The influence of natural organic matter on radionuclide mobility under conditions relevant to cementitious disposal of radioactive wastes: a review of direct evidence

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Supporting Information

The following data are contained within this section:

Figure S1a & b and Table S1. Data showing the increasing organic complexation of neptunyl(V) with organic acids as pH increases. The table details the slope and correlation data for each of the datasets.

Figures S2a & b. Data for uranyl(VI) complexation with organic acids at pH 9 and 10 in the presence of carbonate. Data from Glaus et al. (1997).

Figure S3a -S3g. High pH ternary system data collated from the available literature.

Figure S4. High pH ternary system data in equilibrium with atmospheric CO_2.

Figure S5. High pH ternary system data where the presence of organic acids did not affect sorption.

Data used to plot figures S3, S4 and S5 are included as supporting information in a separate .csv file.

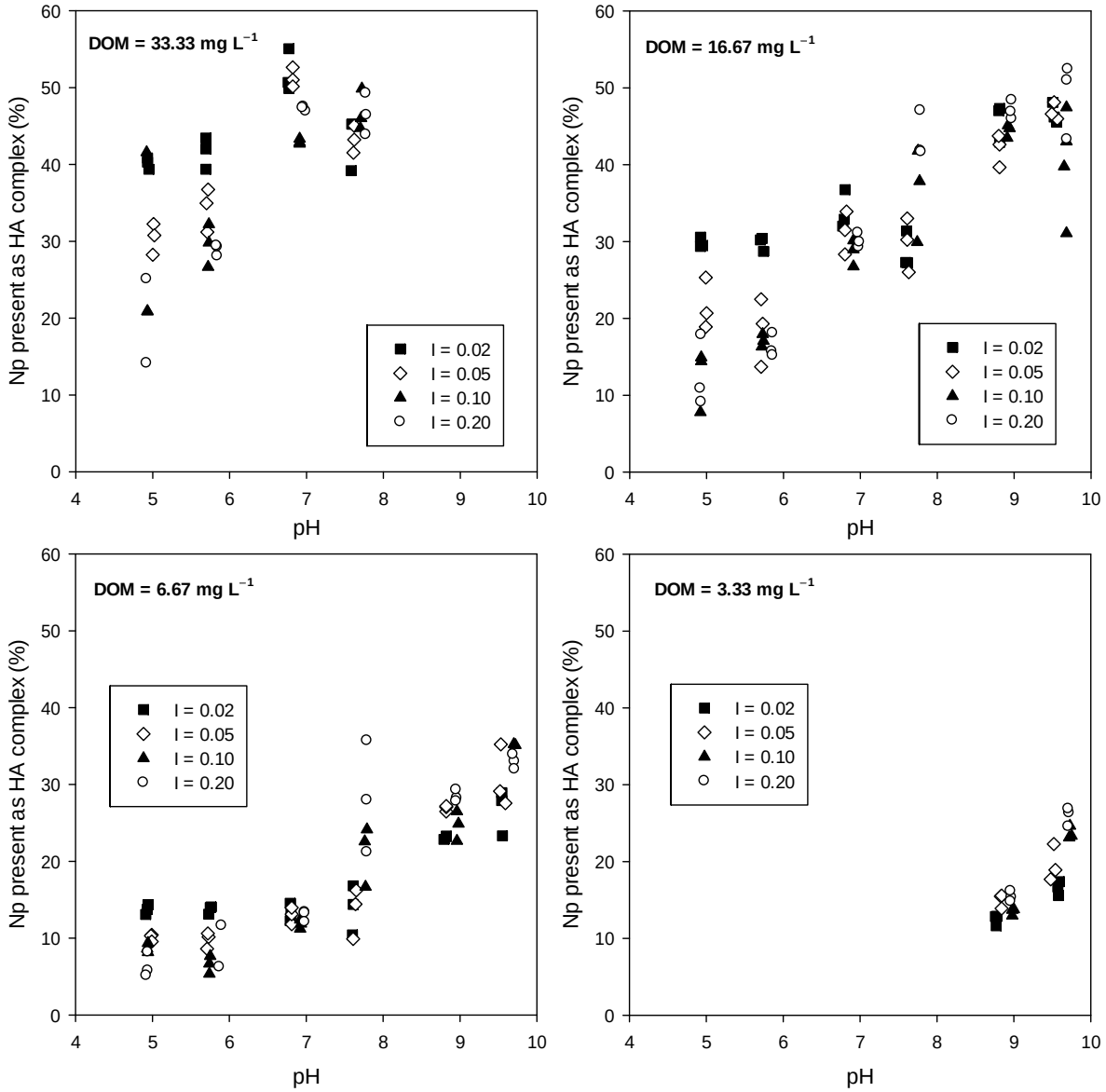

Figure S1a. Neptunium(V) complexation with purified Aldrich humic acid. Data from Glaus et al. (1997; I = mol L^{-1}; Np(V) = 1.2×10^{-8} mol L^{-1}).

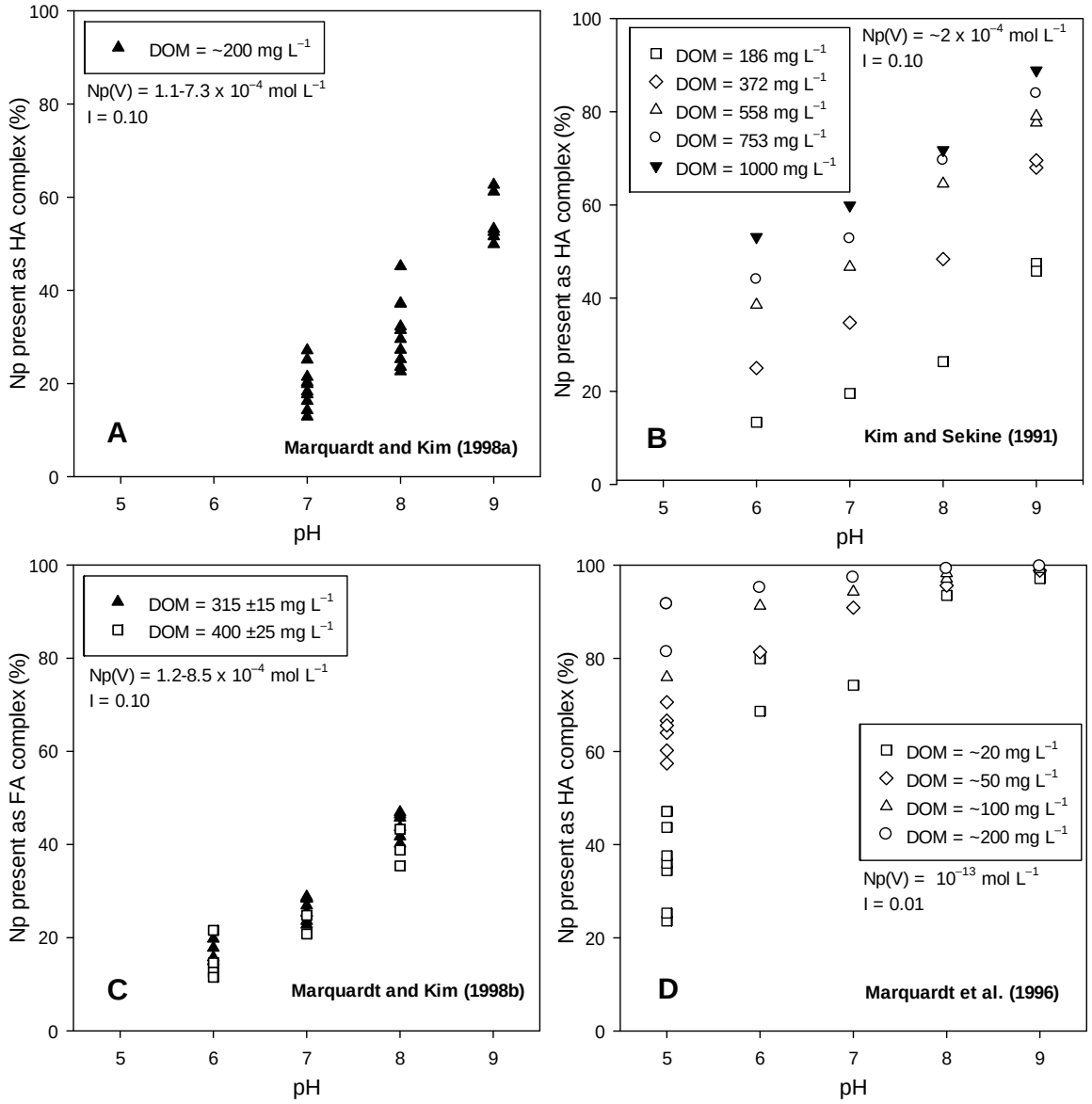

Figure S1b. Neptunium(V) complexation with Gorleben humic acid (A and B), Gorleben fulvic acid (C) and purified Aldrich humic acid (D), (I = mol L^{-1}).

Table S1. Slope (m; change in percent bound per pH unit) and R^2 data for the data in Figure S1 assuming a linear gradient (I = mol L^{-1}).

Reference	[HA] (mg L^{-1})	I	m	R^2	*n*
Glaus et al. (1997)	33.33	0.02	2	0.22	12
		0.05	6	0.61	12
		0.10	8	0.69	12
		0.20	10	0.86	11
	16.67	0.02	4	0.68	18
		0.05	6	0.86	18
		0.10	7	0.83	19
		0.20	8	0.91	18
	6.67	0.02	3	0.68	18
		0.05	5	0.82	18
		0.10	5	0.87	17
		0.20	6	0.84	17
	3.33	0.02	5	0.91	6
		0.05	7	0.73	6
		0.10	14	0.99	6
		0.20	14	0.98	6
Marquardt & Kim (1998a)	~200	0.10	17	0.82	26
Kim & Sekine (1991)	186	0.10	11	0.93	5
	372	0.10	15	0.98	5
	558	0.10	14	0.99	5
	753	0.10	13	0.99	5
	1000	0.10	12	0.96	4
Marquardt & Kim (1998b) (Fulvic acid)	315 ±15	0.10	14	0.92	19
	400 ±25	0.10	12	0.88	9
Marquardt et al. (1996)	~20	0.01	17	0.81	13
	~50	0.01	10	0.89	10
	~100	0.01	5	0.82	6
	~200	0.01	3	0.64	7

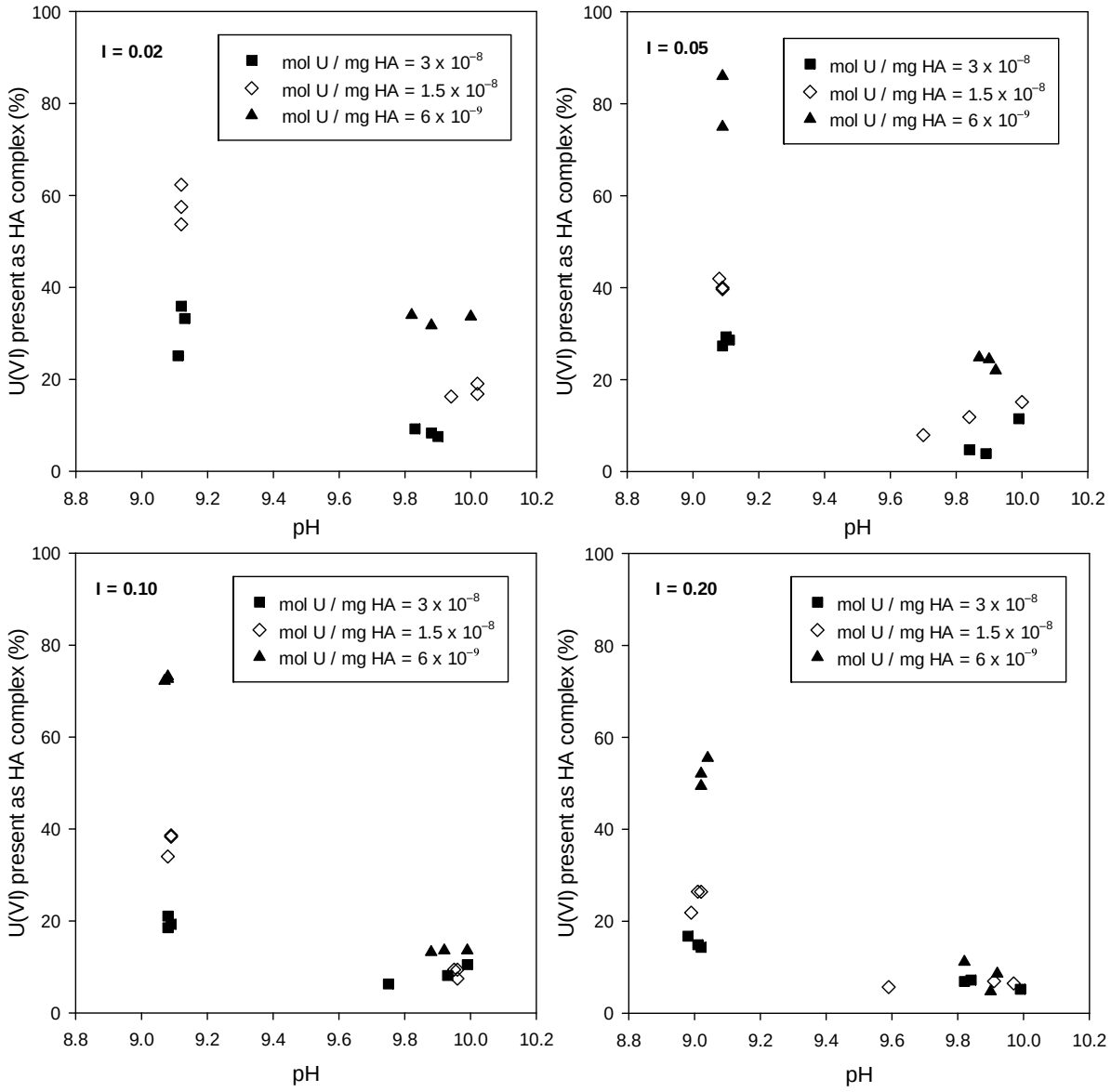

Figure S2a. Uranyl complexation with purified Aldrich humic acid at high pH (data from Glaus et al., 1997). Carbonate concentration = 2×10^{-4} mol l^{-1}, I = mol L^{-1}.

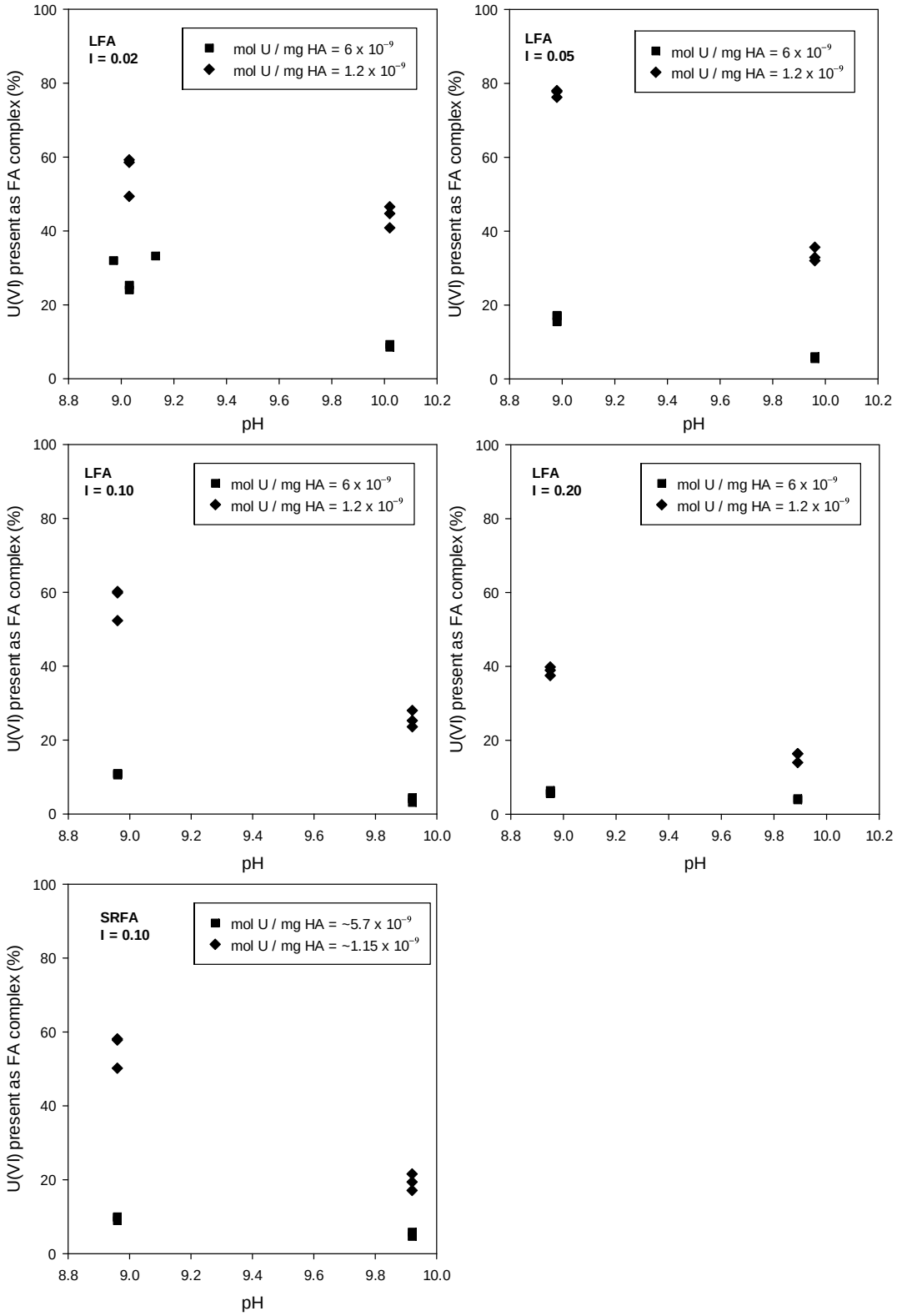

Figure S2b. Uranyl complexation with purified Laurentian soil fulvic acid (LFA) and Suwannee River fulvic acid (SRFA) at high pH (data from Glaus et al., 1997). Carbonate concentration = 1.5×10^{-3} mol l^{-1}.(pH ~9) and 1×10^{-3} mol l^{-1}.(pH ~10), (I = mol L^{-1}).

Supplementary ternary system data

The following figures illustrate all of the reviewed ternary system data as included in Table 2 of the main manuscript. Data are obtained from digitisation of figures in the published literature as cited. Readers are referred to the source publications for further experimental details and data for lower pH values. Figure S3 details ternary system data for experiments under nitrogen atmosphere or where the atmospheric conditions are not reported. Figure S4 details ternary system data for experiments where the pCO_2 was in equilibrium with the atmosphere. Figure S5 detail those systems where the presence of organic acids did not affect sorption.

- Filled data points show binary radionuclide – mineral sorption.
- Open data points show radionuclide – mineral sorption in the presence of organic acids (HA and/or FA).
- Fulvic acid data are denoted with a centre dot in the symbol.
- Where experiments have been performed on multiple concentrations of HA/FA these are denoted by decreasing greyscale intensity as the concentration increases.
- The legend contains; reference, radionuclide concentration, HA/FA concentration, mass to volume ratio of the mineral (m/v), and ionic strength (mol L^{-1}).
- Where other system components were varied the definitions for the symbols used are defined in the figure legends.

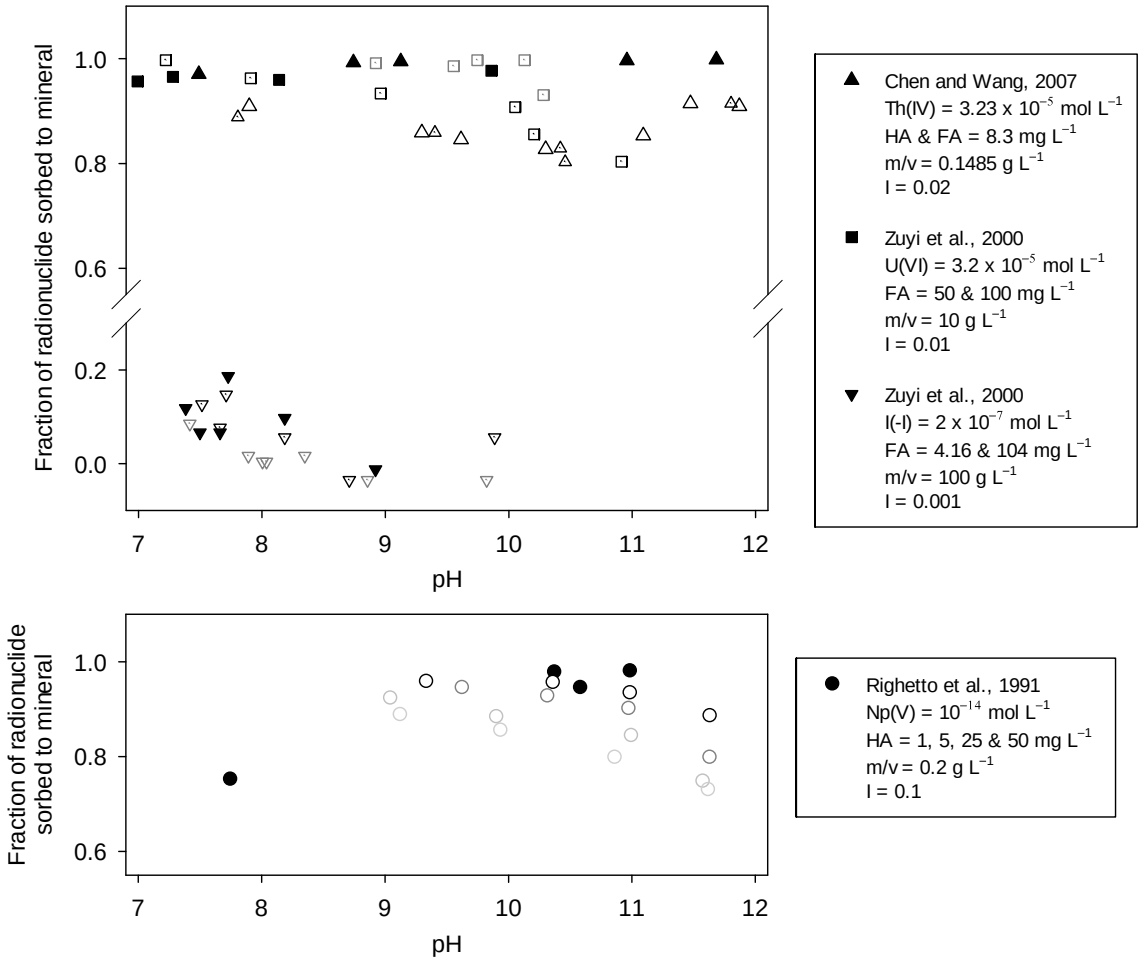

Figure S3a. Ternary system data for γ-Al_2O_3.

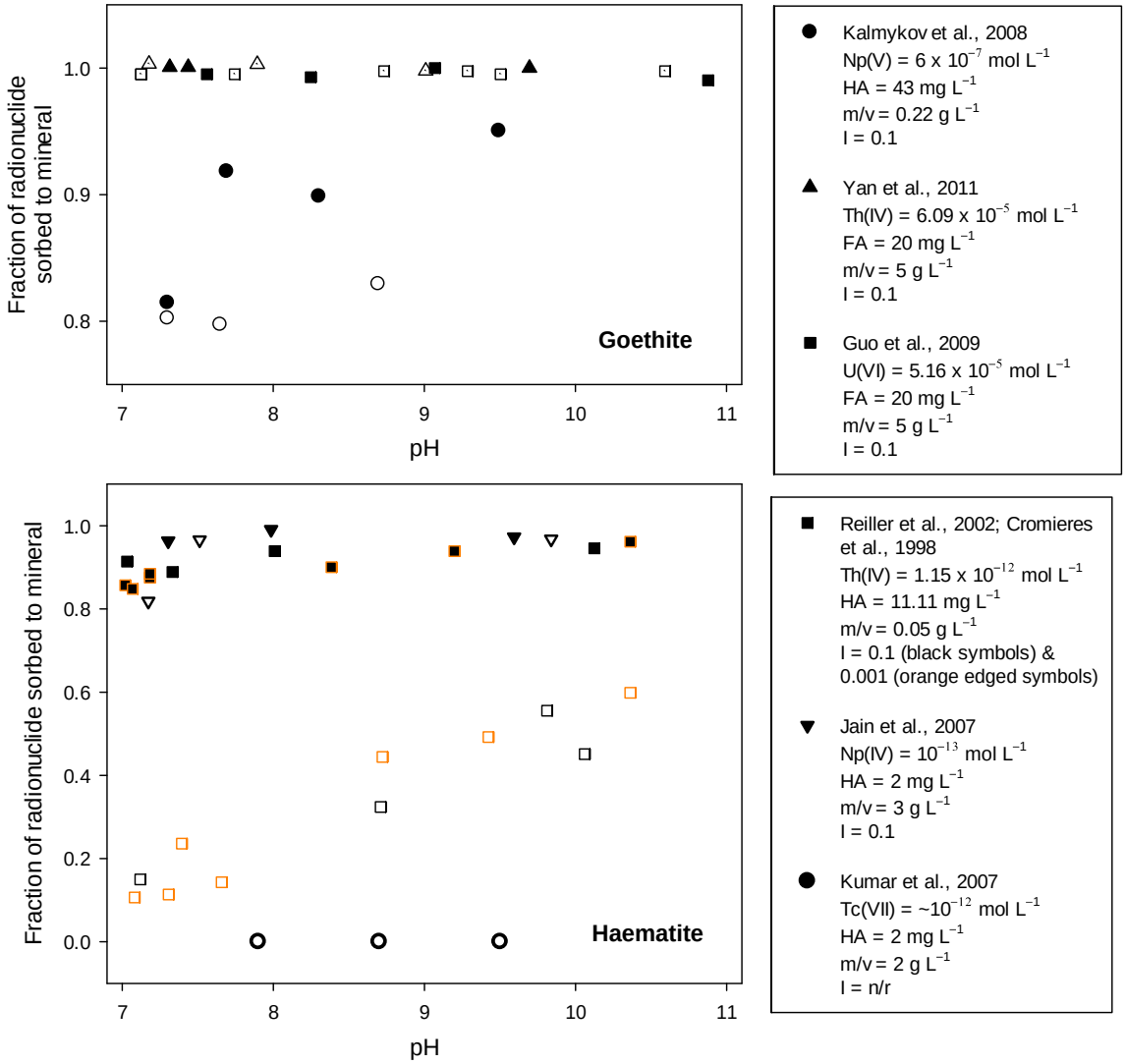

Figure S3b. Ternary system data for the iron oxides goethite and haematite.

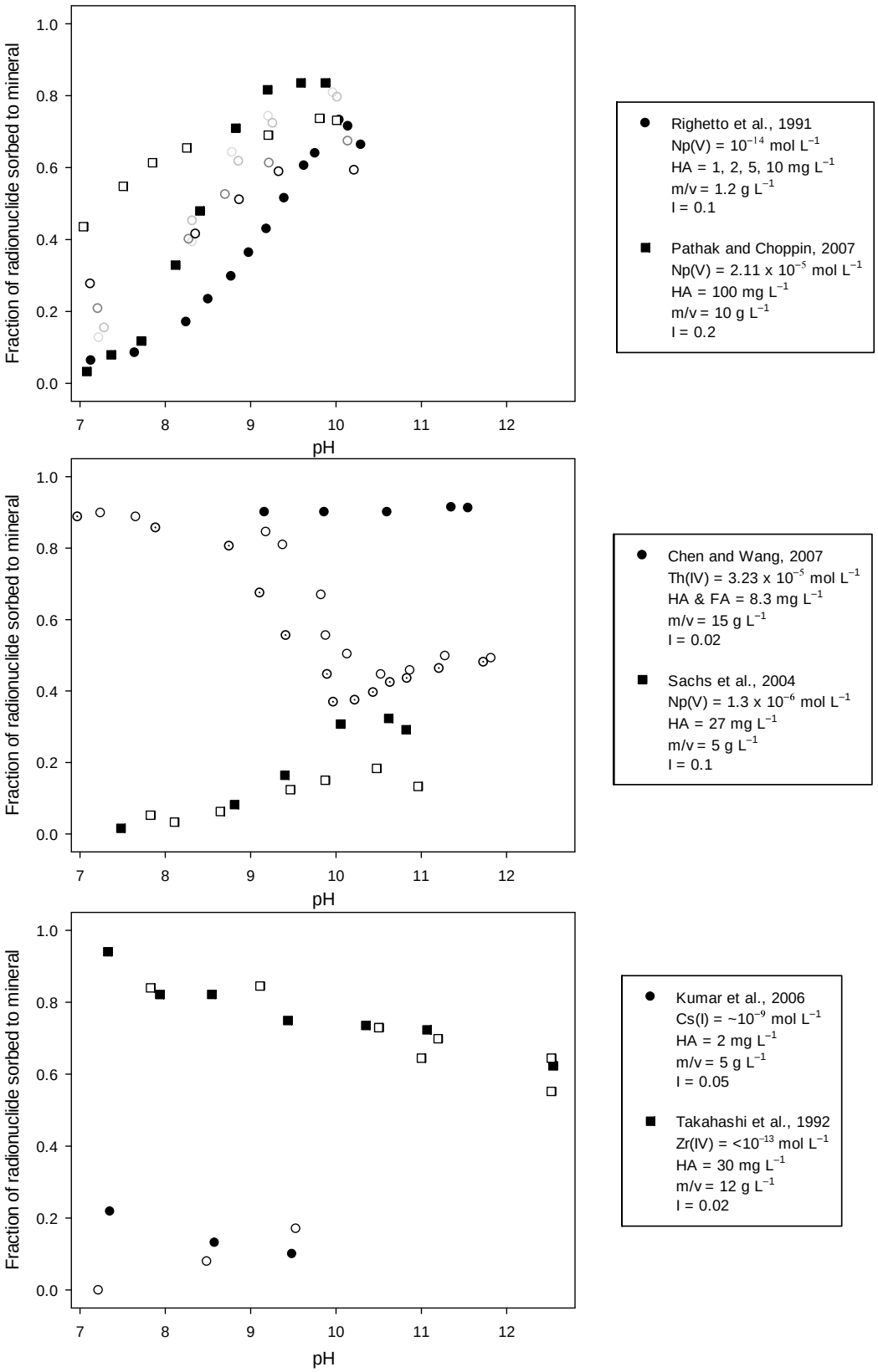

Figure S3c. Ternary system data for silica (quartz).

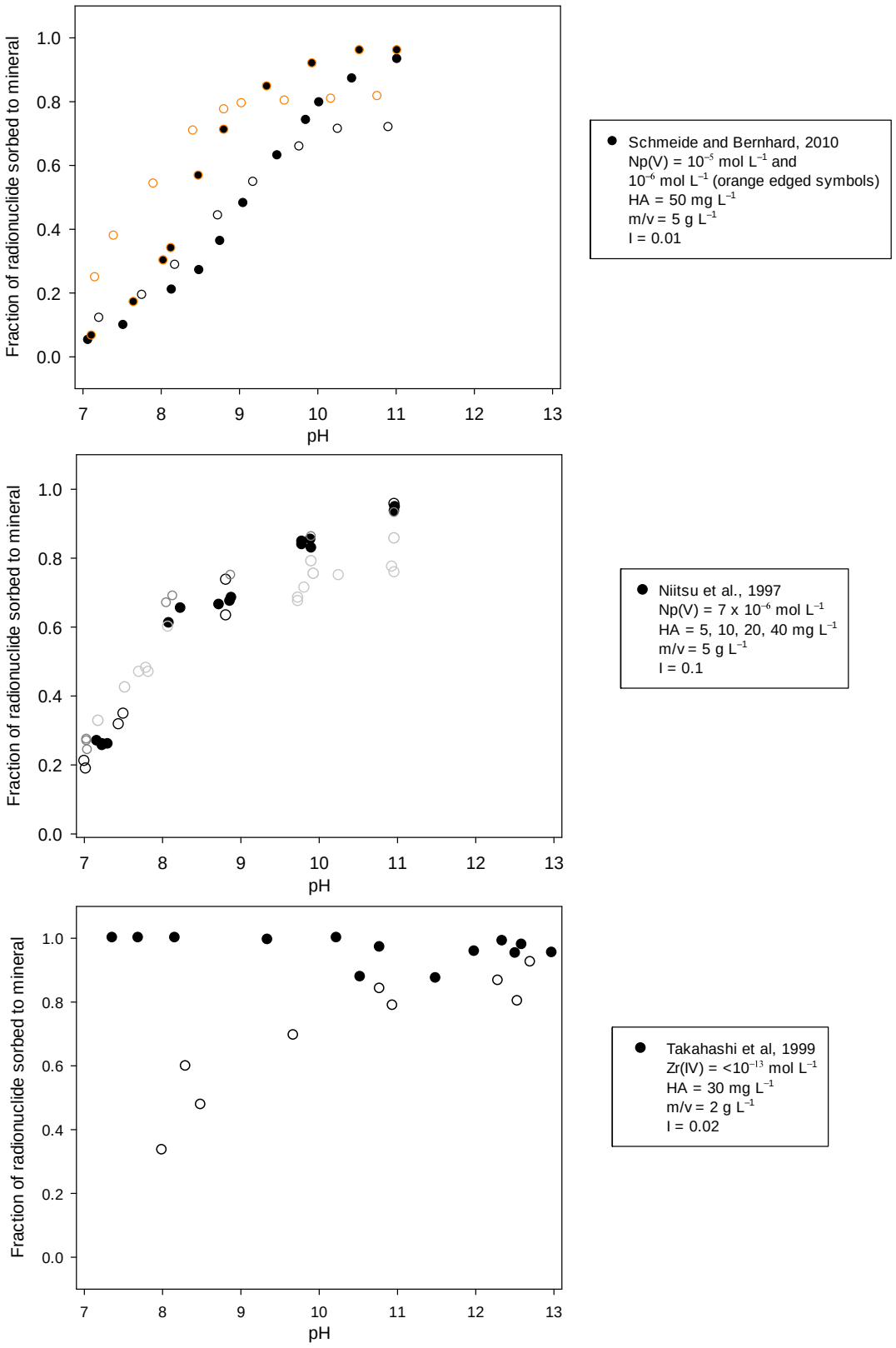

Figure S3d. Ternary system data for kaolinite.

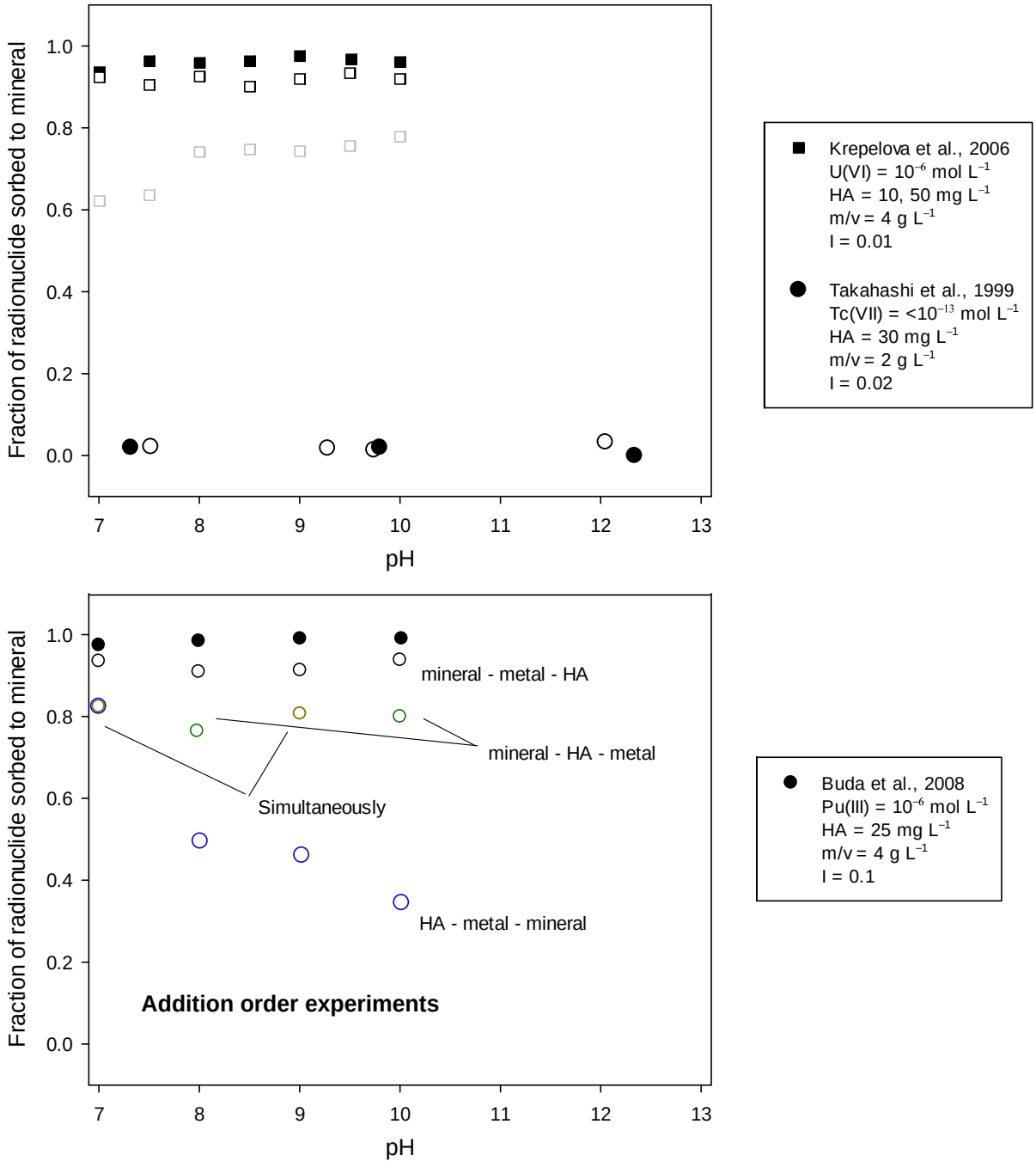

Figure S3d cont.

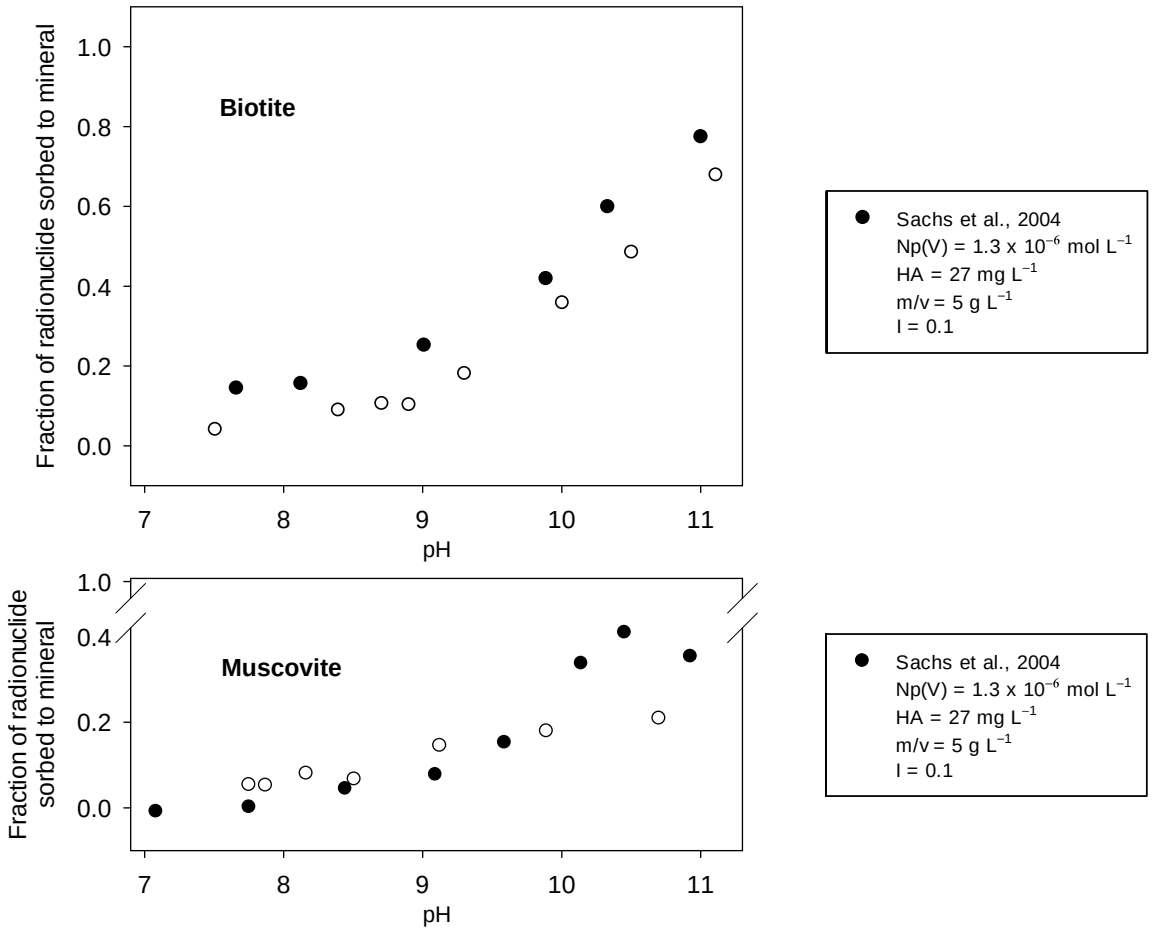

Figure S3e. Ternary system data for biotite and muscovite.

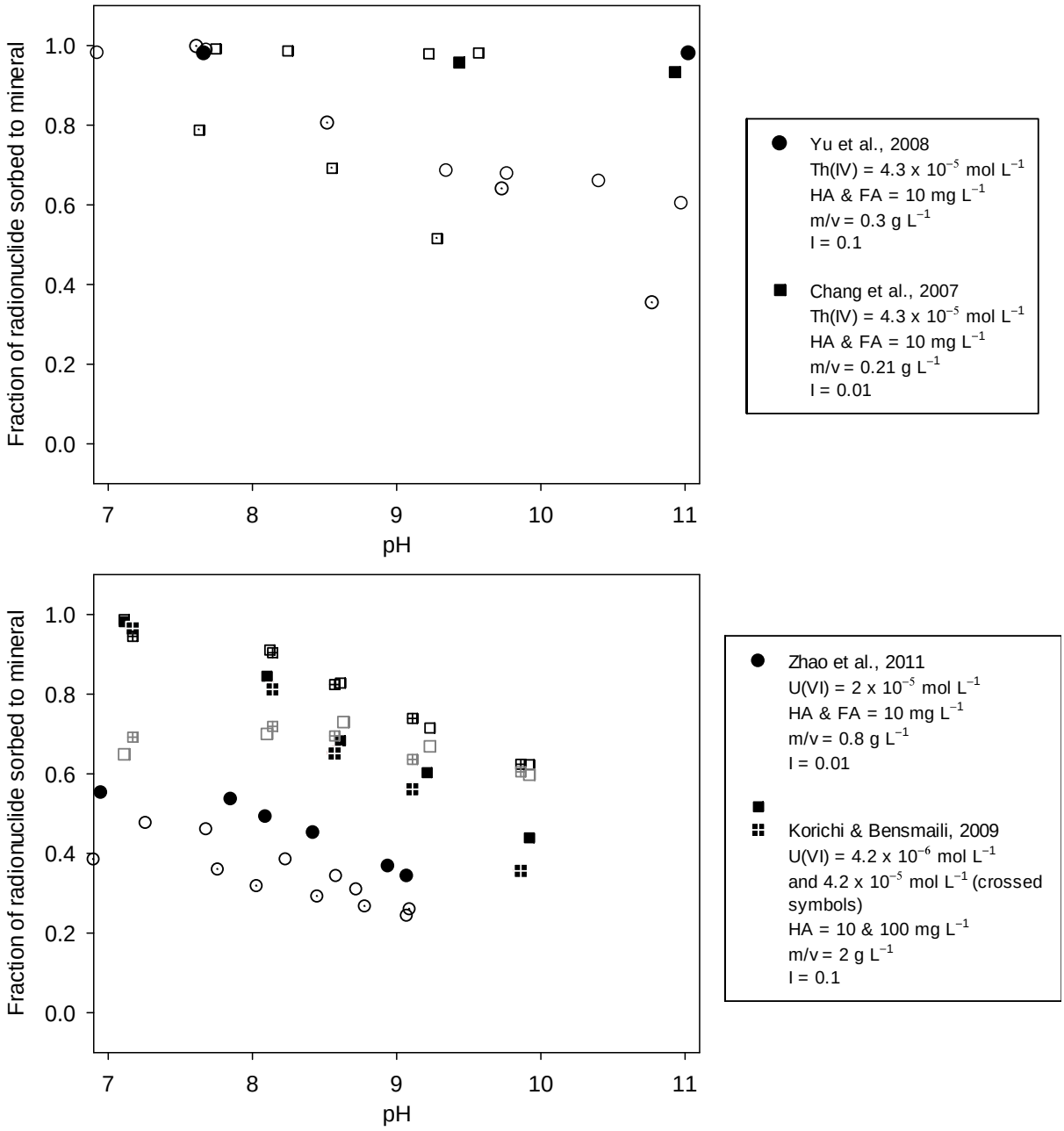

Figure S3f. Ternary system data for rectorite.

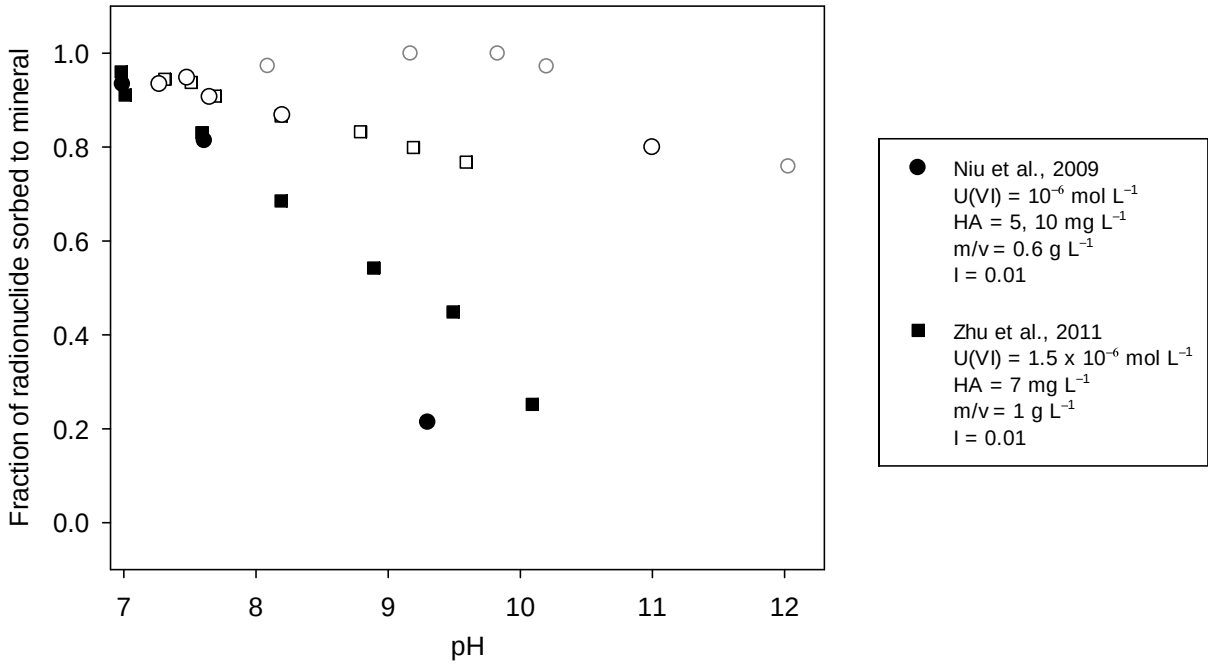

Figure S3g. Ternary system data for palygorskite.

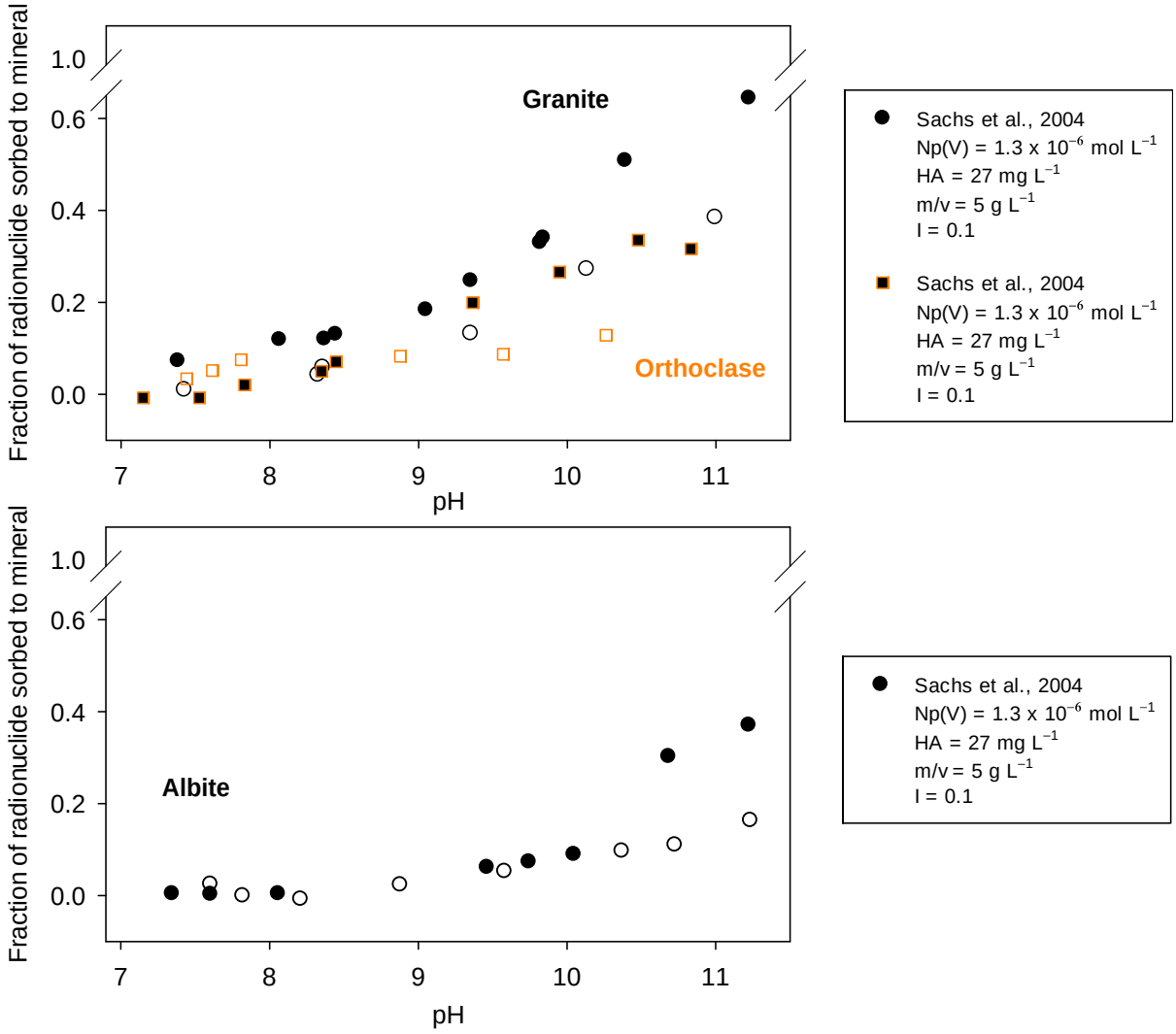

Figure S3h. Ternary system data for and granite, orthoclase, and albite.

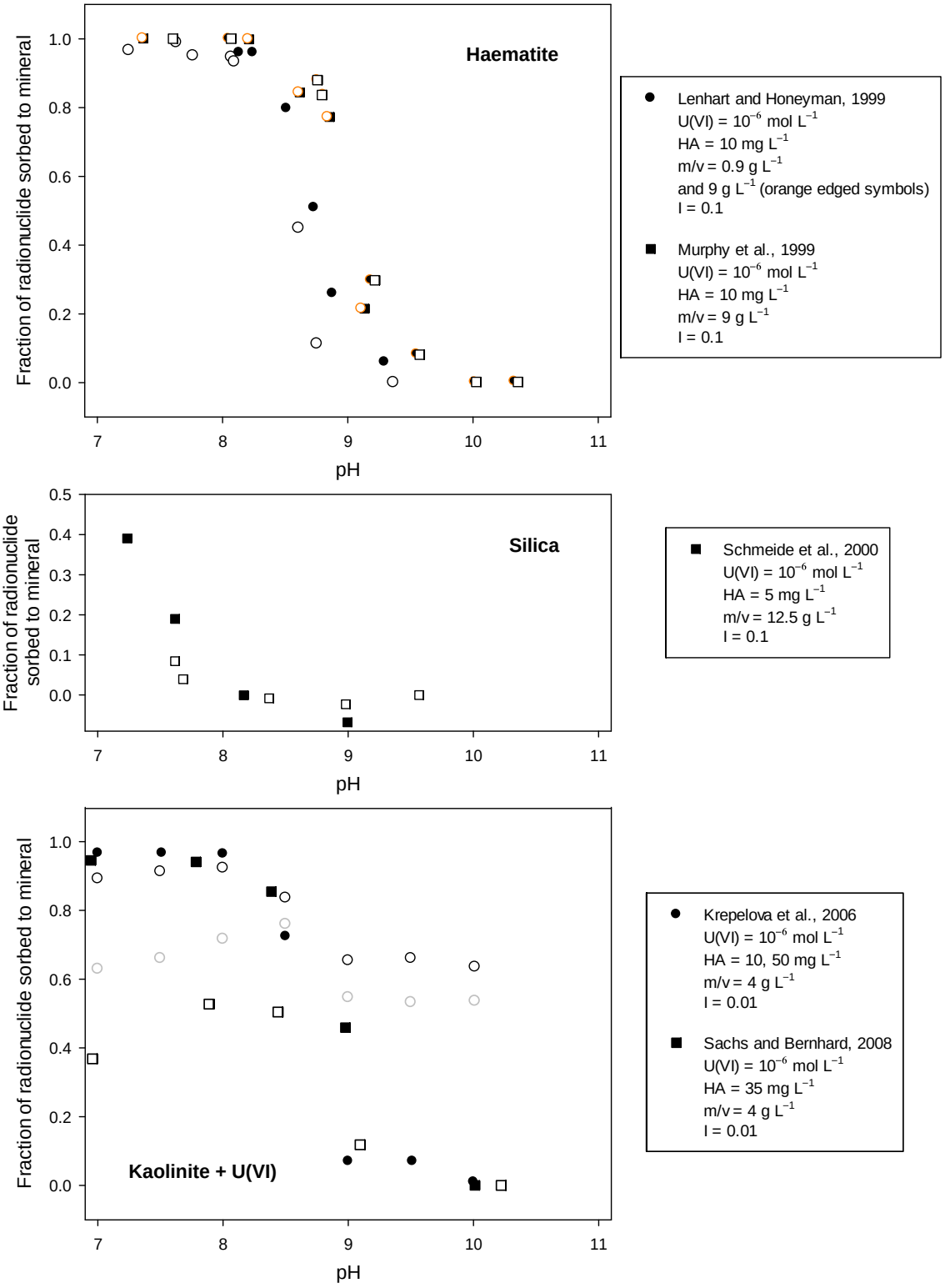

Figure S4. Ternary system data where experiments have been performed at atmospheric equilibrium with CO_2.

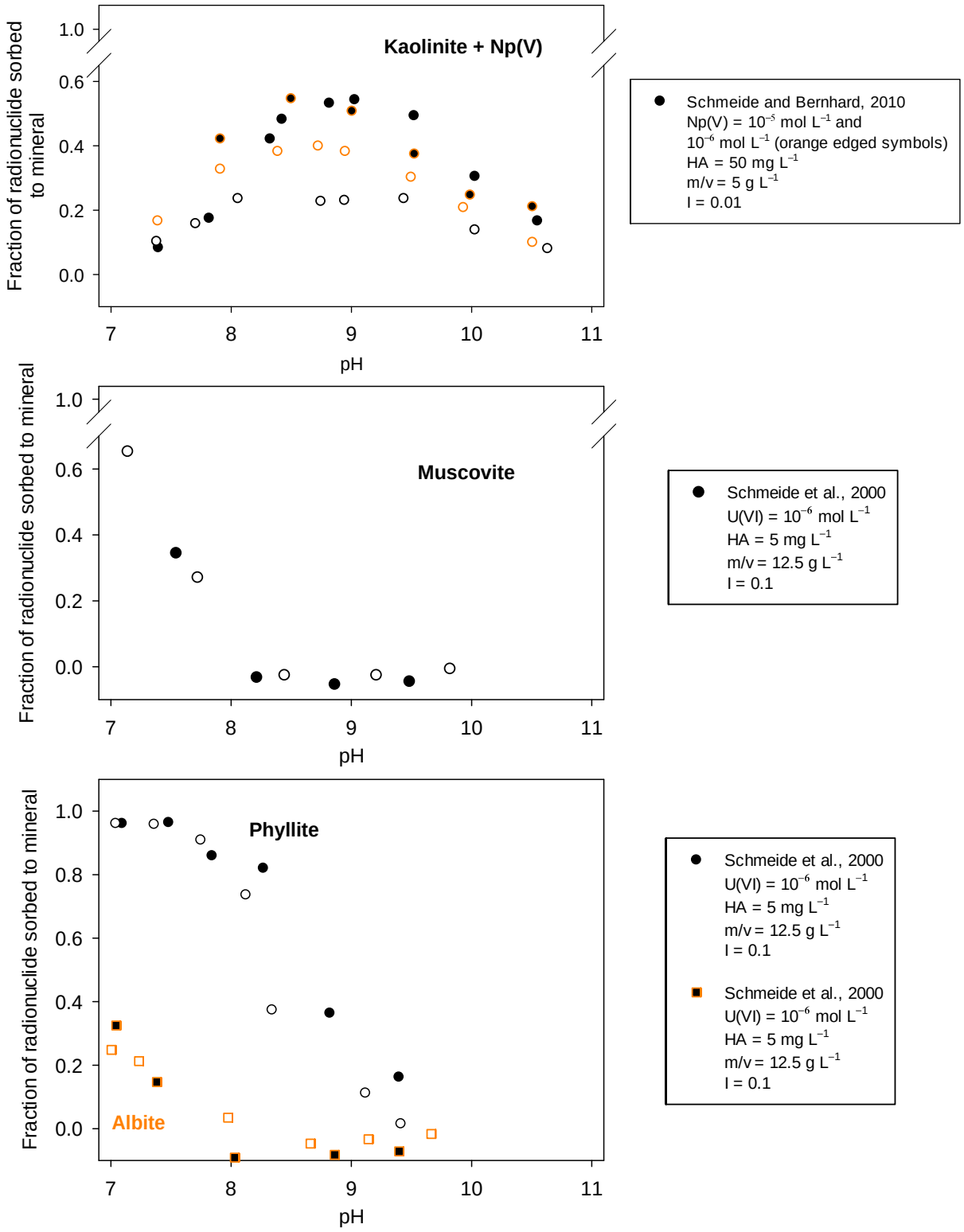

Figure S4 cont.

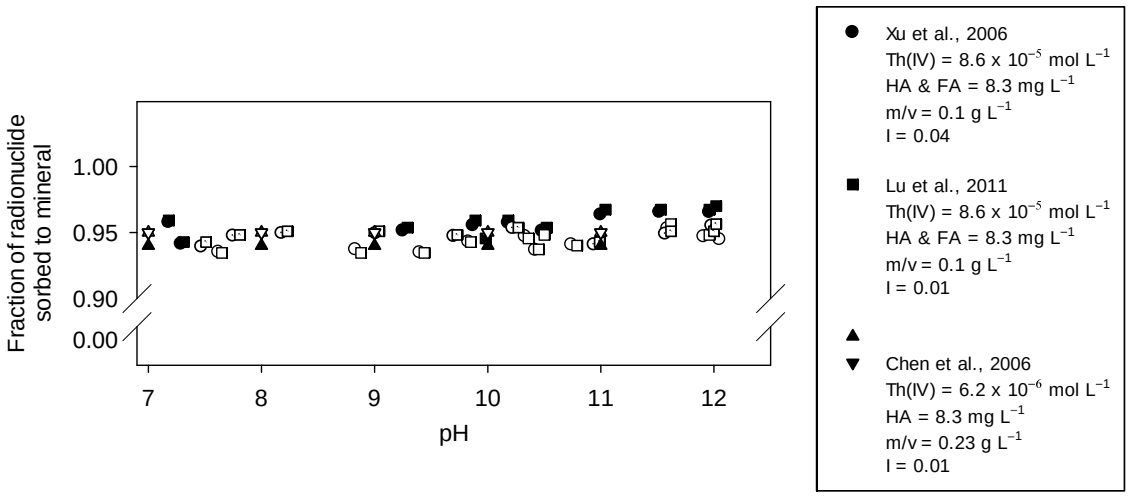

Figure S5a. Ternary system data for MX80 bentonite (Xu et al., and Lu et al.) and montmorillonite (Chen et al., upward triangles montmorillonite, downward triangles cross linked montmorillonite).

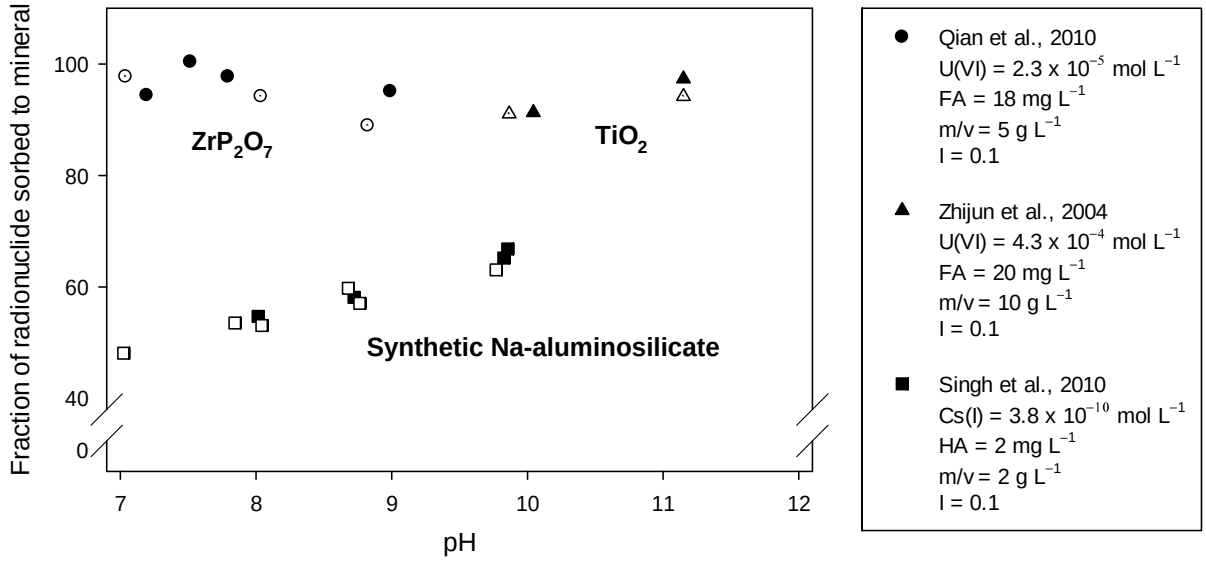

Figure S5b. Ternary system data for ZrP_2O_7, TiO_2 and synthetic Na-aluminosilicate.