Supporting Information for

Geochemical Modeling of Reactions and Partitioning of Trace Metals and Radionuclides during Titration of Contaminated Acidic Sediments

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Aqueous complexation reactions

Table S1. Aqueous complexation reactions considered in the model

Aqueous Complexation Reaction	log K	References
$H_2O = H^+ + OH^-$	-14.0	*(1)*
$H_2CO_3^0 = H^+ + HCO_3^-$	-6.352	*(1)*
$HCO_3^- = H^+ + CO_3^{2-}$	-10.329	*(1)*
$H^+ + SO_4^{2-} = HSO_4^-$	1.988	*(1)*
$Na^+ + H_2O = NaOH^0 + H^+$	-14.18	*(1)*
$Na^+ + HCO_3^- = NaHCO_3^0$	-0.25	*(1)*
$Na^+ + CO_3^{2-} = NaCO_3^-$	1.27	*(1)*
$Na^+ + SO_4^{2-} = NaSO_4^-$	0.70	*(1)*
$K^+ + H_2O = KOH^0 + H^+$	-14.46	*(1)*
$K^+ + SO_4^{2-} = KSO_4^-$	0.85	*(1)*
$Ca^{2+} + H_2O = CaOH^+ + H^+$	-12.78	*(1)*
$Ca^{2+} + HCO_3^- = CaHCO_3^+$	1.106	*(1)*
$Ca^{2+} + CO_3^{2-} = CaCO_3^0$	3.224	*(1)*
$Ca^{2+} + SO_4^{2-} = CaSO_4^0$	2.30	*(1)*
$Mg^{2+} + H_2O = MgOH^+ + H^+$	-11.44	*(1)*
$Mg^{2+} + HCO_3^- = MgHCO_3^+$	1.07	*(1)*
$Mg^{2+} + CO_3^{2-} = MgCO_3^0$	2.98	*(1)*
$Mg^{2+} + SO_4^{2-} = MgSO_4^0$	2.37	*(1)*
$Sr^{2+} + H_2O = SrOH^+ + H^+$	-13.29	*(1)*
$Sr^{2+} + HCO_3^- = SrHCO_3^+$	1.18	*(1)*
$Sr^{2+} + CO_3^{2-} = SrCO_3^0$	2.81	*(1)*
$Sr^{2+} + SO_4^{2-} = SrSO_4^0$	2.29	*(1)*
$Mn^{2+} + H_2O = MnOH^+ + H^+$	-10.59	*(1)*
$Mn^{2+} + HCO_3^- = MnHCO_3^+$	1.95	*(1)*
$Mn^{2+} + CO_3^{2-} = MnCO_3^0$	4.90	*(1)*
$Mn^{2+} + SO_4^{2-} = MnSO_4^0$	2.25	*(1)*

$Mn^{2+} + Cl^- = MnCl^+$	0.61	*(1)*
$Mn^{2+} + 2Cl^- = MnCl_2^0$	0.25	*(1)*
$Mn^{2+} + 3Cl^- = MnCl_3^-$	-0.31	*(1)*
$Fe^{3+} + H_2O = FeOH^{2+} + H^+$	-2.19	*(1)*
$Fe^{3+} + 2H_2O = Fe(OH)_2^+ + 2H^+$	-5.67	*(1)*
$Fe^{3+} + 3H_2O = Fe(OH)_3^0 + 3H^+$	-12.56	*(1)*
$Fe^{3+} + 4H_2O = Fe(OH)_4^- + 4H^+$	-21.6	*(1)*
$2Fe^{3+} + 2H_2O = Fe_2(OH)_2^{4+} + 2H^+$	-2.95	*(1)*
$3Fe^{3+} + 4H_2O = Fe_3(OH)_4^{5+} + 4H^+$	-6.3	*(1)*
$Fe^{3+} + Cl^- = FeCl^{2+}$	1.48	*(1)*
$Fe^{3+} + 2Cl^- = FeCl_2^+$	2.13	*(1)*
$Fe^{3+} + 3Cl^- = FeCl_3^0$	1.13	*(1)*
$Fe^{3+} + SO_4^{2-} = FeSO_4^+$	4.04	*(1)*
$Fe^{3+} + 2SO_4^{2-} = Fe(SO_4)_2^-$	5.38	*(1)*
$Fe^{3+} + HSO_4^- = FeHSO_4^{2+}$	2.48	*(1)*
$Al^{3+} + H_2O = AlOH^{2+} + H^+$	-5.00	*(1)*
$Al^{3+} + 2H_2O = Al(OH)_2^+ + 2H^+$	-10.1	*(1)*
$Al^{3+} + 3H_2O = Al(OH)_3^0 + 3H^+$	-16.9	*(1)*
$Al^{3+} + 4H_2O = Al(OH)_4^- + 4H^+$	-22.7	*(1)*
$Al^{3+} + SO_4^{2-} = AlSO_4^+$	3.02	*(1)*
$Al^{3+} + 2SO_4^{2-} = Al(SO_4)_2^-$	4.92	*(1)*
$Al^{3+} + HSO_4^- = AlHSO_4^{2+}$	0.46	*(1)*
$Si(OH)_4^0 = SiO(OH)_3^- + H^+$	-9.83	*(1)*
$Si(OH)_4^0 = SiO_2(OH)_2^{2-} + 2H^+$	-23.0	*(1)*
$UO_2^{2+} + H_2O = UO_2OH^+ + H^+$	-5.25	*(2)*
$UO_2^{2+} + 2H_2O = UO_2(OH)_2^0 + 2H^+$	-12.15	*(2)*
$UO_2^{2+} + 3H_2O = UO_2(OH)_3^- + 3H^+$	-20.25	*(2)*
$UO_2^{2+} + 4H_2O = UO_2(OH)_4^{2-} + 4H^+$	-32.4	*(2)*
$2UO_2^{2+} + H_2O = (UO_2)_2OH^{3+} + H^+$	-2.7	*(2)*

$2UO_2^{2+} + 2H_2O = (UO_2)_2(OH)_2^{2+} + 2H^+$	-5.62	*(2)*
$3UO_2^{2+} + 4H_2O = (UO_2)_3(OH)_4^{+} + 4H^+$	-11.9	*(2)*
$3UO_2^{2+} + 5H_2O = (UO_2)_3(OH)_5^{+} + 5H^+$	-15.55	*(2)*
$3UO_2^{2+} + 7H_2O = (UO_2)_3(OH)_7^{-} + 7H^+$	-32.2	*(2)*
$4UO_2^{2+} + 7H_2O = (UO_2)_4(OH)_7^{+} + 7H^+$	-21.9	*(2)*
$UO_2^{2+} + CO_3^{2-} = UO_2CO_3^{0}$	9.94	*(2)*
$UO_2^{2+} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$	16.61	*(2)*
$UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$	21.84	*(2)*
$UO_2^{2+} + 6CO_3^{2-} = UO_2(CO_3)_6^{6-}$	54.0	*(2)*
$2UO_2^{2+} + CO_3^{2-} + 3H_2O = (UO_2)_2CO_3(OH)_3^{-} + 3H^+$	-0.861	*(2)*
$3UO_2^{2+} + CO_3^{2-} + 3H_2O = (UO_2)_3O(OH)_2HCO_3^{+} + 3H^+$	0.649	*(2)*
$11UO_2^{2+} + 6\ CO_3^{2-} + 12H_2O = (UO_2)_{11}(CO_3)_6(OH)_{12}^{2-} + 12H^+$	36.394	*(2)*
$UO_2^{2+} + NO_3^{-} = UO_2NO_3^{+}$	0.300	*(2)*
$UO_2^{2+} + SO_4^{2-} = UO_2SO_4^{0}$	3.15	*(2)*
$UO_2^{2+} + 2SO_4^{2-} = UO_2(SO_4)_2^{2-}$	4.14	*(2)*
$UO_2^{2+} + Cl^- = UO_2Cl^+$	0.17	*(2)*
$UO_2^{2+} + 2Cl^- = UO_2Cl_2^{0}$	-1.1	*(2)*

Precipitation/dissolution reactions

Table S2. Solid phases and solubility in the equilibrium reaction path model.

Reactions	**logK**	**References**
$Al(OH)_{3(s)}$ (Gibbsite) $+ 3H^+ = Al^{3+} + 3H_2O$	8.11	*(1)*
$AlSO_4(OH) \cdot 5H_2O_{(s)}$ (Jurbanite) $+ H^+ = Al^{3+} + SO_4^{2-} + 6H_2O$	-3.2	*(3)*
$Al_4SO_4(OH)_{10} \cdot 5H_2O_{(s)}$ (Basaluminite) $+ 10H^+ = 4Al^{3+} + SO_4^{2-} + 15H_2O$	22.5	*(3)*
$SiO_{2(s)}$ (Amorphous silica) $+ 2H_2O = Si(OH)_4^{0}$	-2.71	*(1)*
$Al_2Si_2O_5(OH)_{4(s)}$ (Kaolinite) $+ 6H^+ = 2Al^{3+} + 2Si(OH)_4^{0} + H_2O$	7.435	*(1)*
$Ca(OH)_{2(s)}$ (Portlandite) $+ 2H^+ = Ca^{2+} + 2H_2O$	22.8	*(1)*
$CaCO_3 \cdot H_2O_{(s)}$ (Monohydrocalcite) $= Ca^{2+} + CO_3^{2-} + H_2O$	-7.60	*(4)*

$CaSO_4 \cdot 2H_2O_{(s)}$ (Gypsum) = $Ca^{2+} + SO_4^{2-} + 2H_2O$	-4.58	*(1)*
$Mg(OH)_{2(s)}$ (Brucite) + $2H^+ = Mg^{2+} + 2H_2O$	16.84	*(1)*
$MgCO_{3(s)}$ (Magnesite) = $Mg^{2+} + CO_3^{2-}$	-7.5	*(1)*
$CaMg(CO_3)_{2(s)}$ (Disordered dolomite)= $Ca^{2+} + CO_3^{2-}$	-16.54	*(4)*
$Mn(OH)_{2(s)}$ (Pyrolusite)+ $2H^+ = Mn^{2+} + 2H_2O$	15.2	*(1)*
$MnCO_{3(s)}$ (Synthetic rhodocrosite)= $Mn^{2+} + CO_3^{2-}$	-10.39	*(4)*
$Ni(OH)_{2(s)}$ (Theophrastite) + $2H^+ = Ni^{2+} + 2H_2O$	10.8	*(4)*
$NiCO_{3(s)} + H^+ = Ni^{2+} + HCO_3^-$	3.512	*(4)*
$UO_2CO_{3(s)}$ (Rutherfordine)+ $H^+ = UO_2^{2+} + HCO_3^-$	-4.143	*(4)*
$CaUO_{4(s)} + 4H^+ = Ca^{2+} + UO_2^{2+} + 2H_2O$	15.931	*(4)*
$UO_3 \cdot 2H_2O_{(s)}$ (Schoepite) + $2H^+ = UO_2^{2+} + 3H_2O$	4.812	*(4)*
$(UO_2)_2SiO_4 \cdot 2H_2O_{(s)}$ (Soddyite)+ $4H^+ = 2UO_2^{2+} + Si(OH)_4^0 + 2H_2O$	5.74	*(5)*
$Ca(UO_2)_2(SiO_3OH)_2 \cdot 5H_2O_{(s)}$ (Uranophane)+ $6H^+ = Ca^{2+} + 2UO_2^{2+} + 2Si(OH)_4^0 + 5H_2O$	9.42	*(5)*
$Na_2(UO_2)_2(Si_2O_5)_3 \cdot 5H_2O_{(s)}$ (Na-weeksite)+ $6H^+ + 5H_2O = 2Na^+ + 2UO_2^{2+} + 6Si(OH)_4^0$	1.51	*(5)*
$Co(OH)_{2(s)} + 2H^+ = Co^{2+} + 2H_2O$	12.1	*(4)*
$CoCO_{3(s)}$ (Sphaerocobaltite) + $H^+ = Co^{2+} + HCO_3^-$	-0.233	*(4)*
$SrCO_{3(s)}$ (Strontianite)= $Sr^{2+} + CO_3^{2-}$	-9.271	*(1)*
$SrSO_{4(s)}$ (Celestite)= $Sr^{2+} + SO_4^{2-}$	-6.63	*(1)*
$FeCo_{0.1}(OH)_{3.2(s)} + 3.2H^+ = Fe^{3+} + 0.1Co^{2+} + 3.2H_2O$	5.7	*(4)*

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