Investigation of dominant hydrogeochemical processes influencing uranium transport in groundwater at a retired explosives test site

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Abstract

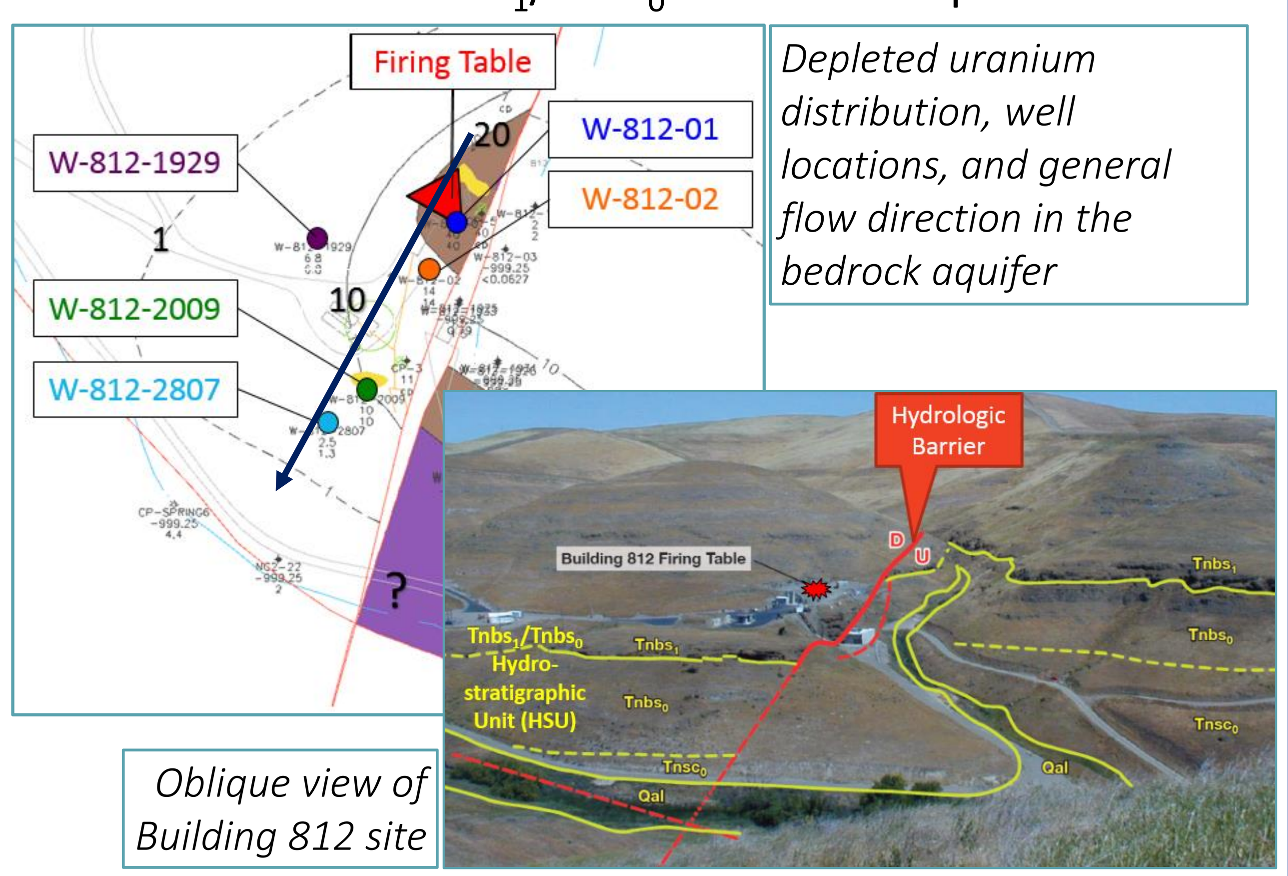
The Lawrence Livermore National Laboratory (LLNL) Site 300 facility was established in 1955 to support US weapons research and development. Open-air explosives tests, that used depleted uranium as a proxy, were conducted at the Building 812 site.

Photo of explosives test shot

Depleted uranium was released into the environment, eventually migrating into the underlying sandstone bedrock aquifer. At the site, groundwater concentrations of uranium exceed the California Maximum Contaminant Level of 20 picoCuries per liter (pCi/L). However, the groundwater contaminant plume appears to attenuate within 60 meters of the contaminant source, beyond which no depleted uranium is observed. The goal of this research is to determine the processes (i.e., sorption to mineral surfaces, biotransformation, diffusion, dispersion, and mineral precipitation) controlling attenuation of uranium in local groundwater.

Site Description

5 monitoring wells are screened 50-70 feet below ground surface within the $Tnbs_1/Tnbs_0$ sandstone aquifer.



Depleted uranium distribution, well locations, and general flow direction in the bedrock aquifer

Oblique view of Building 812 site

Research Questions

1. What are the dominant processes causing the observed attenuation of the depleted uranium?
2. Can a reactive transport model be used to simulate uranium transport in the aquifer?
3. If so, do the simulations indicate the system has the ability to attenuate the uranium indefinitely?

Objectives and Methods

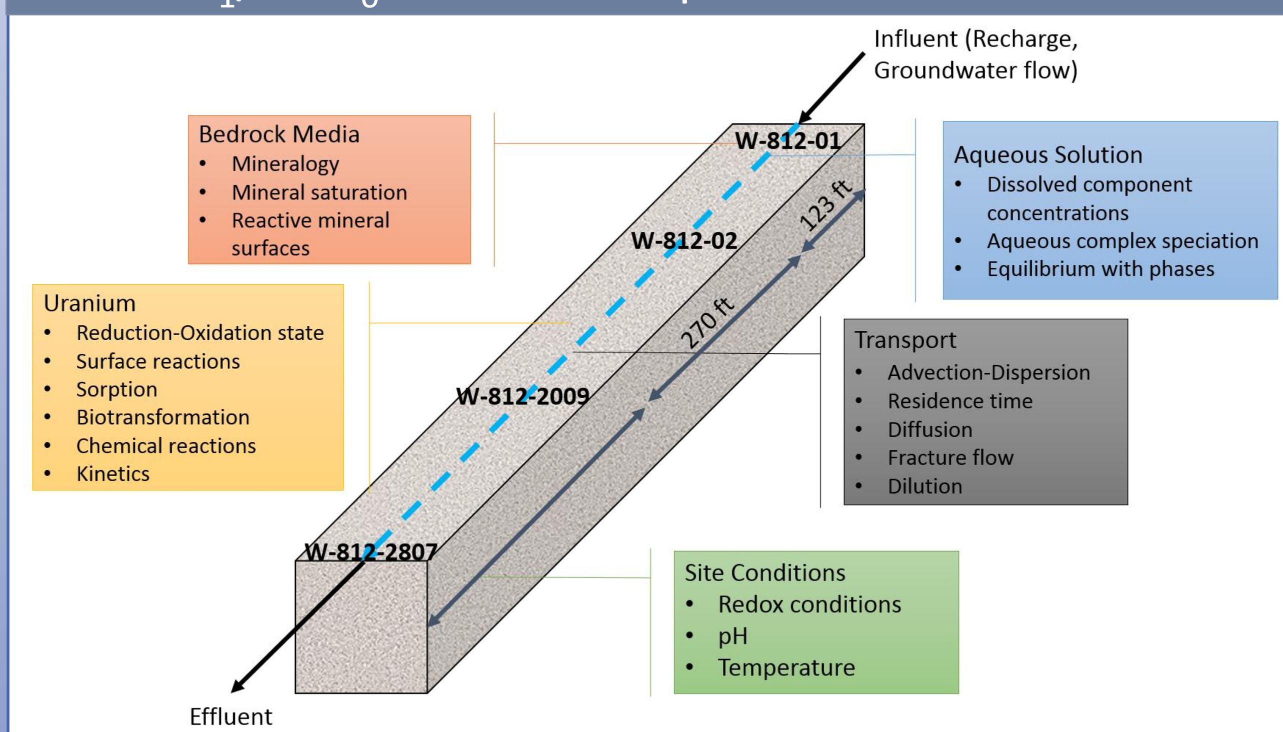
Determining the dominant processes influencing uranium transport will be completed by the following objectives.

A. Evaluate 14 years of hydrogeologic/chemical data

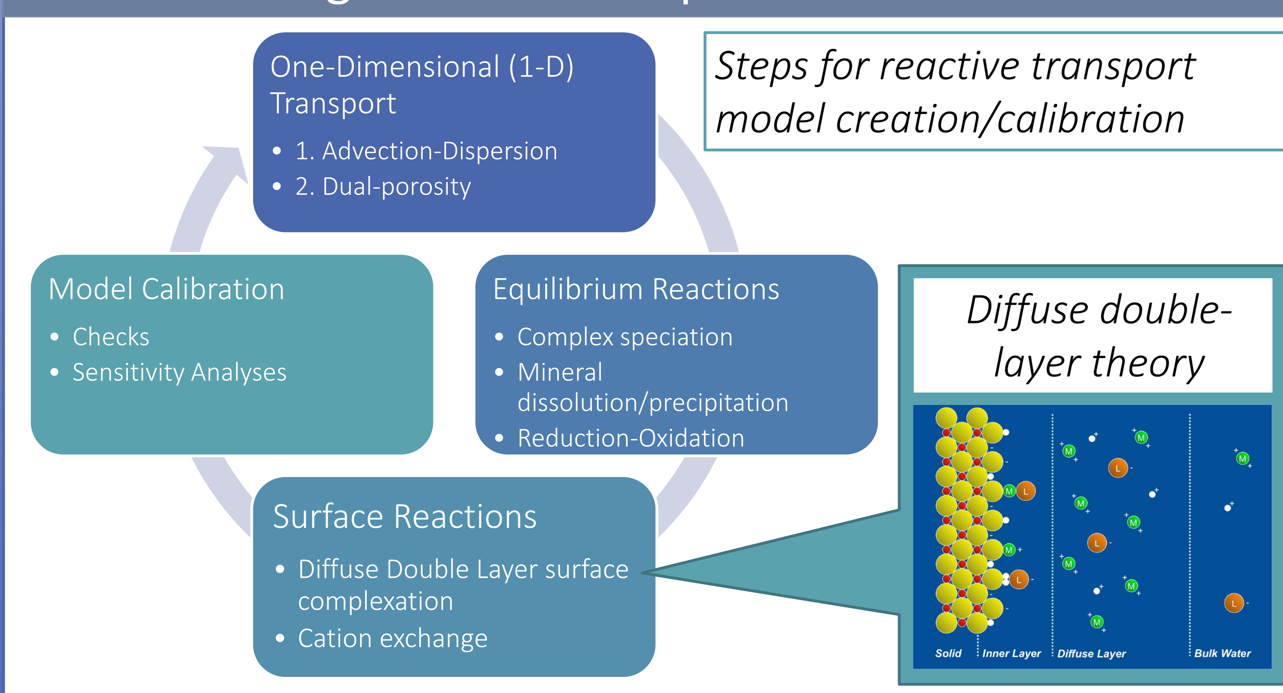
Monitoring Well	Field pH	Field Temperature (Degrees C)	Field Dissolved Oxygen (ppm)	Field Oxidation Potential (Eh) (mVolts)	Specific Conductance (umhos/cm)	Ca^{2+} (mg/L)	Mg^{2+} (mg/L)	Na^{+} (mg/L)	K^{+} (mg/L)	Bicarbonate Alkalinity (mg/L as CaCO3)	Cl^{-} (mg/L)	Nitrate (as NO_3) (mg/L)	PO_4^{3-} (mg/L)	SiO_2 (mg/L)	SO_4^{2-} (mg/L)	Uranium (ug/L)
W-812-01	7.6	20.8	5.9	82.2	801.6	32.2	24.7	80.8	7.0	186.3	61.8	61.9	0.1	67.0	26.8	68.4
W-812-02	7.9	21.1	7.3	54.2	464.9	26.0	20.2	40.8	6.6	148.0	27.0	54.8	0.1	68.0	23.3	46.2
W-812-1929	8.0	21.2	6.6	44.3	763.9	28.0	17.0	100.0	8.6	181.7	76.5	52.2	0.1	64.0	51.0	7.9
W-812-2009	7.4	20.4	5.6	34.7	787.4	56.6	30.5	35.4	4.3	149.8	62.8	20.8	0.3	77.0	98.7	8.4
W-812-2807	8.2	20.7		167.0	394.0	24.0	13.0	28.0	3.4	94.0	19.0	32.5	1.0	73.0	27.0	1.1

Average water chemistry values for the 5 monitoring wells

B. Create a hydrogeologic conceptual model of the $Tnbs_1/Tnbs_0$ sandstone aquifer



C. Apply a geochemical and reactive transport model, Geochemist's Workbench V.10, to identify specific processes, aquifer properties, and site conditions controlling uranium transport

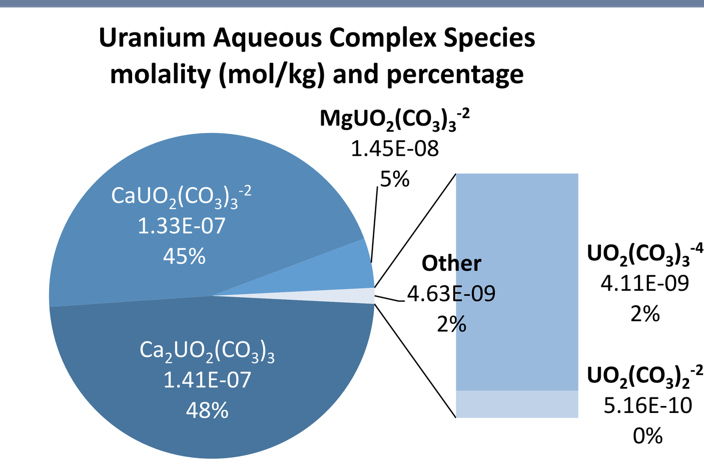


Steps for reactive transport model creation/calibration

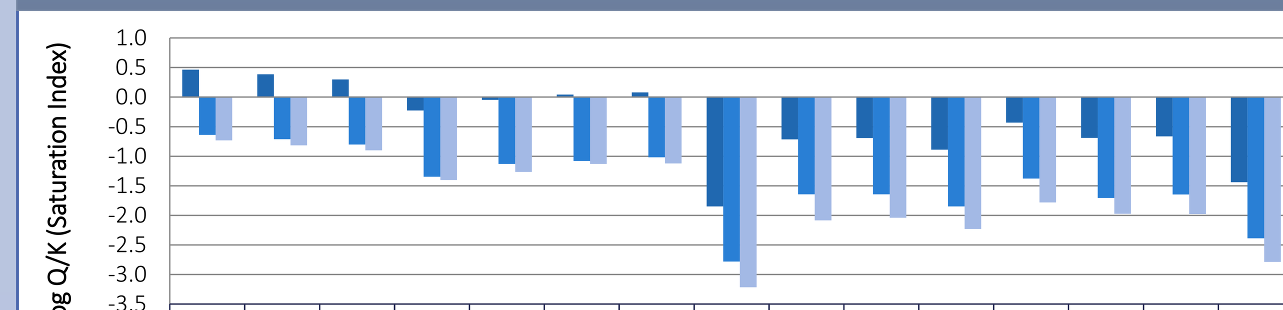
Results

Aqueous Speciation

Bicarbonate (HCO_3^-) complexes bond strongly with uranyl ions (UO_2^{2+}) in ground water. Uranium speciation in solution is dominated by uranyl carbonate complexes.



Mineral Saturation Indices



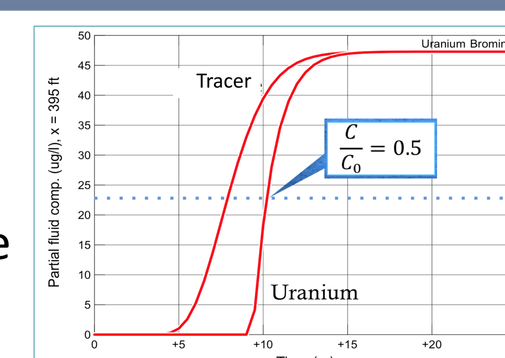
	9/29/00 W-812-01	5/26/04 W-812-01	6/21/05 W-812-01	9/28/00 W-812-02	5/26/04 W-812-02	6/21/05 W-812-02	10/12/06 W-812-02	2/22/05 W-812-2009	5/31/05 W-812-2009	10/6/05 W-812-2009	12/13/05 W-812-2009	12/23/06 W-812-2009	11/5/04 W-812-1929	5/31/05 W-812-1929	1/10/13 W-812-2807
Swartzite	0.5	0.4	0.3	-0.2	0.0	0.0	0.1	-1.8	-0.7	-0.7	-0.9	-0.4	-0.7	-0.7	-1.4
Liebigite	-0.6	-0.7	-0.8	-1.3	-1.1	-1.1	-1.0	-2.8	-1.6	-1.6	-1.8	-1.4	-1.7	-1.6	-2.4
Bayelite	-0.7	-0.8	-0.9	-1.4	-1.3	-1.1	-1.1	-3.2	-2.1	-2.0	-2.2	-1.8	-2.0	-2.0	-2.8

Saturation Indices for Uranium-bearing secondary phases in the aquifer

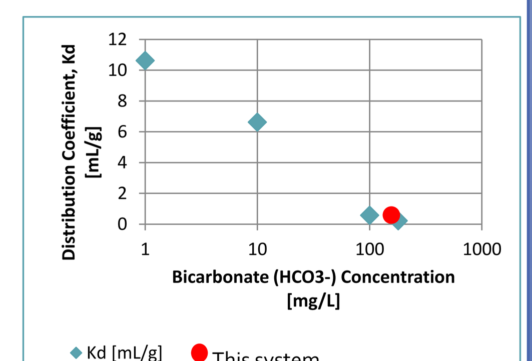
Preliminary Reactive Transport

Preliminary simulations of 1-D advective reactive transport along a transect from W-812-01 to W-812-2009 were conducted with the following parameters:

- UO_2^{2+} sorption to HFO modeled after Dzombak and Morel (1990)
- Diffuse double-layer surface complexation theory
- Sorption of uranyl carbonates to HFO % Volume of 0.01
- Retardation factors and distribution coefficients (K_d) calculated from breakthrough curves of UO_2^{2+} and a conservative tracer
- Various HCO_3^- concentrations were used to show effect on sorption



U and Tracer breakthrough curves at distance to W-812-2009 [HFO % Volume = 0.01}



K_d (and UO_2^{2+}) sorption decrease with increasing HCO_3^- concentration.

Summary

The hydrogeological conceptual model and the reactive transport model are preliminary. From the initial geochemical and reactive transport model outputs, the following suggestions can be made about the site.

- The chemistry of the aquifer is governed by characteristic silicate weathering reactions.
- The Aquifer is highly oxidized, implying uranium will likely exist in the more mobile oxidized state.
- The Majority of uranyl ion will complex with carbonate ions
- Uranium-bearing secondary phases are dominantly undersaturated, except for Swartzite in water in W-812-01 and W-812-02
- Uranyl carbonates will affect the amount of sorption/interaction with a surface
- Sorption may be dominant process, but refinement of model is needed.

Future Work

Further research includes the following activities and objectives:

- Further evaluation of site-specific data
- Refinement of advective-dispersive transport model
- Further evaluation and elimination of attenuation processes
- Input of different processes into the reactive transport model to determine their relative importance
- Inverse-modeling to determine additional potential processes
- Determine actual reactive mineral content in $Tnbs_1/Tnbs_0$ rocks (whole rock mineralogy analysis)
- Calibrate model to site data and conduct sensitivity analyses
- Determine if dual porosity models hydraulics effectively

Acknowledgements

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