

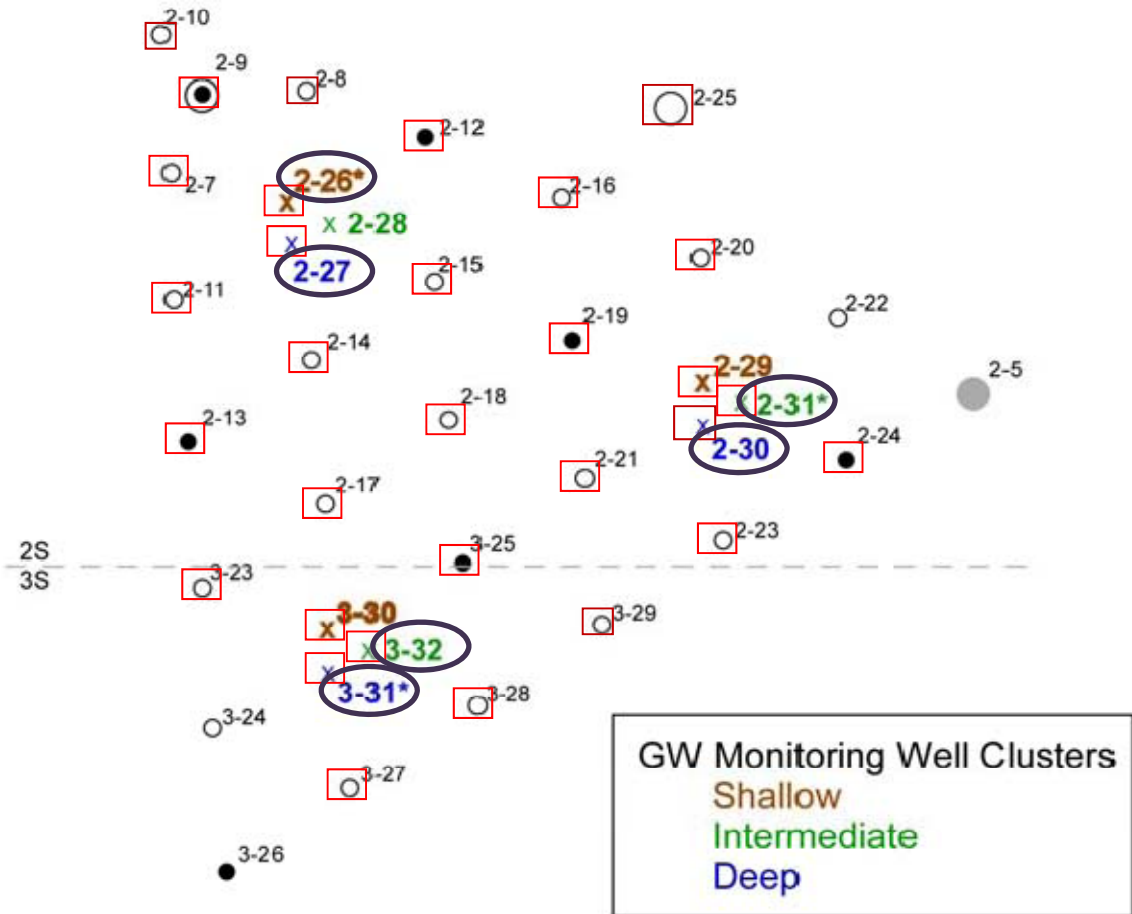
Smear-zone geochemical & hydrologic model:

On-going experiments & products

IFRC All Hands Meeting
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Doug Kent and Deb Stoliker
U. S. Geological Survey

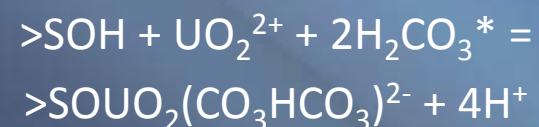
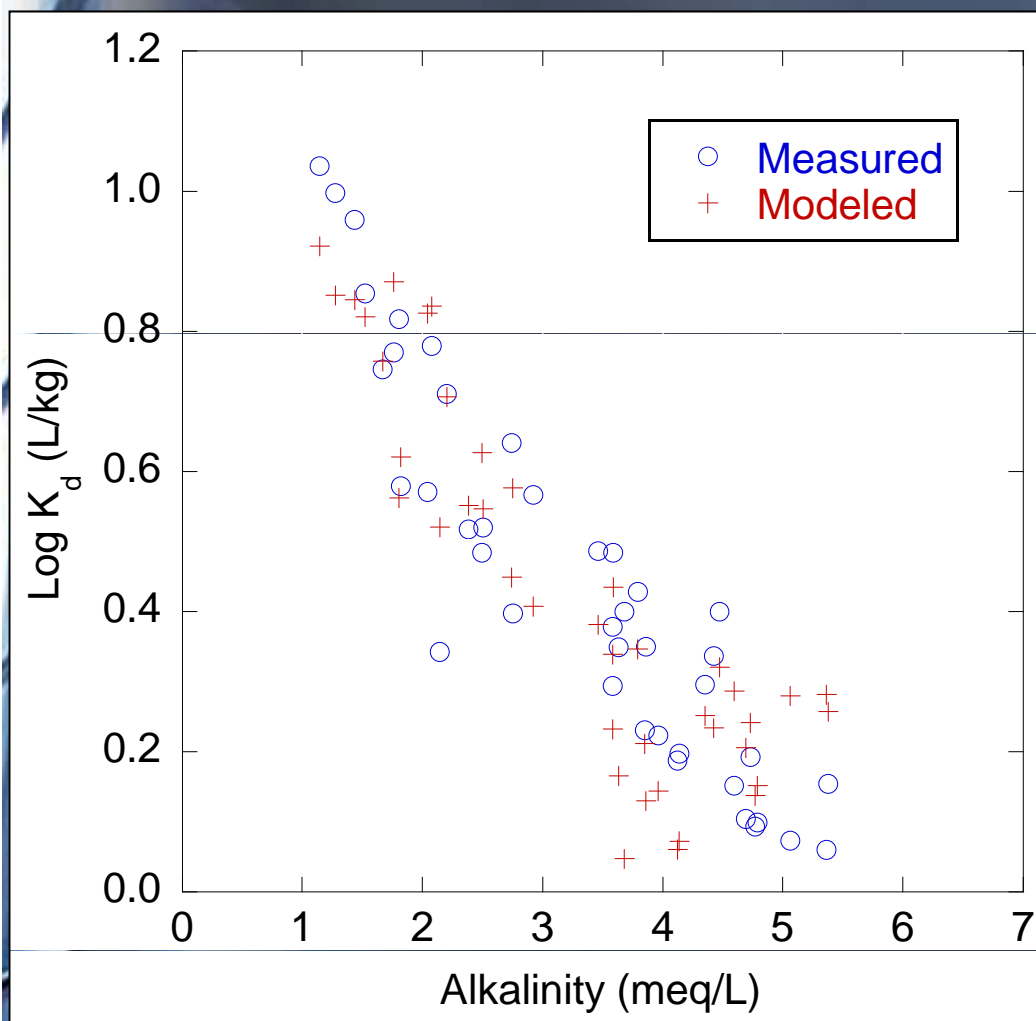
Individual samples and Site-Wide Composite (SWC)



Sample	Elevation Range (m above sea level)
2-26	105.8-107.3
2-27	106.1-107.3
2-30	106.4-107.3
2-31	106.5-107.3
3-31	106.7-107.6
3-32	106.3-107.3
SWC	102.6-108.0

Static Batch Reactors

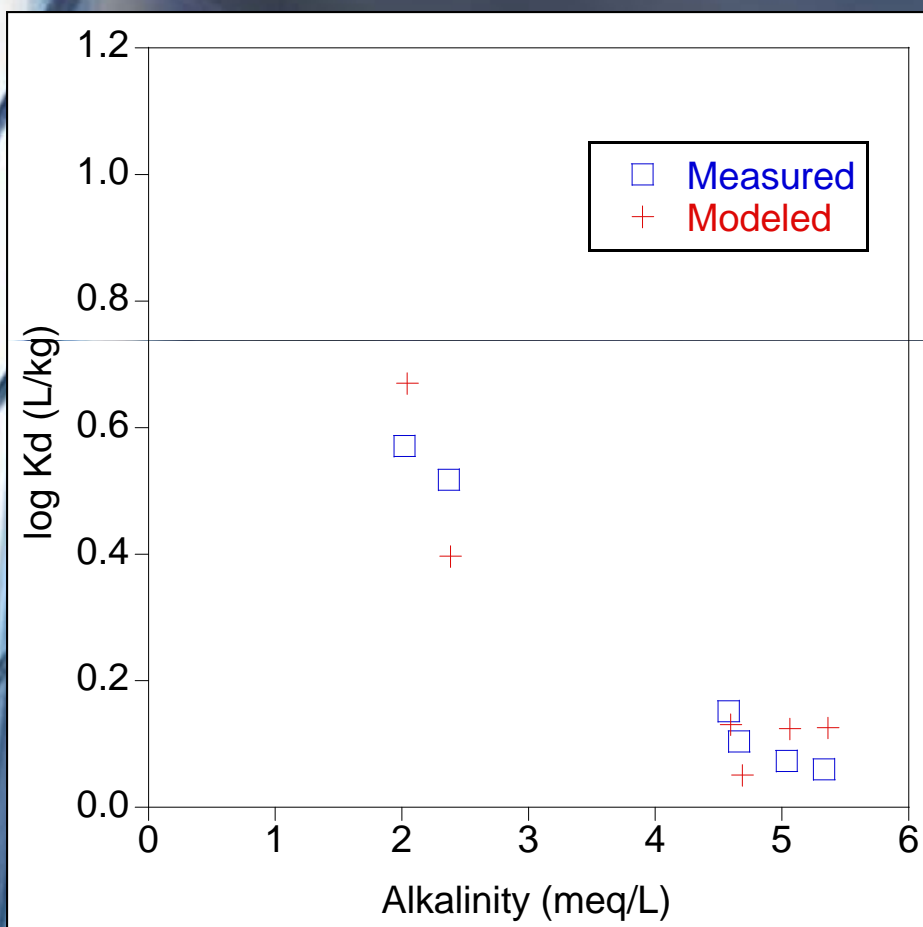
Surface Complexation Modeling



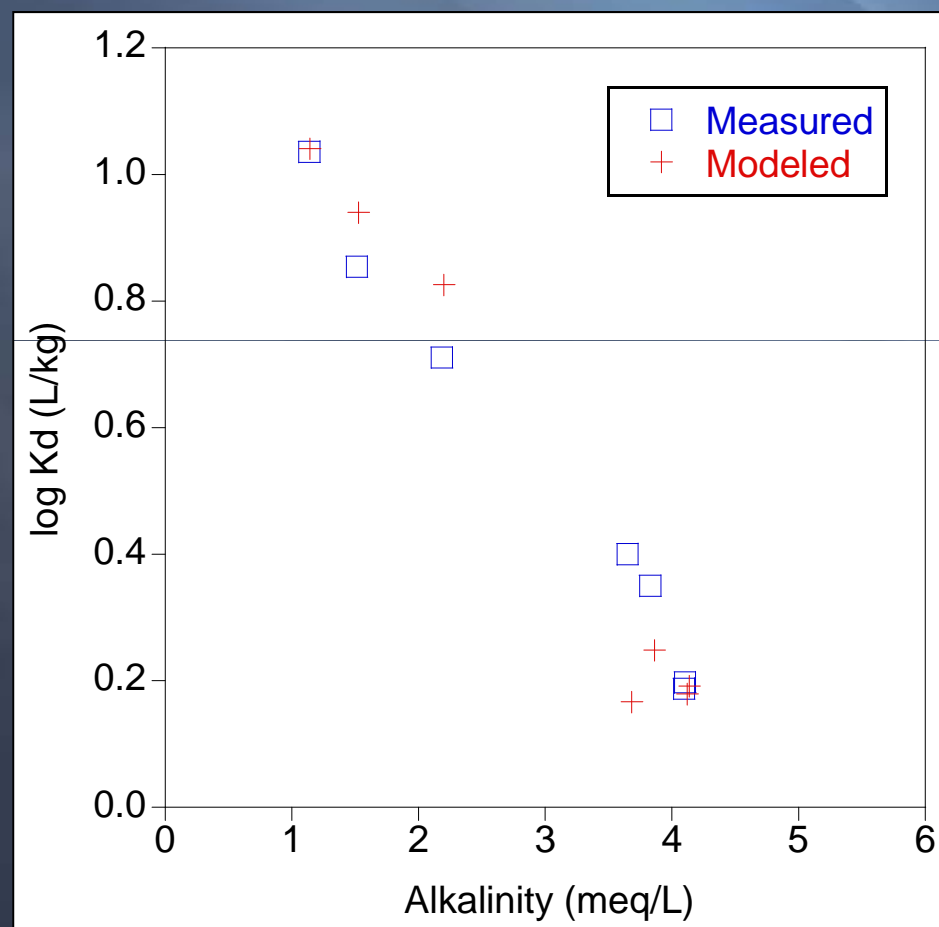
Sample	Surface Area (m ² /g)	Log K	WSOS / DF
All	12.5*	-11.72 ± 0.04	4.0
SWC-1	14.1 ± 0.9	-11.73 ± 0.11	2.8
2-26	11.7 ± 0.6	-11.78 ± 0.11	2.4
2-27	13.4 ± 0.9	-11.61 ± 0.12	2.1
2-30	13.3 ± 0.9	-11.69 ± 0.12	2.2
2-31	12.1 ± 0.6	-11.60 ± 0.11	2.5
3-31	11.7 ± 0.6	-11.80 ± 0.13	1.6
* 3-32	11.5 ± 0.6	-11.88 ± 0.11	2.0

Static Batch Reactors

Surface Complexation Modeling



3-32 composite
 $\log K = -11.88 \pm 0.11$
 $11.5 \pm 0.6 \text{ m}^2/\text{g}$



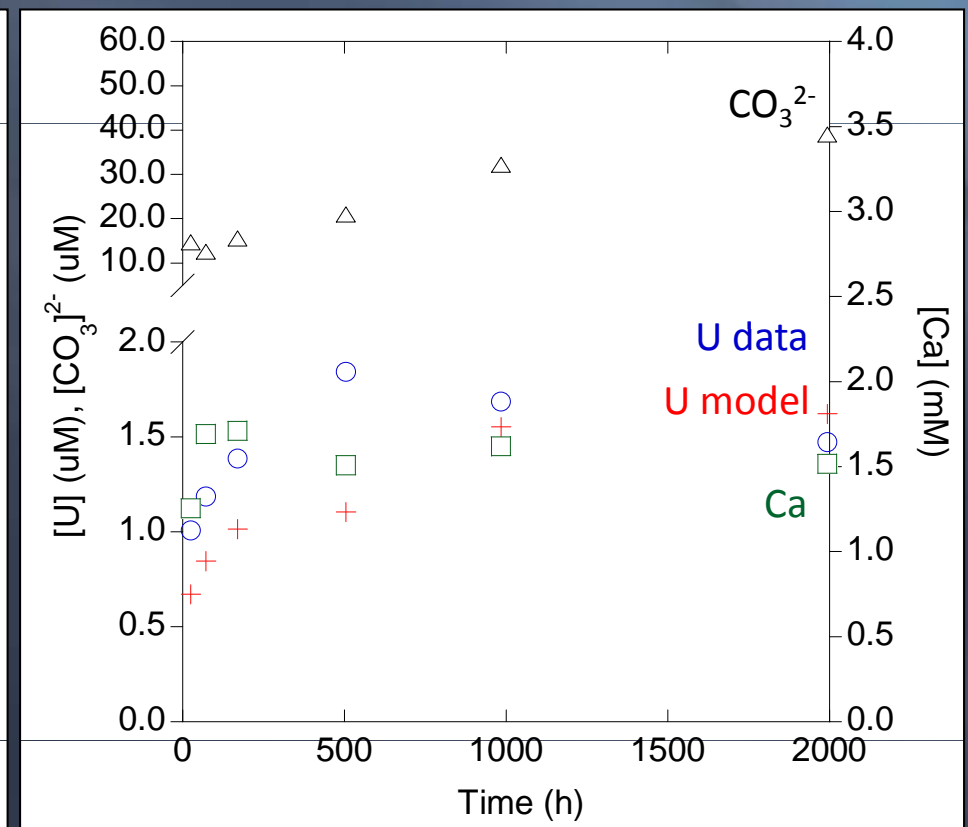
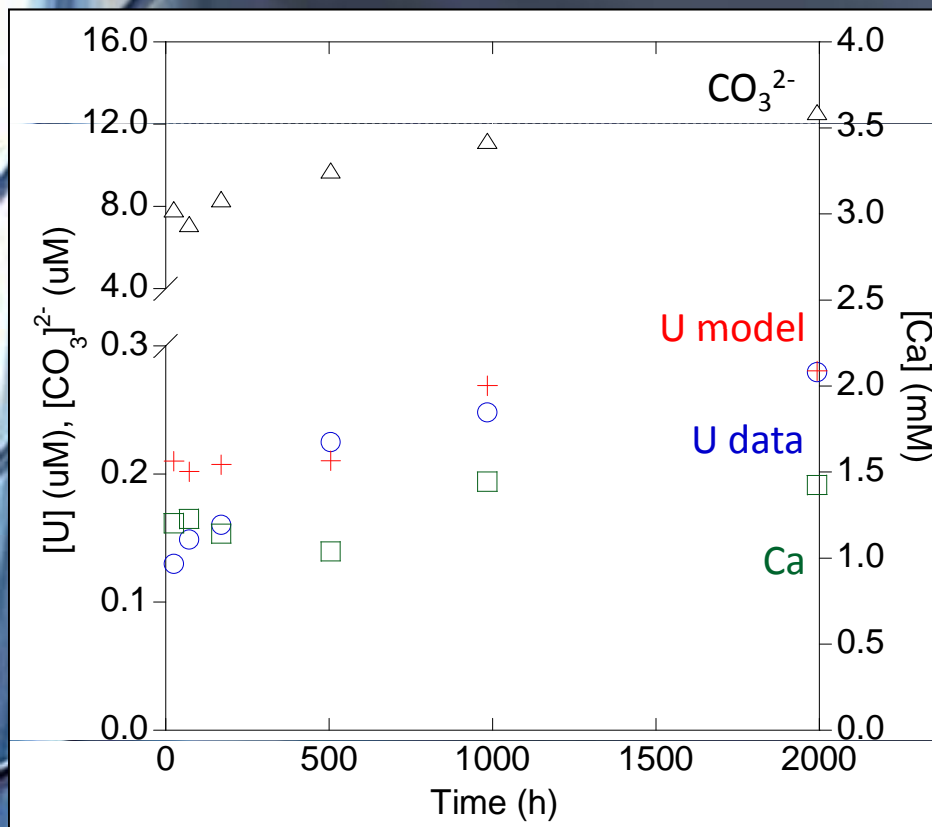
2-31 composite
 $\log K = -11.60 \pm 0.11$
 $12.1 \pm 0.6 \text{ m}^2/\text{g}$

Static Batch Reactors

Bulk Water Chemical Variations

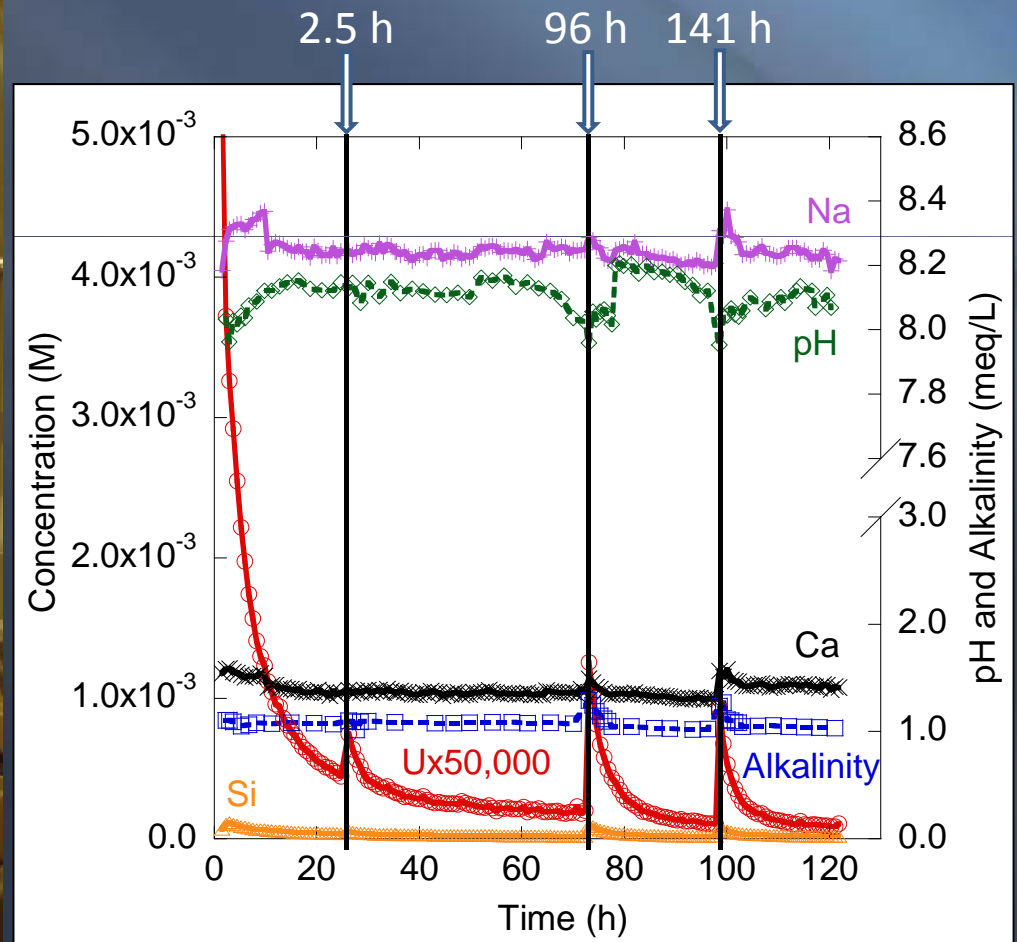
SWC, 100 g/L, 1 meq/L

SWC, 1000 g/L, 4 meq/L

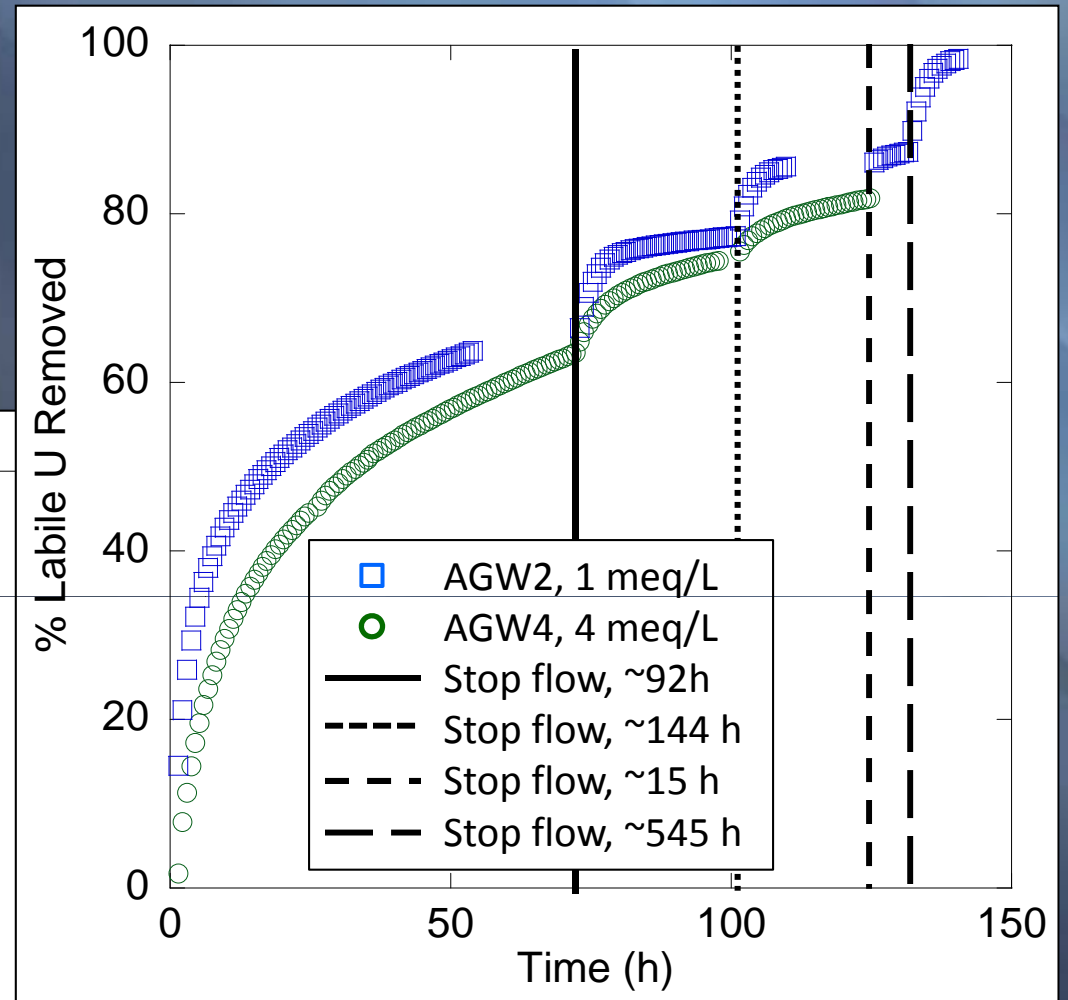
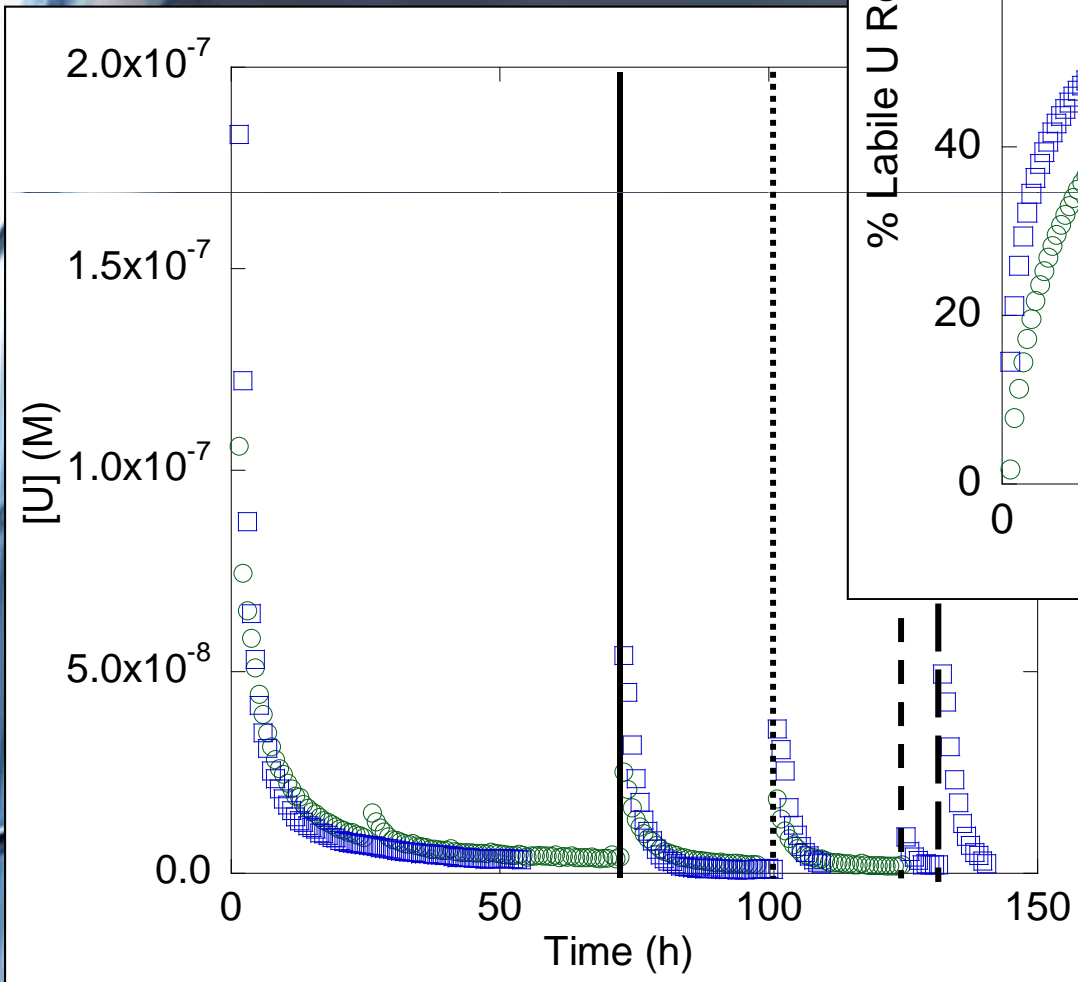


Flow-through Batch Reactors

SWC #2, <2 mm, 5 g / 45 mL, Alk = 1 meq/L

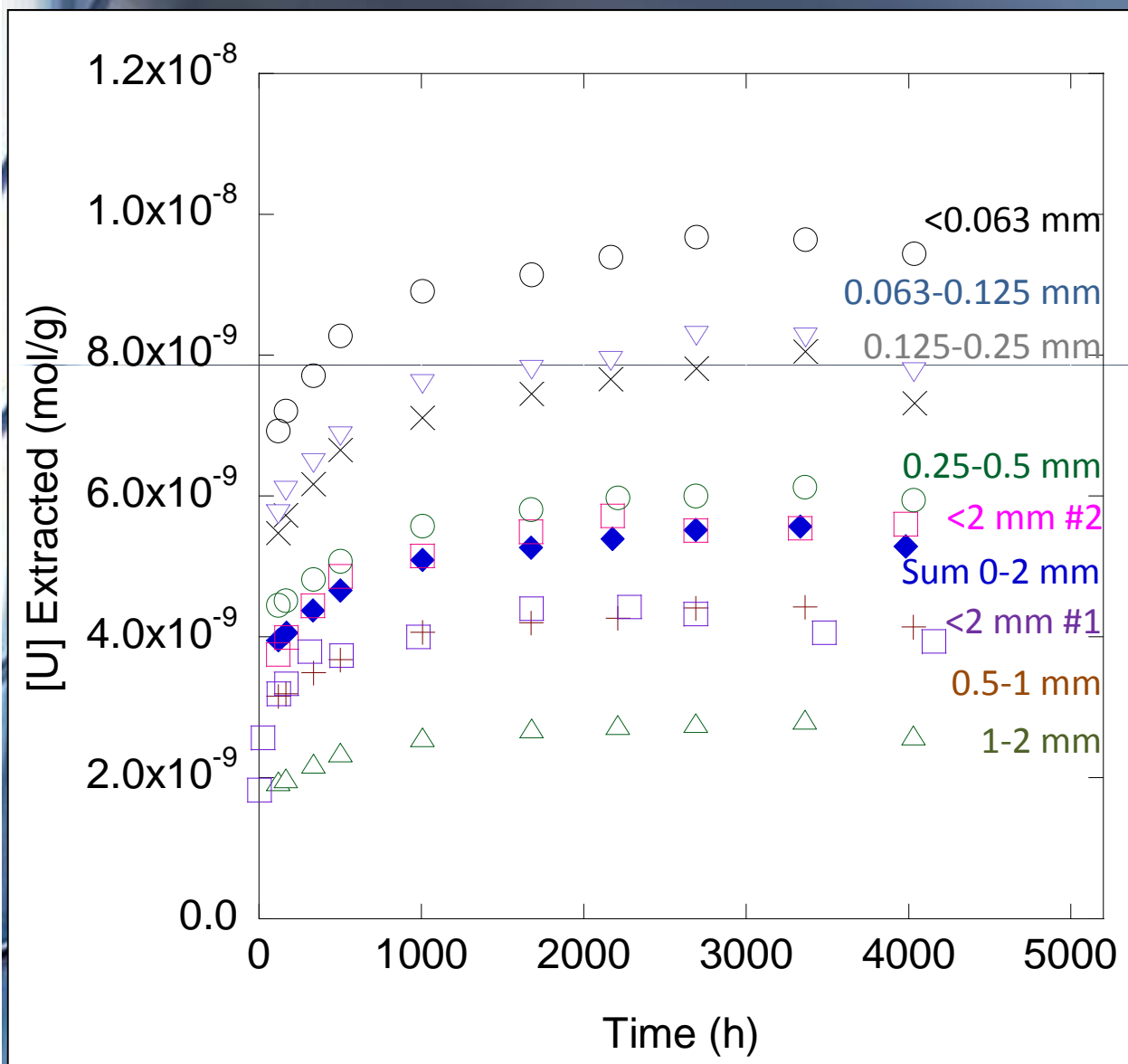


Flow Through Batch Reactors Alkalinity Effects



x-axes adjusted to overlay onset of elution after a stop-flow event

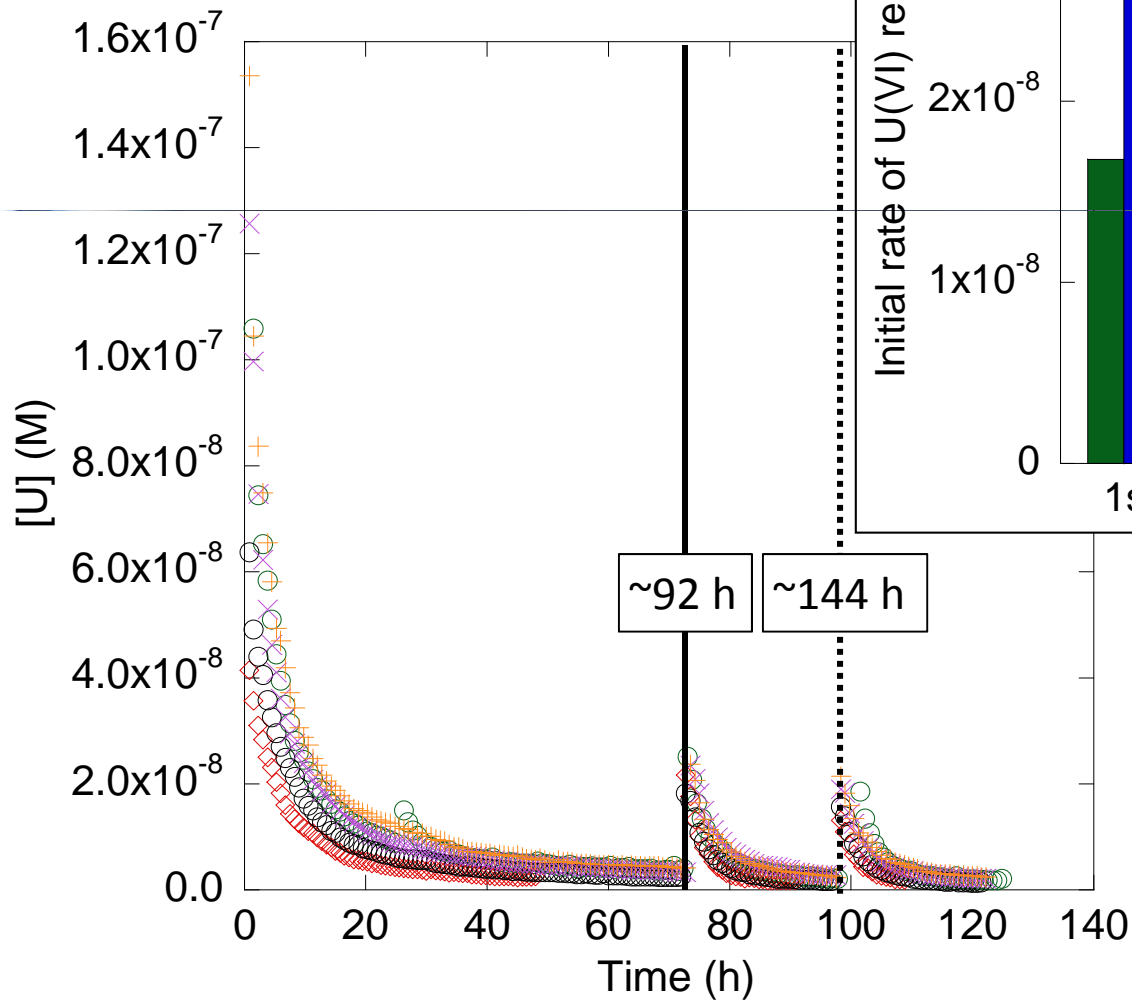
Carbonate Extraction - SWC Size Fractions



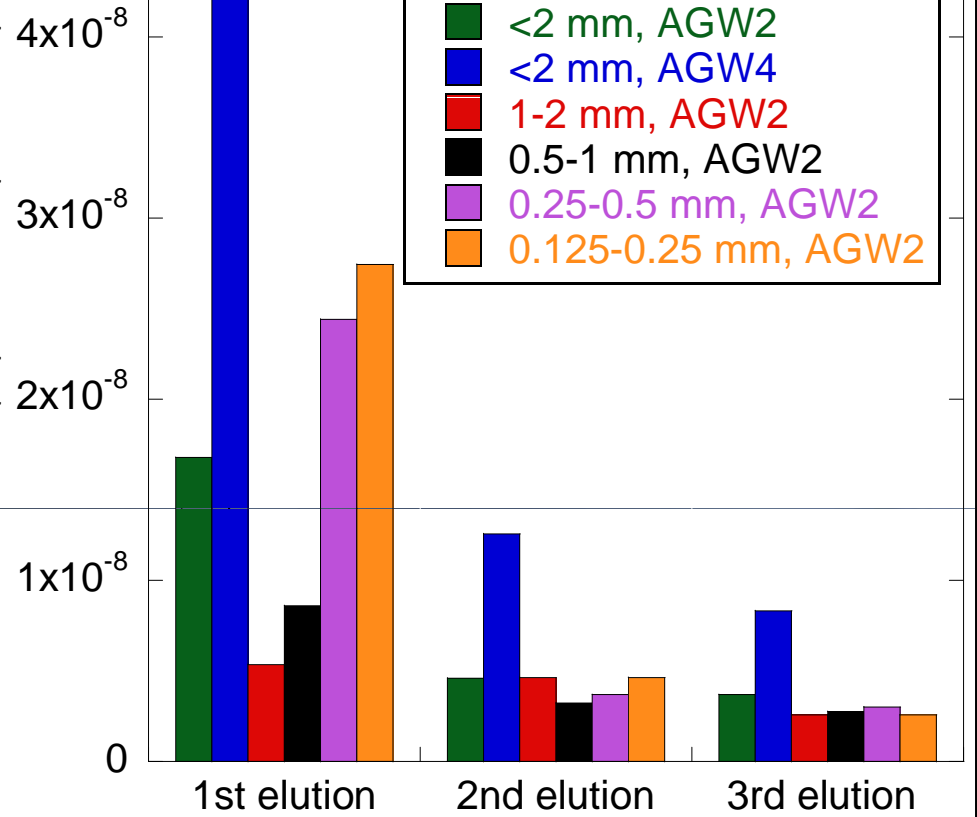
Size Fraction (mm)	Surface Area (m ² /g)	Weight % of <2 mm
<2 #1	14.1	-
<2 #2	18.0	-
1-2	13.4	27.0
0.5-1	15.0	25.2
0.25-0.5	17.5	21.1
0.125-0.25	18.2	5.1
0.063-0.125	19.6	7.3
<0.063	23.1	14.3

25-40% of U_{total} removed

Flow Through Batch Reactors Particle Size Effects



Initial rate of U(VI) release (mol/L/hr)



x-axes adjusted to overlay onset of elution after a stop-flow event

Experiment vs theory

Ratio of initial rates	Measured	Multi-rate SCM
AGW4/AGW2	2.5	2.2
Ratio to initial rate for 1-2 mm fraction		
0.5-1 mm	1.6	6.1
0.25-0.5 mm	4.6	7.1
0.125-0.25 mm	5.1	9.5

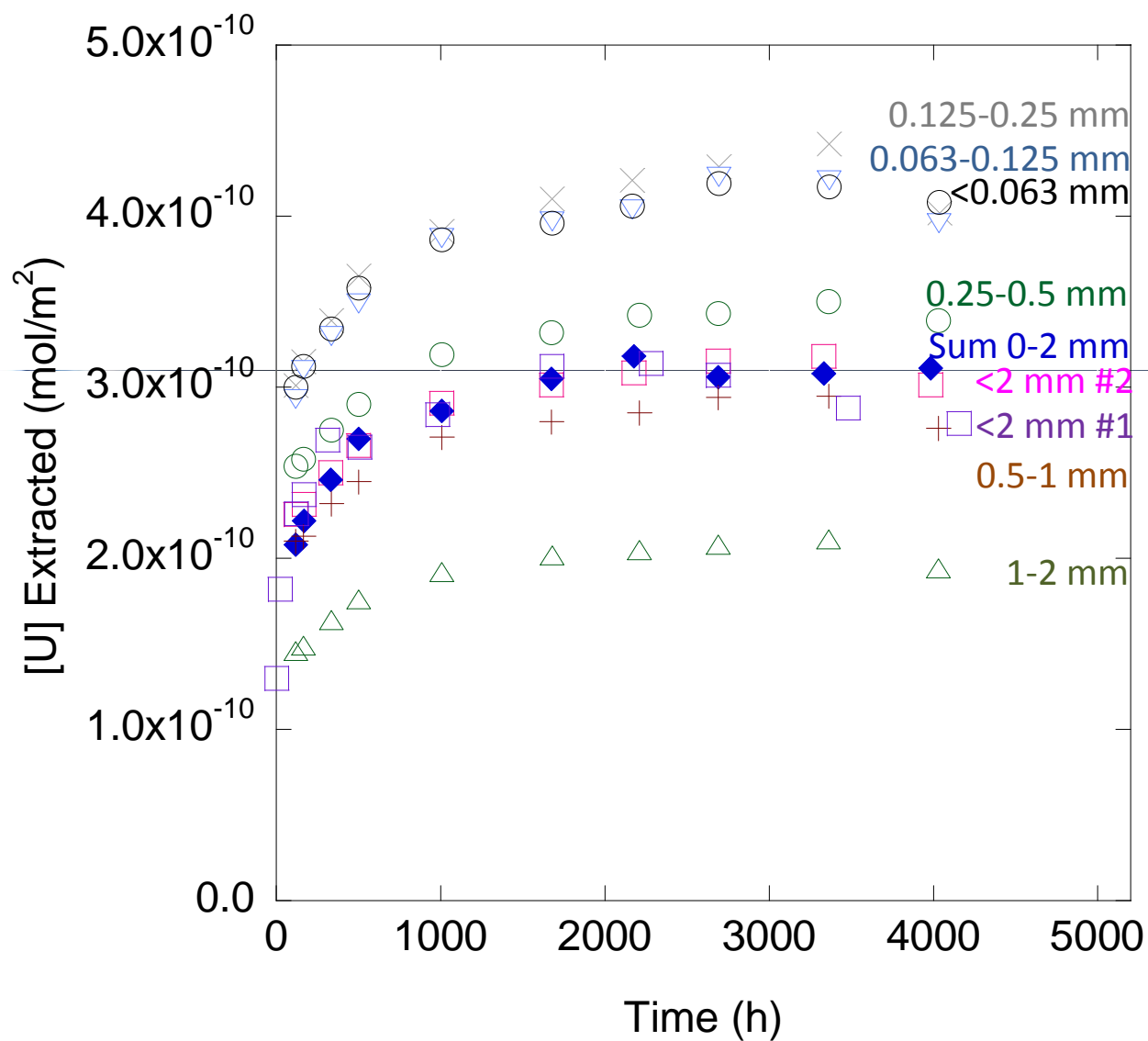
Conclusions

- Differences in U(VI) adsorption equilibrium constants determined by SCM vary by a factor of 2
- Differences in measured surface area vary by a factor of 1.3
- In static batch reactors, U(VI) adsorption equilibria is reached after 500-1000 h though chemical conditions continue to change for many thousands of hours
- In flow-through batch reactors, U(VI) desorption rates increase with increasing carbonate concentration
- In flow-through batch reactors, *initial* U(VI) desorption rate increases with decreasing particle size

Future Work

- Complete data set using flow-through batch reactor for all SWC size fractions at 1 meq/L
- Non-reactive tracer (Br) pulse to determine residency time for modeling
- Expand the range of chemical conditions to represent field-relevant alkalinity, pH, and [Ca]
- Uranium adsorption experiments
- Modeling
- More targeted experiments with upper saturated zone material from specific wells (limited quantities)

Carbonate Extraction - SWC Size Fractions



Size Fraction (mm)	S.A. (m ² /g)	Weight %
<2 #1	14.1	-
<2 #2	18.0	-
1-2	13.4	27.0
0.5-1	15.0	25.2
0.25-0.5	17.5	21.1
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0.063-0.125	19.6	7.3
<0.063	23.1	14.3

25-40% of U_{total} removed