_k -9.00

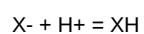
EXCHANGE_MASTER_SPECIES

X X- # X- = kaolinite exchange site

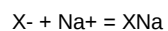
EXCHANGE_SPECIES



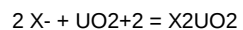
log_k 0.0



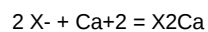
log_k 4.5



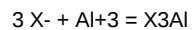
log_k 2.9



log_k 7.1



log_k 6.8



log_k 8.0

SURFACE 1

GOH-0.5 4.2e-5 38.5 0.22 # total number of the site in moles; specific surface area (m²/g); mass (g)

-diffuse_layer 1e-8 # thickness (m) of diffuse layer

-only_counter_ions

KsOH-0.5 6.0e-5 35.4 0.44

-diffuse_layer 1e-8

-only_counter_ions

QOH 1.47e-5 0.20 19.24

-diffuse_layer 1e-8

-only_counter_ions

EXCHANGE 1

X- 7.4e-6 # total X- exchange site in moles

SOLUTION 1

-units mmol/kgw

pH 3.05

U(+6) 0.00114 # include added 1uM U(VI) and the carbonate-extractable U from sediment-A (0.14uM)

Na 10 charge # when equilibrium with CO₂ (g) at higher pH, dissolved (bi)carbonate is balanced by Na

N(5) 10

Ca 0.000

Al 0.0631 # equilibrium 0.063mM

Si 0.077

SOLUTION 2

-units mmol/kgw

pH 3.57

U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.0365 # equilibrium 0.036mM
Si	0.065

SOLUTION 3

-units	mmol/kgw
pH	4.18
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.0228 # equilibrium 0.021mM
Si	0.057

SOLUTION 4

-units	mmol/kgw
pH	4.74
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000
Si	0.053

SOLUTION 5

-units	mmol/kgw
pH	5.15
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000
Si	0.033

SOLUTION 6

-units	mmol/kgw
pH	5.95
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000
Si	0.038

SOLUTION 7

-units	mmol/kgw
pH	7.02
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000
Si	0.036

SOLUTION 8

-units	mmol/kgw
pH	7.48
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.004
Al	0.000
Si	0.027

SOLUTION 9

-units	mmol/kgw
pH	7.99
U(+6)	0.00114
Na	10 charge
N(5)	10.00
Ca	0.304
Al	0.000
Si	0.030

SOLUTION 10

-units	mmol/kgw
pH	8.44
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000
Si	0.029

SOLUTION 11

-units	mmol/kgw
pH	9.06
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.000

Si	0.027
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SOLUTION 12

-units	mmol/kgw
pH	9.44
U(+6)	0.00114
Na	10 charge
N(5)	10
Ca	0.000
Al	0.001
Si	0.027

PHASES

Fix_H+
H+ = H+
log_k 0.0

END

SELECTED_OUTPUT

-file Sediment-A.sel

#

USE solution 1

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-3.05	NaOH	10.0
CO2(g)	-3.5	# equilibrium with air CO2	

END

USE solution 2

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-3.57	NaOH	10.0
CO2(g)	-3.5		

END

USE solution 3

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-4.18	NaOH	10.0
CO2(g)	-3.5		

END

USE solution 4

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-4.74	NaOH	10.0
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CO2(g)	-3.5
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END

USE solution 5

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-5.15	NaOH	10.0
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CO2(g)	-3.5
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END

USE solution 6

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-5.95	NaOH	10.0
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CO2(g)	-3.5
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END

USE solution 7

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-7.02	NaOH	10.0
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CO2(g)	-3.5
--------	------

END

USE solution 8

USE surface 1

USE EXCHANGE 1

EQUILIBRIUM_PHASES 1

Fix_H+	-7.48	NaOH	10.0
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CO2(g)	-3.5
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END

USE solution 9

USE surface 1
USE EXCHANGE 1
EQUILIBRIUM_PHASES 1
 Fix_H+ -7.99 NaOH 10.0
 CO2(g) -3.5
END

USE solution 10
USE surface 1
USE EXCHANGE 1
EQUILIBRIUM_PHASES 1
 Fix_H+ -8.44 NaOH 10.0
 CO2(g) -3.5
END

USE solution 11
USE surface 1
USE EXCHANGE 1
EQUILIBRIUM_PHASES 1
 Fix_H+ -9.06 NaOH 10.0
 CO2(g) -3.5
END

USE solution 12
USE surface 1
USE EXCHANGE 1
EQUILIBRIUM_PHASES 1
 Fix_H+ -9.44 NaOH 10.0
 CO2(g) -3.5
END