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Evapotranspiration and Geochemical Controls on Groundwater Plumes in Arid Climates:

Lessons from Archetype Uranium Milling Sites

Tuba City Site

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General Background

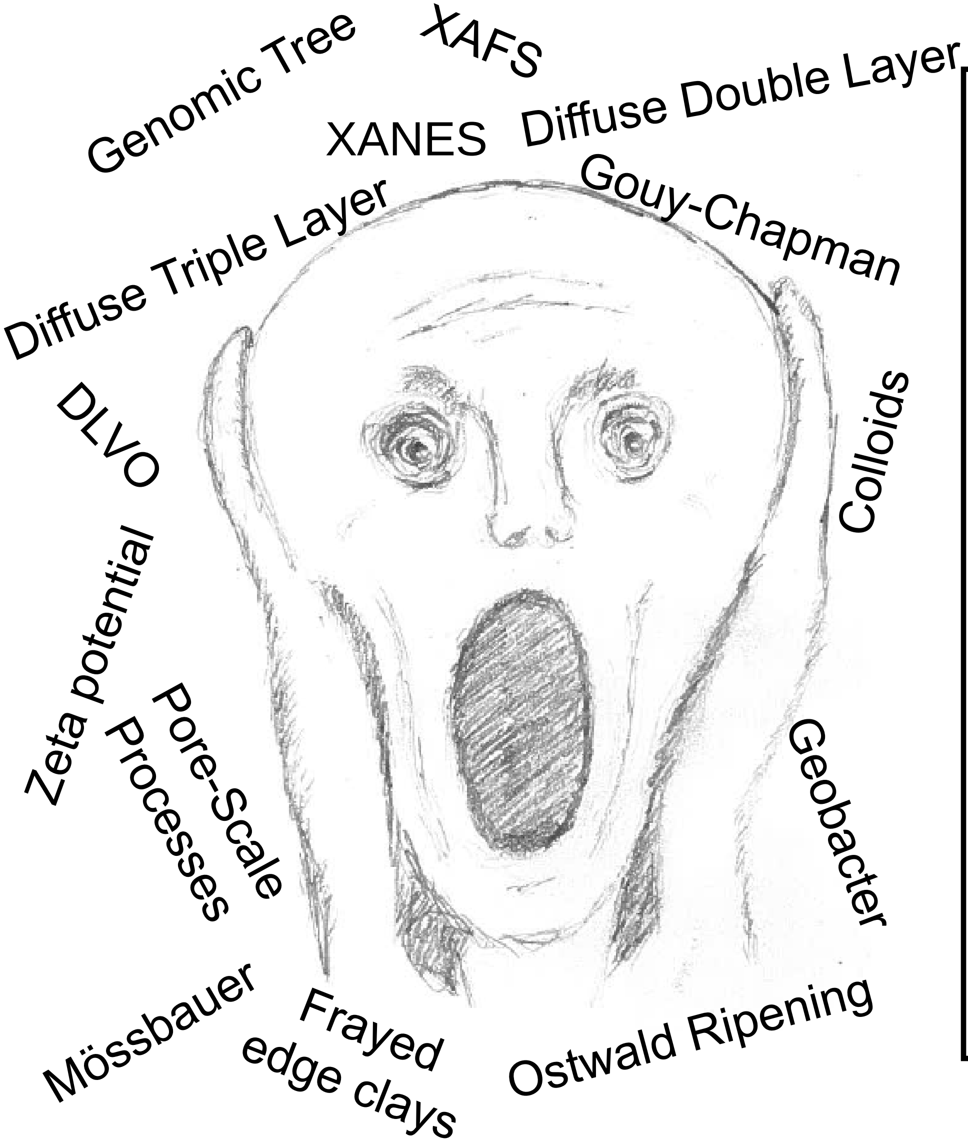
The Department of Energy Office of Legacy Management (DOE-LM) is responsible for a number of former uranium milling and processing sites and the associated tailings disposal areas (“Uranium Mill Tailings Radiation Control Act” – UMTRCA – sites)

A majority of the UMTRCA sites are in arid regions in the western United States

In 2013, DOE-LM sponsored an independent technical evaluation of the UMTRCA groundwater program/strategy

The results for the “case study” Tuba City Mill/Tailings Site and other sites provide key insights related to the challenges and opportunities for UMTRCA groundwater and the potential for site-specific alternative end states

Avoid Paralysis by Perceived Complexity

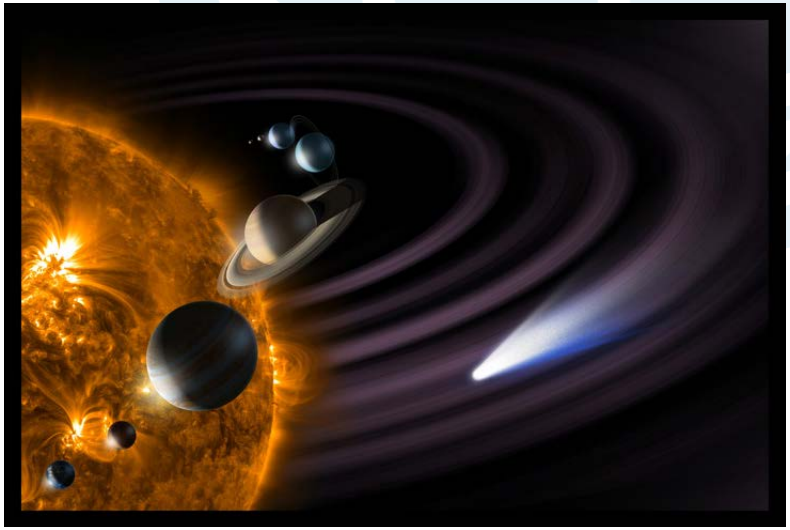


Begin With What You Know

- **Nature of source**
- **Contaminant distribution**
- **Bio-Geo-Chemical conditions of plume**
- **Background Bio-Geo-Chemical conditions**
- **Geological system and Hydrology**
- **General contaminant chemistry**

We often know 90% of what we need to know for Environmental Management Success

Recent UMTRCA groundwater technical evaluation



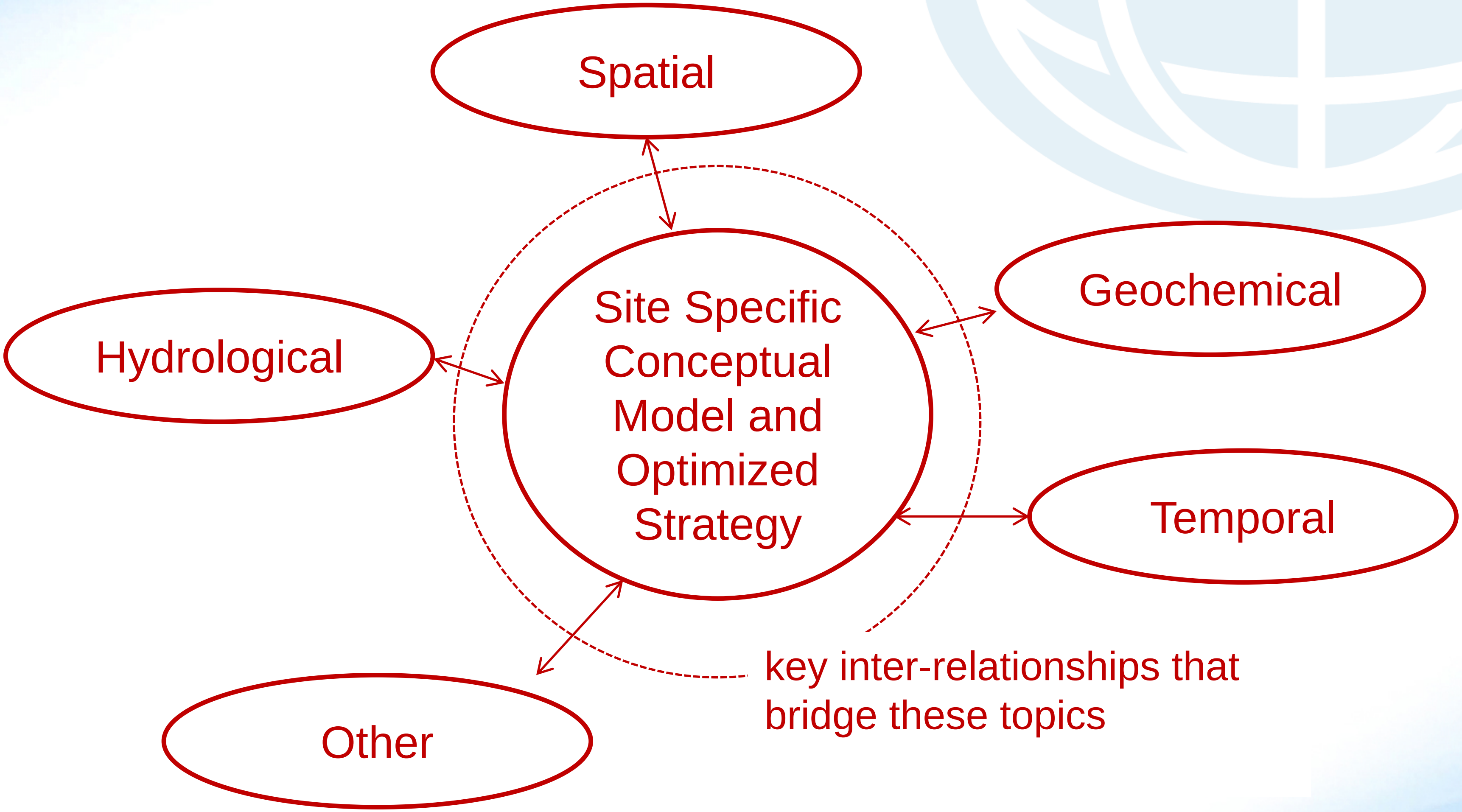
In evaluating the available data, the team developed an overarching set of frameworks

Frameworks provide a consistent way of organizing and interpreting complex data in a manner that supports environmental decision making

The frameworks support and dovetail with existing conceptual models/approaches for UMTRCA sites

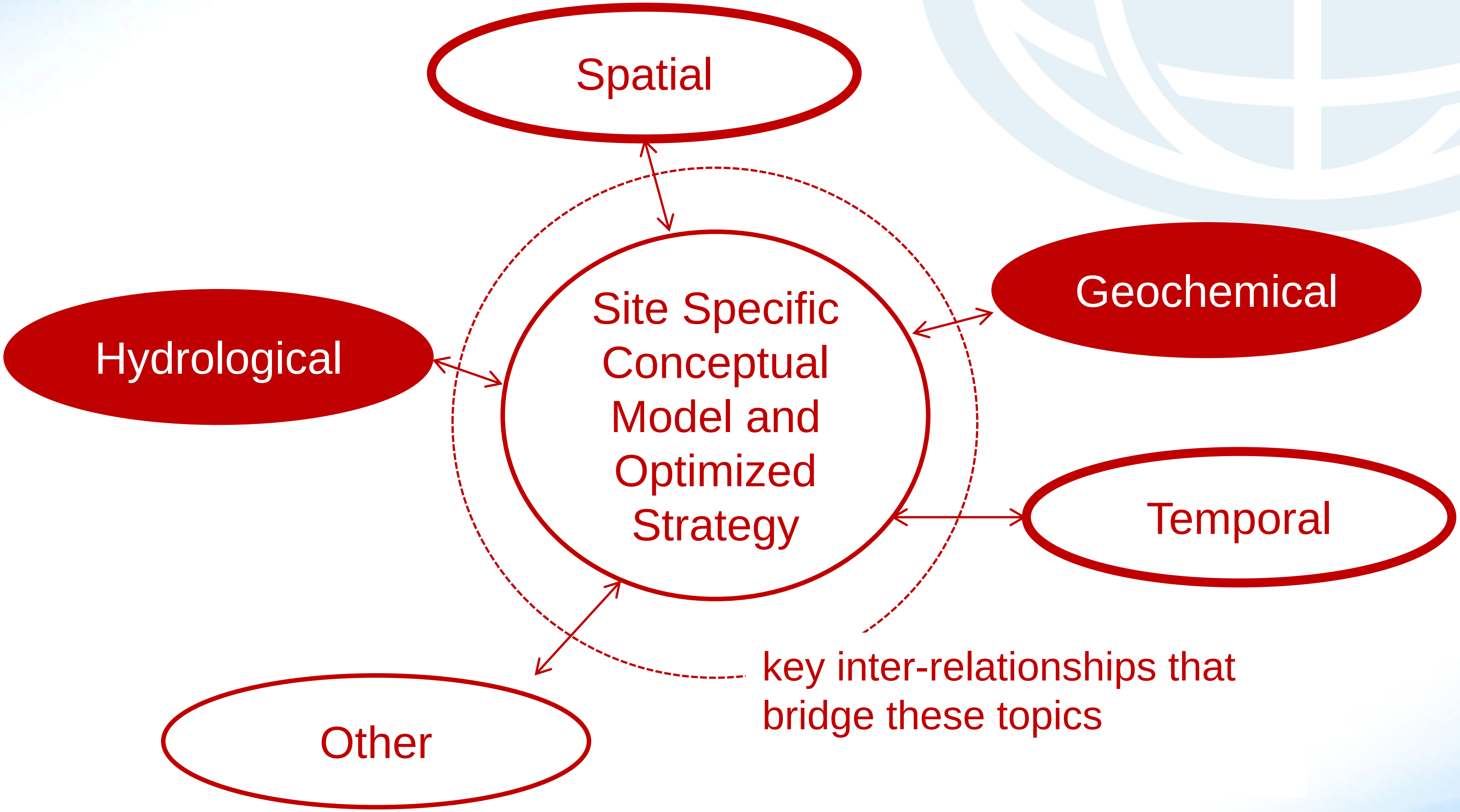
The objective was to identify scientific and technical areas of opportunity for the DOE-LM program.

Example frameworks for UMTRCA groundwater



key inter-relationships that bridge these topics

Example frameworks for UMTRCA groundwater



Spatial

Hydrological

Site Specific Conceptual Model and Optimized Strategy

Geochemical

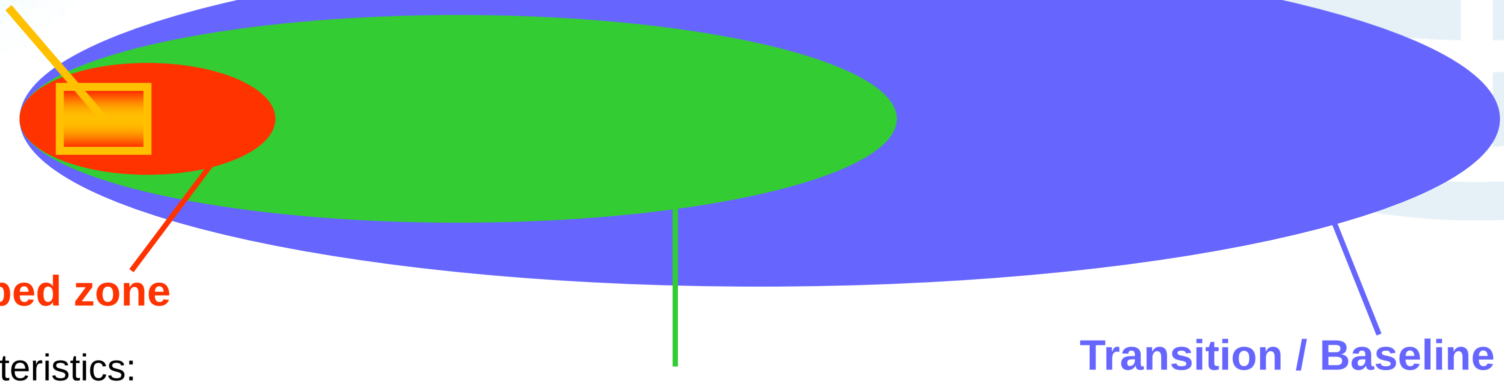
Temporal

Other

key inter-relationships that bridge these topics

Applied Environmental Science Philosophy:

Anatomy of an impacted site



Facility

Disturbed zone

<u>Characteristics</u>:
Perturbed conditions (chemistry, solids, etc.)

<u>Need:</u>
Eliminate or mitigate disturbance by active engineered solution or improved design

Impact zone

<u>Characteristics:</u>
Area with observable and easily detectable facility impacts

<u>Need:</u>
Characterization data to quantify potential impacts and mitigation activities, as needed, to provide environmental protection

Transition / Baseline zone

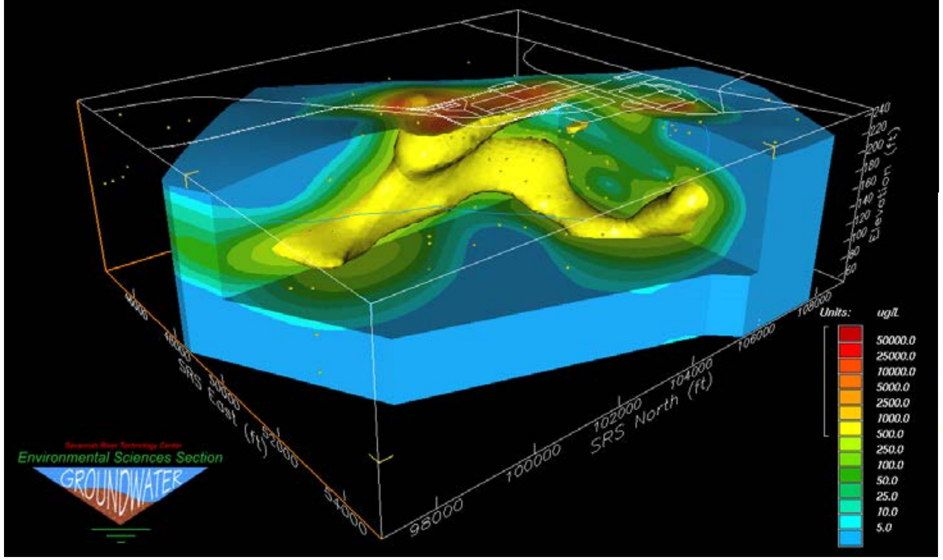
<u>Characteristics:</u>
Area where impacts are minimal or undetectable and conditions are similar to unimpacted settings

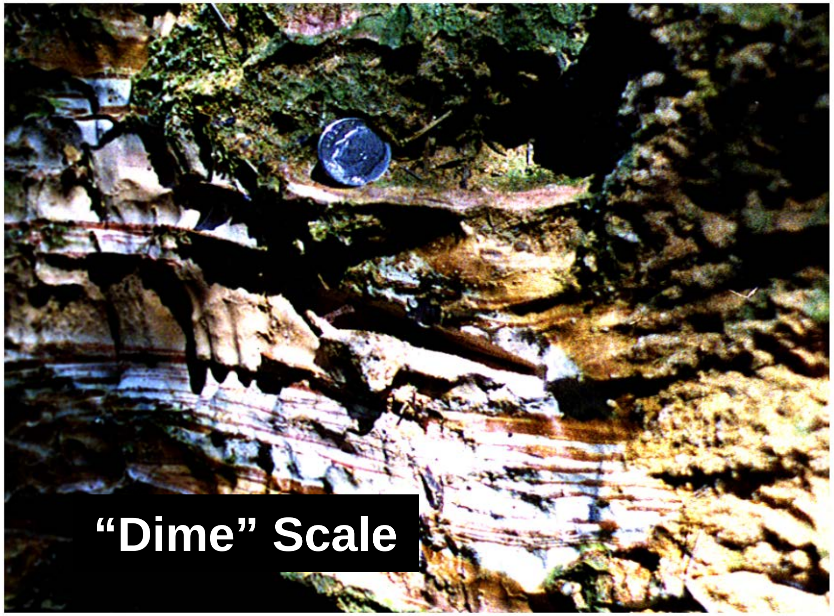
<u>Need:</u>
Careful characterization to provide a baseline for understanding impacts, development. Application of sensitive methods and early warning tools. Fundamental science!

Groundwater

Plume Scale

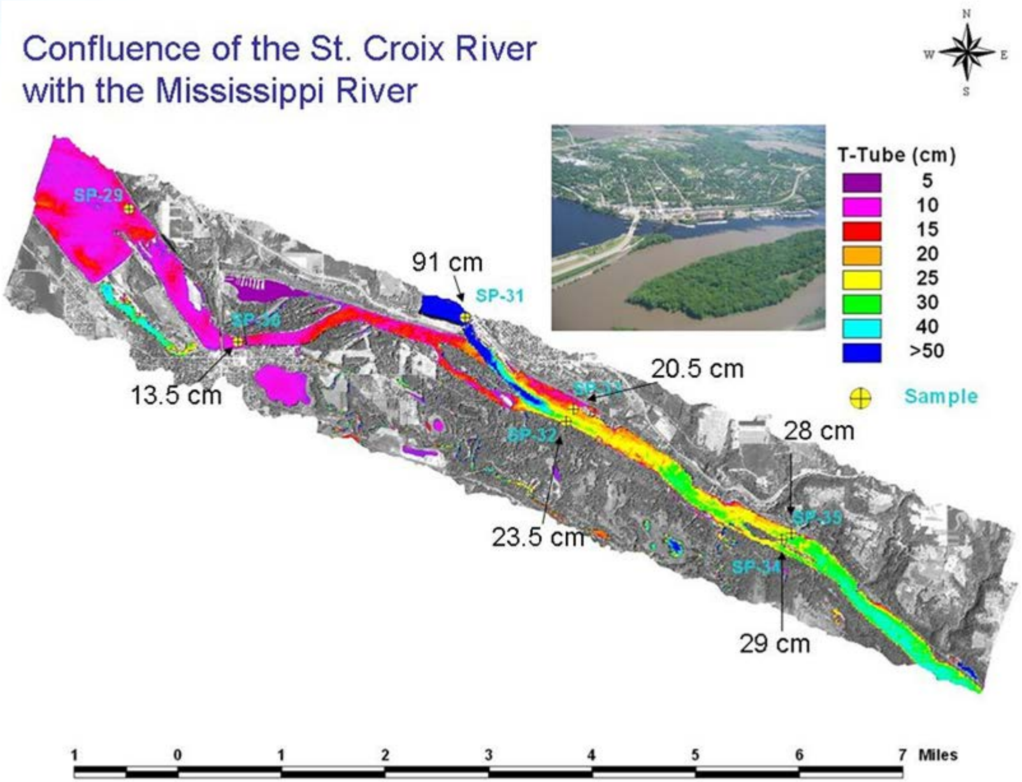
(TCE in groundwater beneath A/M Area of SRS)



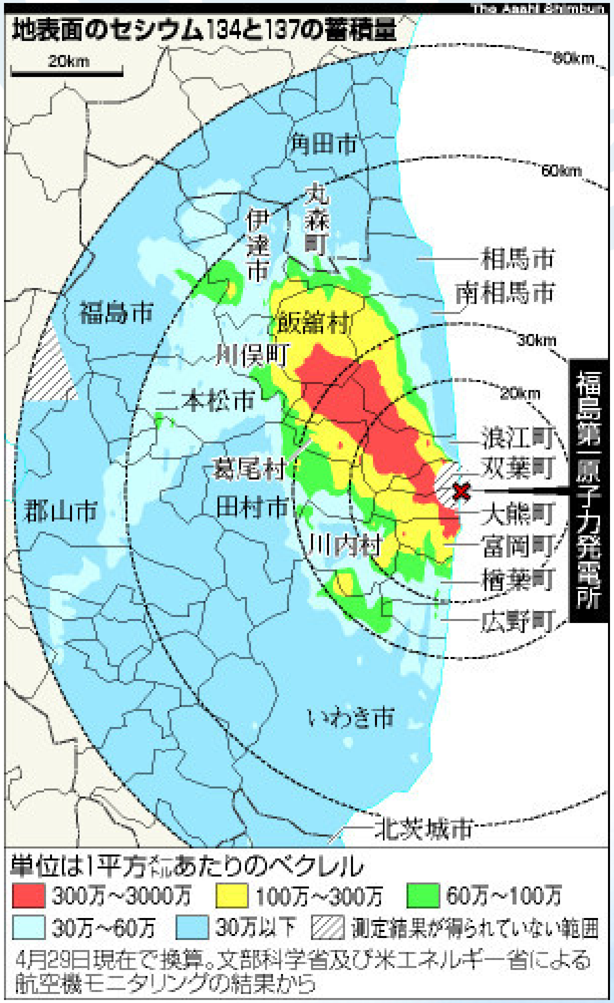


“Dime” Scale

Surface Water and Air



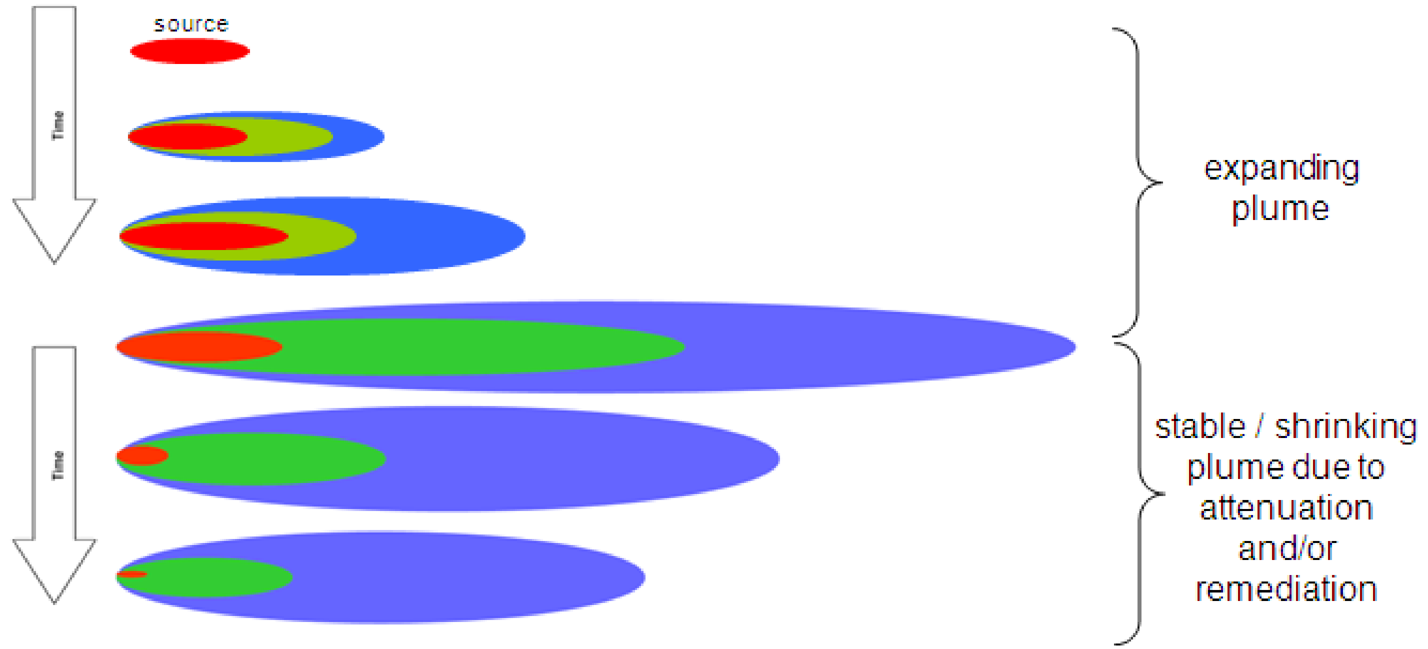
“River” Scale



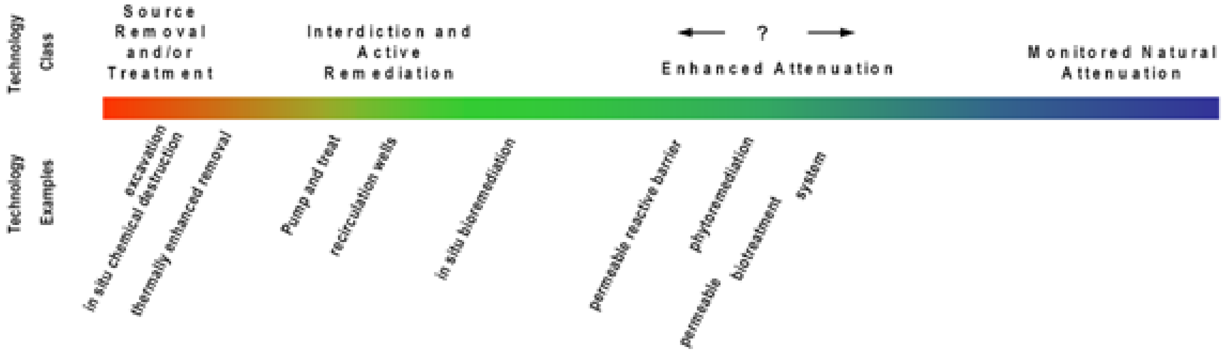
Fukushima

Lifecycle of a Contaminant Plume

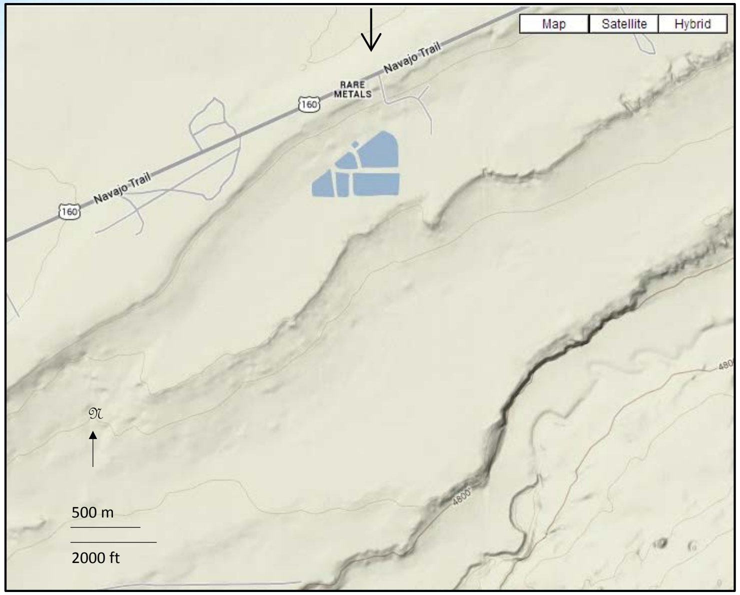
a) simplified representations of a groundwater plume in space and time



b) potential remedial technologies



Tuba City Mill/Tailings Site



Uranium mill operated 1956 through 1966 – various extractions were used

Processed ≅ 800,000 tons of uranium ore

Mill tailings slurried to ponds (33.5 acres or 136,000 m^2) for evaporation /disposal

Surface remediation of site began in 1988 and completed in 1990.

Tailings, debris and windblown material consolidated in engineered tailings cell

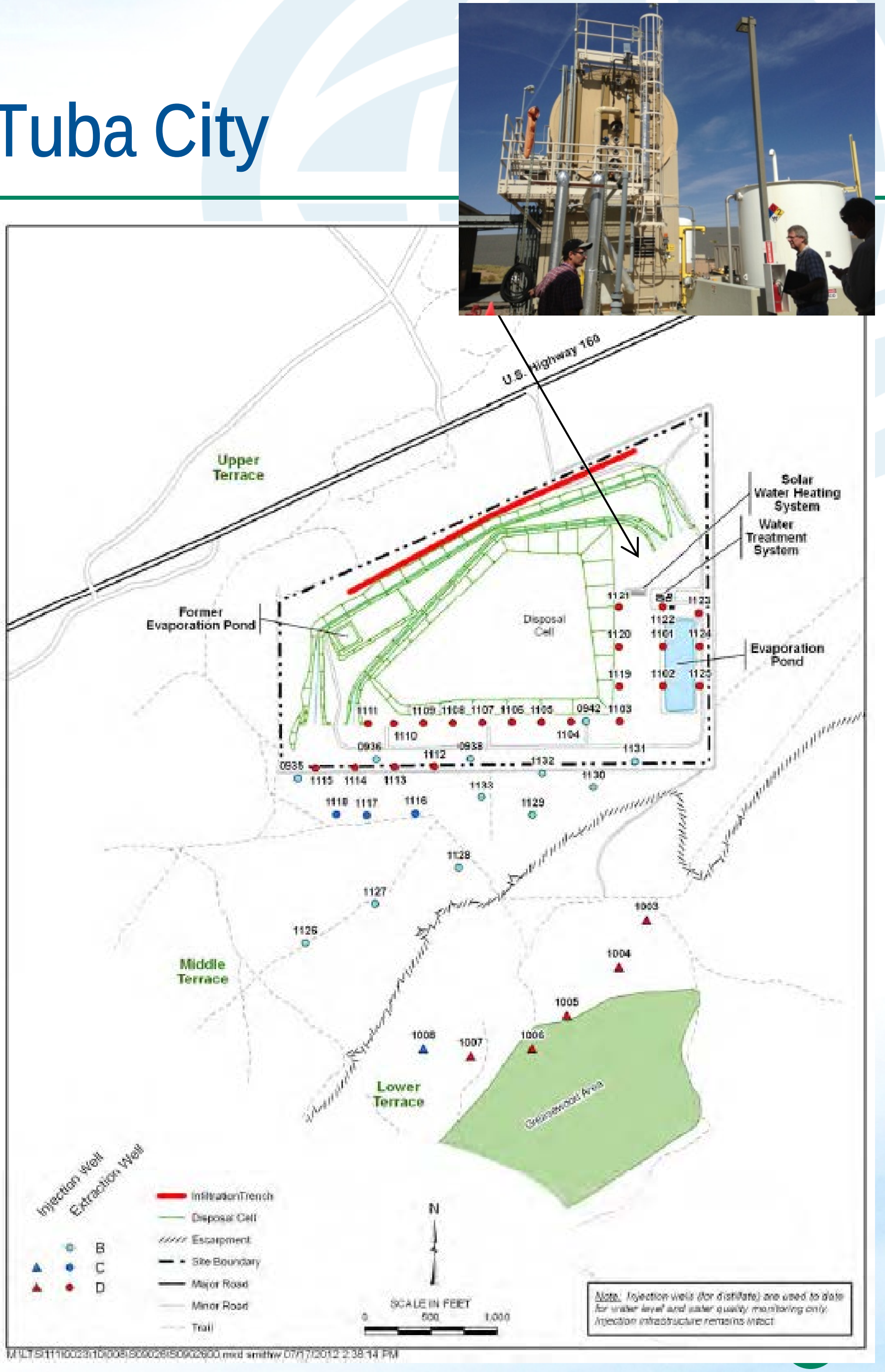
Contaminated groundwater at Tuba City

For contaminated groundwater, 37 recovery wells extract ≅ 100 gpm (375 L/min) which is treated using an advanced distillation process.

The treated water is reinfiltrated and the brine/reject is sent to a lined evaporation pond.

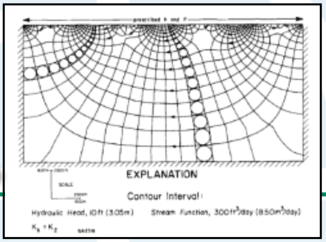
The groundwater treatment system has operated for over 10 years.

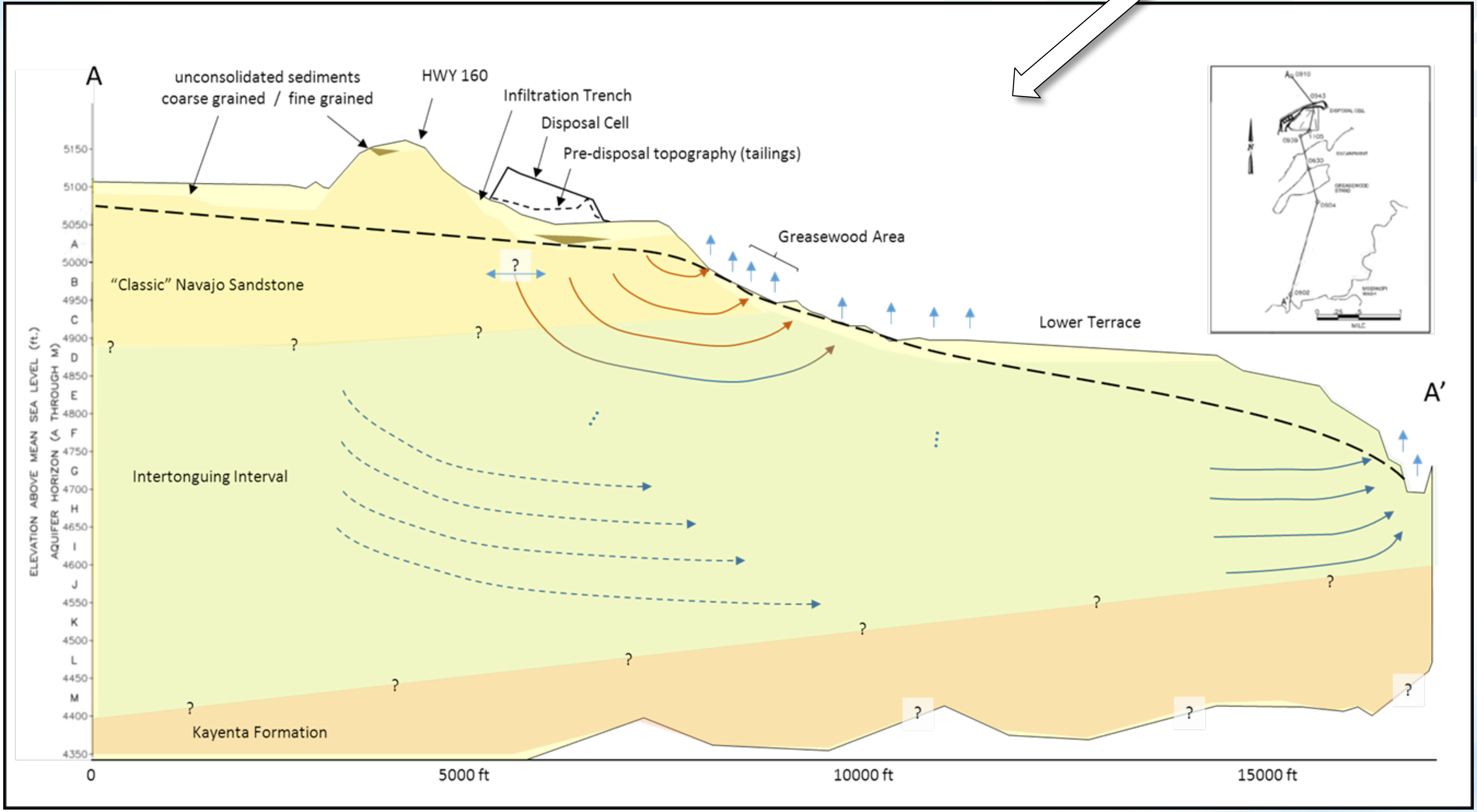
The system has had limited impact on the plume.



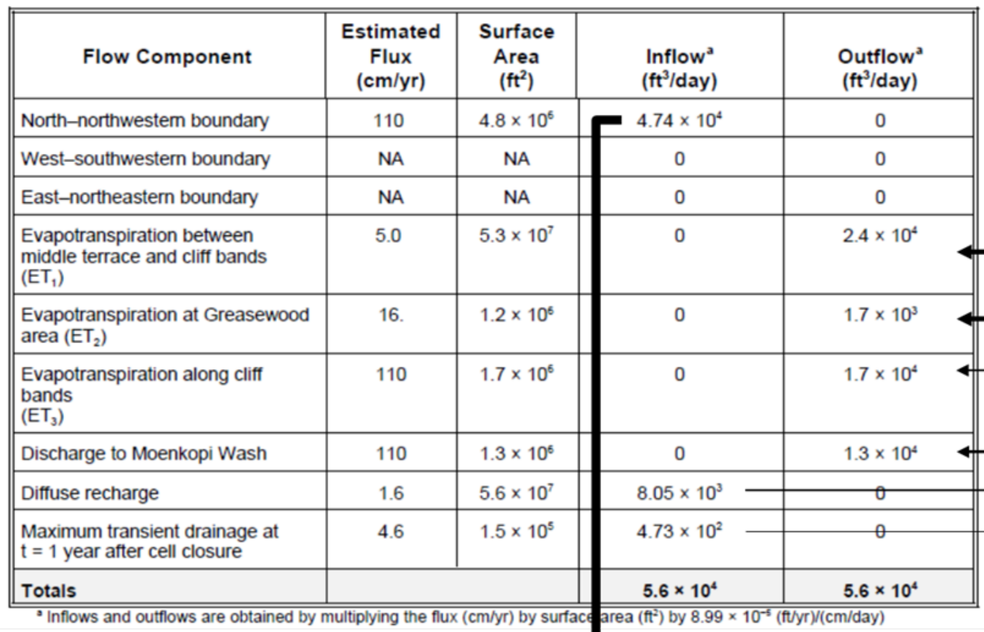
Where does water go?

Hydrological framework – flow lines in an arid terraced setting





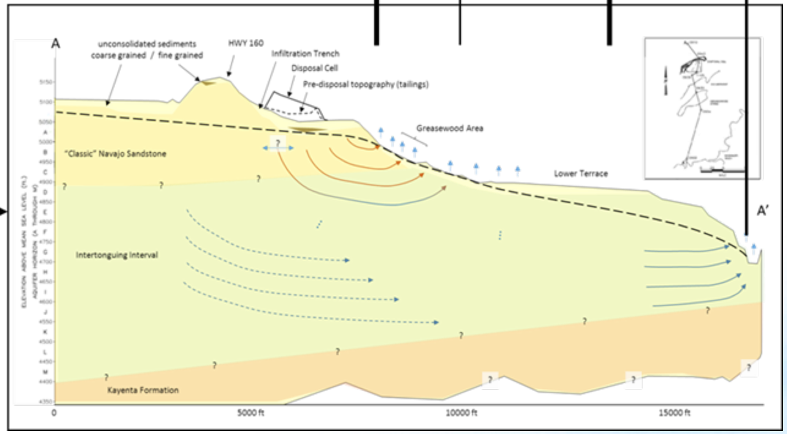
Water Balance Estimate



Flow Component	Estimated Flux (cm/yr)	Surface Area (ft²)	Inflow[a] (ft³/day)	Outflow[a] (ft³/day)
North–northwestern boundary	110	4.8×10^6	4.74×10^4	0
West–southwestern boundary	NA	NA	0	0
East–northeastern boundary	NA	NA	0	0
Evapotranspiration between middle terrace and cliff bands (ET_1)	5.0	5.3×10^7	0	2.4×10^4
Evapotranspiration at Greasewood area (ET_2)	16.	1.2×10^6	0	1.7×10^3
Evapotranspiration along cliff bands (ET_3)	110	1.7×10^6	0	1.7×10^4
Discharge to Moenkopi Wash	110	1.3×10^6	0	1.3×10^4
Diffuse recharge	1.6	5.6×10^7	8.05×10^3	0
Maximum transient drainage at t = 1 year after cell closure	4.6	1.5×10^5	4.73×10^2	0
Totals			**5.6×10^4**	**5.6×10^4**

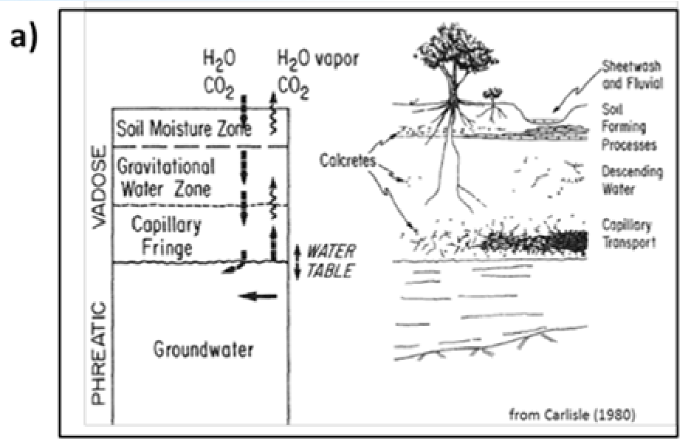
[a] Inflows and outflows are obtained by multiplying the flux (cm/yr) by surface area (ft²) by 8.99×10^{-5} (ft/yr)/(cm/day)

Water Balance (SOWP)

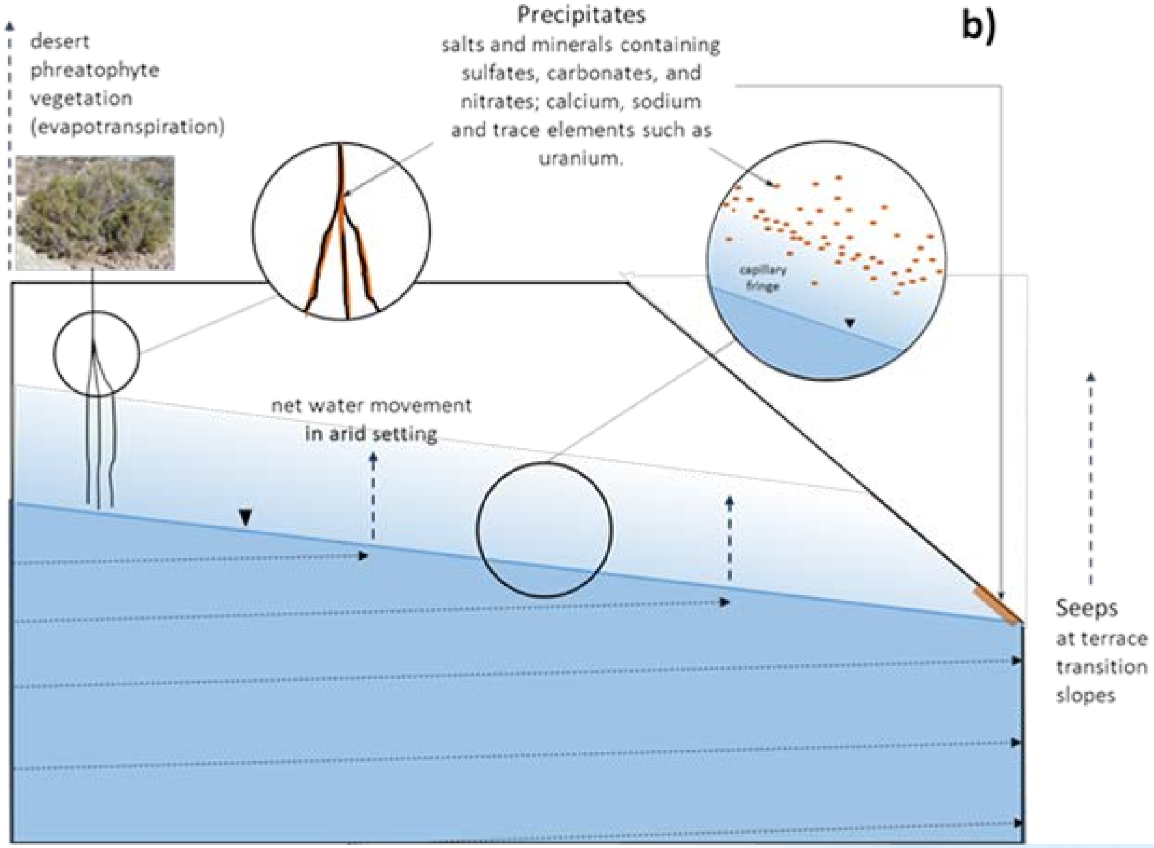


Where do contaminants go?

Linkage of hydrological and geochemical framework...



from Carlisle (1980)



Precipitates
salts and minerals containing sulfates, carbonates, and nitrates; calcium, sodium and trace elements such as uranium.

Summary (Mid-field Plume)

Groundwater contaminants are in the uppermost portion of the aquifer, and exit the groundwater as it flows into and beneath a lower terrace in the landscape

Contaminated water will not reach the Moenkopi Wash, a locally important stream.

Shallow groundwater in arid settings such as Tuba City is transferred into the vadose zone and atmosphere via evaporation, transpiration and diffuse seepage

Dissolved constituents will precipitate as minerals (similar to natural evaporite deposits)

Mineral accumulations will be in the deep vadose zone (near the capillary fringe), around the roots of phreatophyte plants, and near seeps.

Pump and Treat (P&T) Performance

An effective P&T remedy requires effective operation and efficient performance of the two major sub-activities

1) in the subsurface, extraction of contaminants and infiltration of clean water

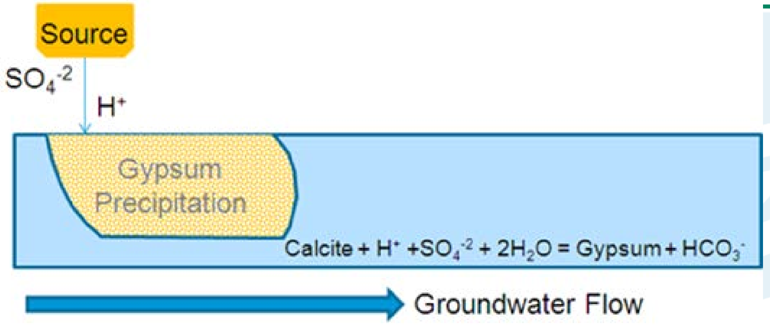
2) above ground, robust and cost effective water treatment and disposal of secondary wastes.

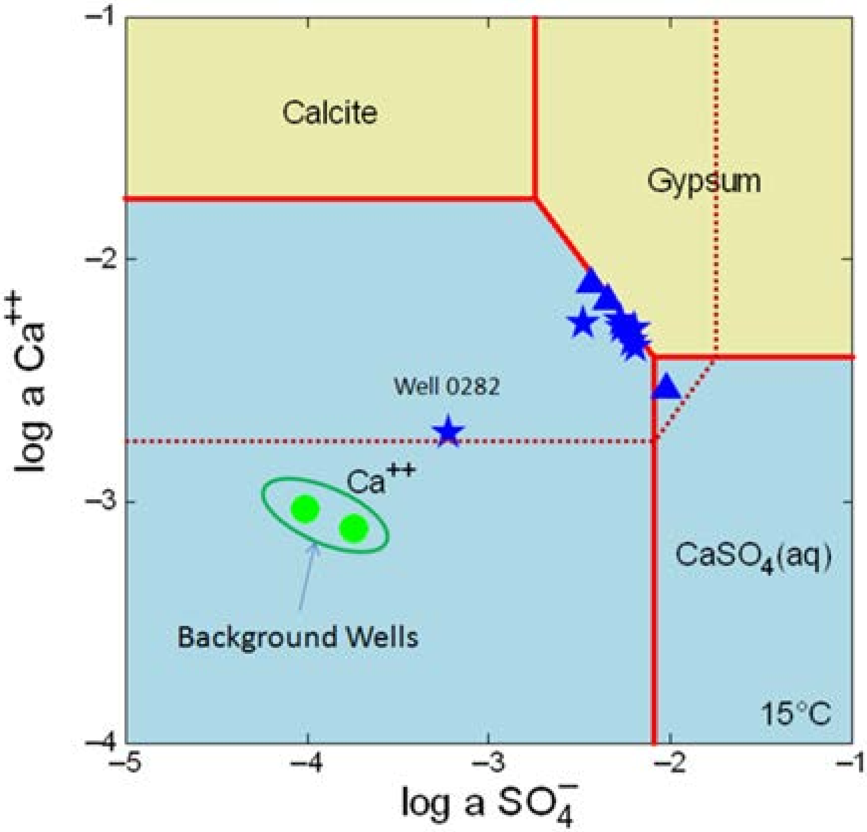
The Tuba City P&T system faces significant challenges, both in the subsurface and above ground

In the subsurface: near-field groundwater geochemistry

Impact on the P&T…

- **Acid infiltration from the original uranium extraction process**
- **Alkaline infiltration from the later carbonate extraction process**
- **Precipitates in the near-field result from complex and perturbed geochemistry**
- **Precipitates clog pores and reduce aquifer permeability, and impact pumping wells**
- **Precipitates serve as long term reservoir of contaminants**





In the subsurface: water extraction experience

Impact on the P&T…

- **Original design 100 gpm (375 L/min) ~~increasing to about 200 gpm (750 L/min)~~**
- **The bulk of the water is extracted from only a few of the recovery wells and a large number of the wells in the system do not produce significant amounts of water (e.g., 0.1 gpm or less).**
- **The uneven hydraulic performance indicates water flow through some areas (i.e., flushing) and not through other areas.**

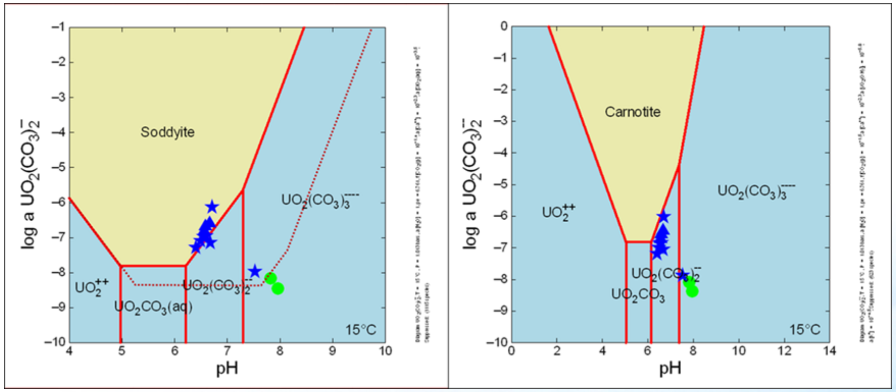
~~"flush one or two "pore volumes" of contaminated groundwater beneath the stabilized tailings…"~~

~~"added driving force of the water from the infiltration trench is intended to increase hydraulic gradients and to accelerate the flushing…"~~

In the subsurface: linkage of hydrology and geochemistry

The Kobayashi Maru…

- **For many contaminants, precipitated solid source material would extend the remediation timeframe until the minerals are dissolved and removed**
- **The portions of the plume that have been impacted by the highest levels of contamination would also have the most solids precipitation and would be the most important to flush. But these areas would produce less water while requiring many more flushing volumes to achieve remedial goals**
- **This discordance between fundamental natural controls/processes in the subsurface and “remediation performance needs” is a significant and overriding challenge**



Above ground: distillation system experience

Snapshot…

- **Contaminated groundwater separated – clean water and brine**
- **Uses a state-of-practice distillation system (a "falling-film vapor recompression distillation process") including a number of components/actions to optimize**
- **Despite high quality efforts, the treatment system has underperformed (e.g., operating less than 20% of the time in 2012) with high operation and maintenance costs**

- **If P&T operations continue DOE has several options to reduce cost and improve operational robustness (e.g., reverse osmosis)**

Next step – confirming the alternative conceptual model

- Confirming the evapotranspiration discharge boundary zone and incorporating it into the site conceptual model are keys to the development of protective and cost effective site strategies.
- A focused characterization study of the critical transition region from the middle to lower terrace and the lower terrace to the Greasewood area → technical basis for productive stakeholder and regulatory interactions.
- The listed "arid-site" processes produce distinctive geochemical signatures
- A few examples from several sites follow…

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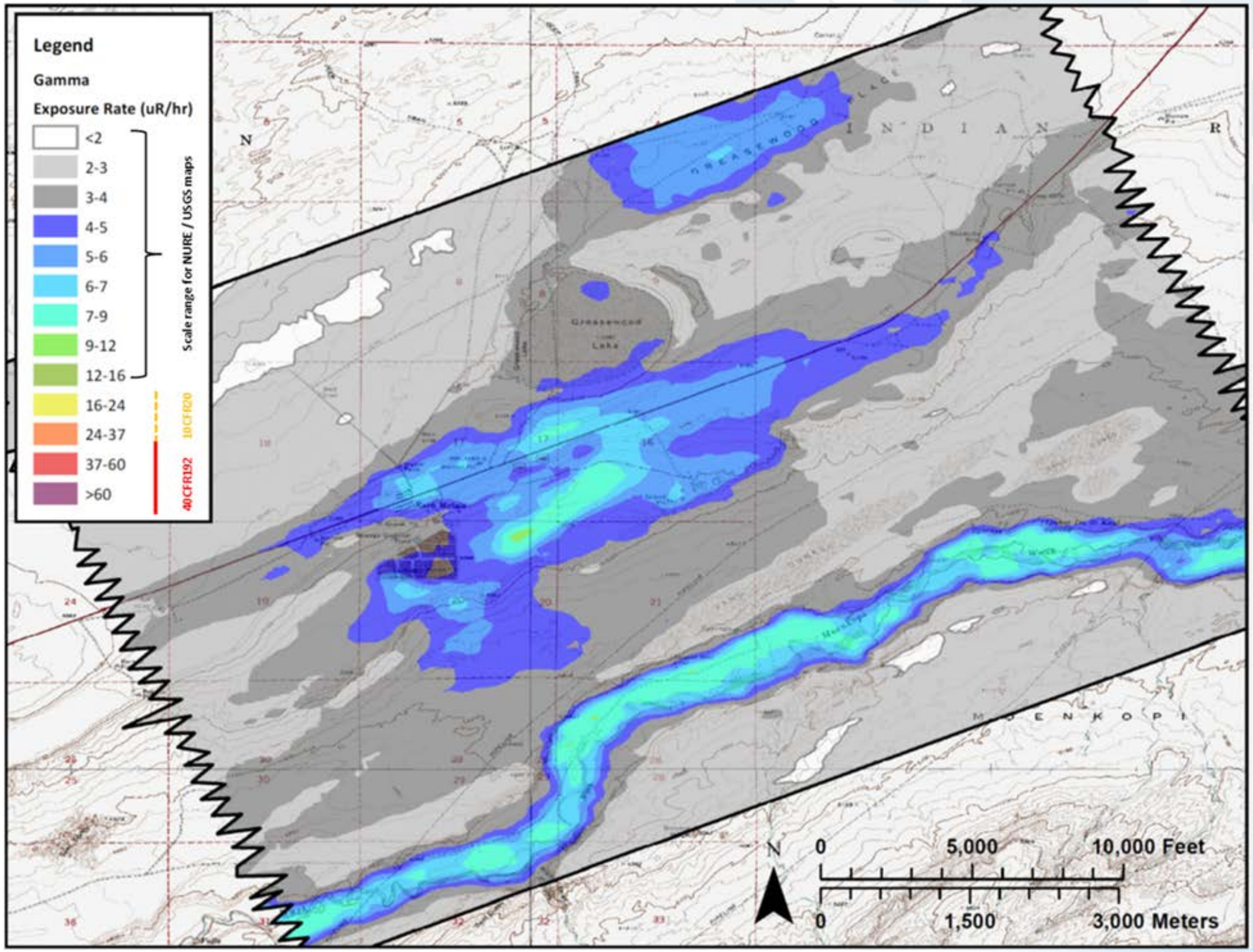
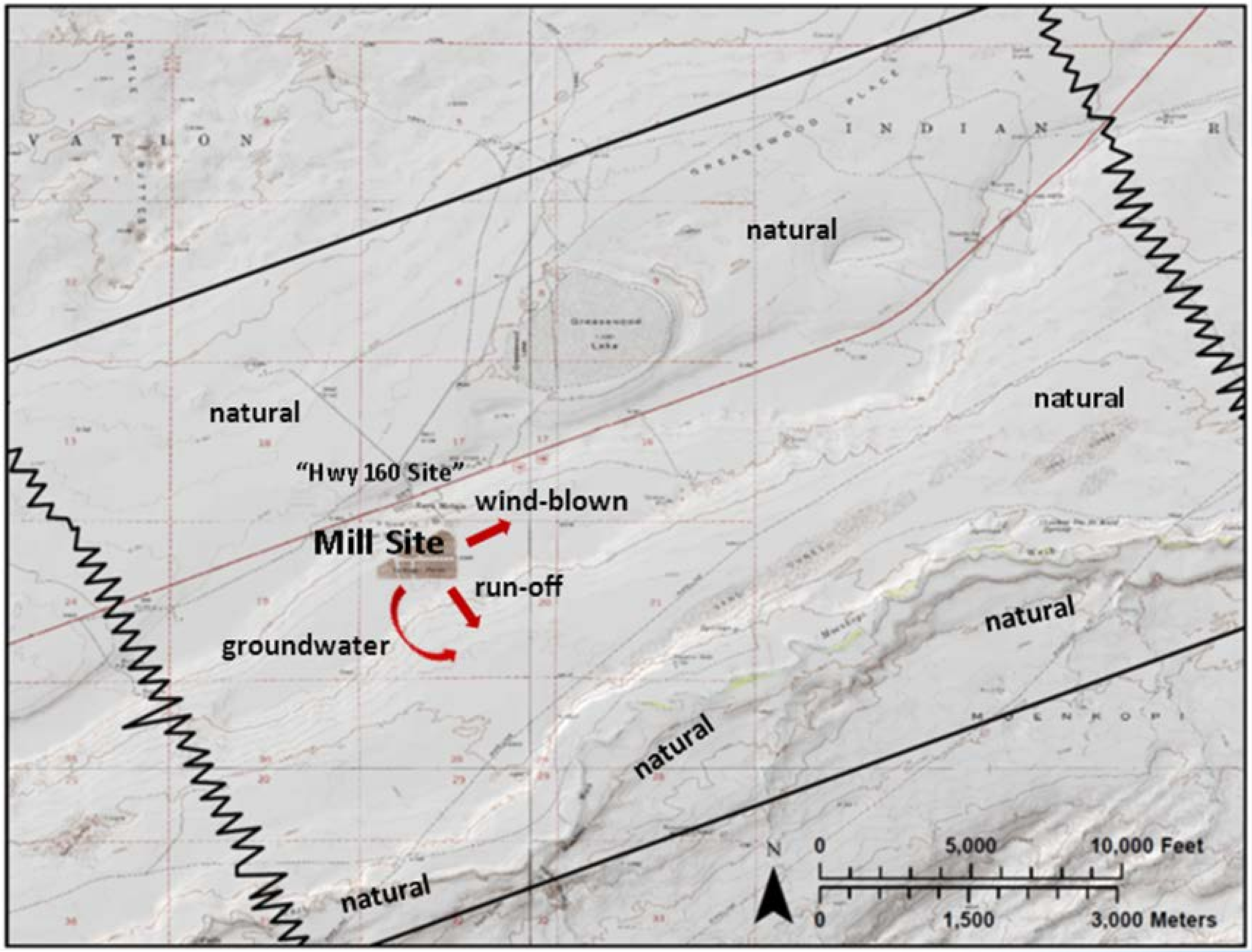
Confirming Transport Pathways – Tuba City

Aerial Gamma Survey



Confirming Transport Pathways – Tuba City

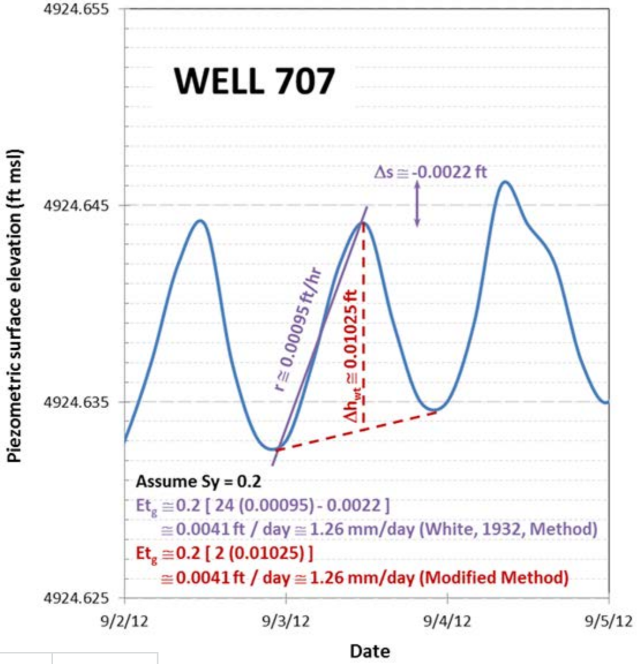
Aerial Gamma Survey Implications

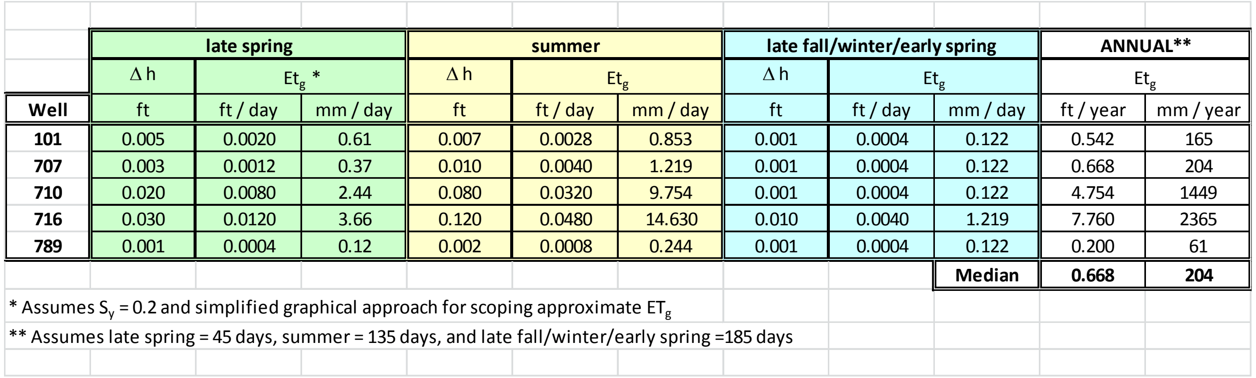


Confirming ET– Riverton WY

Confirming ET – Riverton WY

Diurnal water table oscillations confirm evapotranspiration and provide some quantitative information



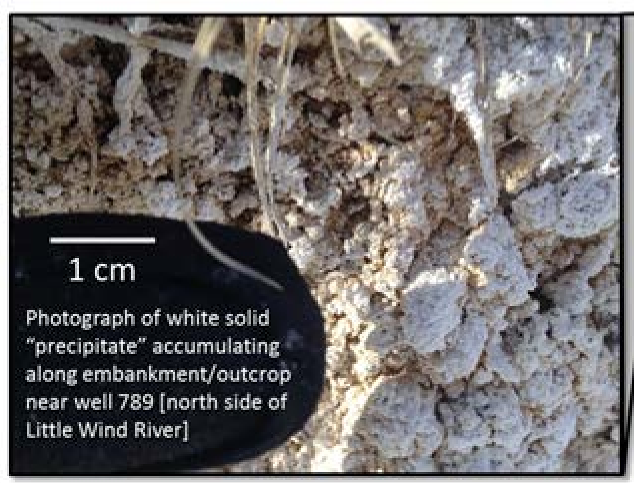


	late spring			summer			late fall/winter/early spring			ANNUAL**	
	Δ h	Et_g *		Δ h	Et_g		Δ h	Et_g		Et_g	
Well	ft	ft / day	mm / day	ft	ft / day	mm / day	ft	ft / day	mm / day	ft / year	mm / year
101	0.005	0.0020	0.61	0.007	0.0028	0.853	0.001	0.0004	0.122	0.542	165
707	0.003	0.0012	0.37	0.010	0.0040	1.219	0.001	0.0004	0.122	0.668	204
710	0.020	0.0080	2.44	0.080	0.0320	9.754	0.001	0.0004	0.122	4.754	1449
716	0.030	0.0120	3.66	0.120	0.0480	14.630	0.010	0.0040	1.219	7.760	2365
789	0.001	0.0004	0.12	0.002	0.0008	0.244	0.001	0.0004	0.122	0.200	61
									Median	**0.668**	**204**

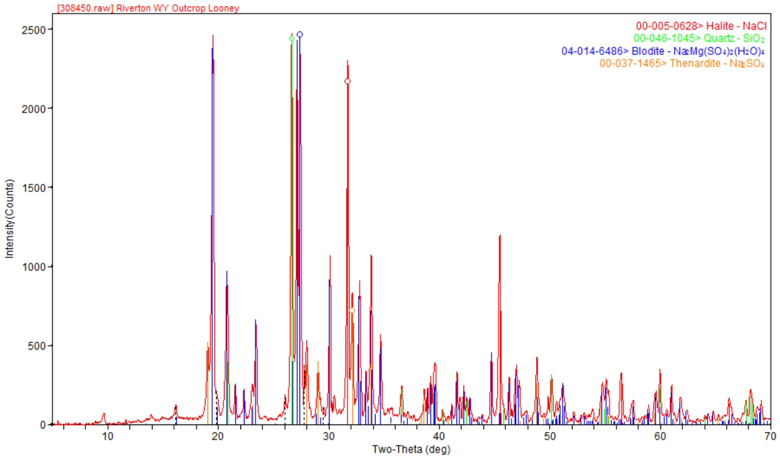
* Assumes $S_y = 0.2$ and simplified graphical approach for scoping approximate ET_g

** Assumes late spring = 45 days, summer = 135 days, and late fall/winter/early spring =185 days

Confirming mineral precipitation – Riverton WY



Photograph of white solid "precipitate" accumulating along embankment/outcrop near well 789 [north side of Little Wind River]



Sample mineral composition (mass fractions)

		Raw Sample Results (XRD and XRF)		**Best Estimates (XRD and XRF) *****		
		total (normalized)	fine powder excl "soil"	**fine powder excl "soil"**		
soil contribution*	**quartz -- SiO_2**	**0.14**	--	**--**	**--**	**--**
	Al2O3	**0.02**	--	**--**	**--**	**--**
	Fe2O3	**0.01**	--	**--**	**--**	**--**
Halite	**NaCl**	**0.04**	0.05	**0.04**	**to**	**0.05**
Blödite	**$Na_2Mg(SO_4)_2 : 4H_2O$**	**0.63**	0.76	**0.66**	**to**	**0.76**
Thenardite / Mirabilite	**Na_2SO_4 / $Na_2SO_4 : 10H_2O$**	**0.10**	0.12	**0.24**	**to**	**0.12**
Other evaporites**		**0.06**	0.07	**0.06**	**to**	**0.07**

Assumptions:

shaded cells are assumed values

* quartz grains (SiO2), Al2O3 and Fe2O3 represent native soil; remaining fine powder material best represents evaporite mineral encrustation

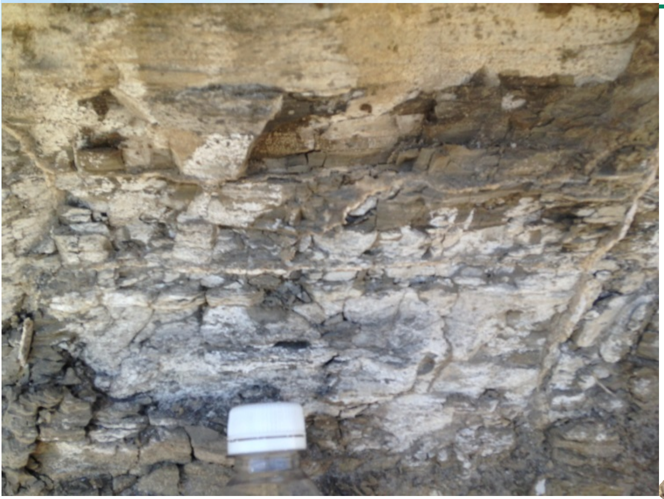
** Other = minor unquantified XRD peaks (e.g., glauberite, various sodium magnesium calcium sulfates and chlorides and related evaporite minerals)

*** Best estimate ranges based on thenardite / mirabolite hydration end members and inclusion of assumed fraction of "Other"

Element	LAB XRF		PORTABLE XRF	
S	**188591**	ppm	82000	ppm
Na	**162022**	ppm	--	
Si	**77735**	ppm	44100	ppm
Cl	**52845**	ppm	49700	ppm
Mg	**47759**	ppm	13400	ppm
Ca	**21125**	ppm	12600	ppm
Al	**14328**	ppm	4720	ppm
Fe	**6150**	ppm	2800	ppm
K	--		3360	ppm
P	1612	ppm	< 400	ppm
Ti	380	ppm	472	ppm
Ba	271	ppm	< 32	ppm
Zr	208	ppm	120	ppm
Sr	194	ppm	100	ppm
Mn	132	ppm	44	ppm
Br	91	ppm	--	
U	*64*	ppm	*73*	ppm
Sc	--		46	ppm
Co	40	ppm	< 32	ppm
Rb	27	ppm	20	ppm

…

Confirming mineral precipitation – Shiprock NM



48 samples of evaporite minerals collected and reported by Stoller in 2000. Despite differences in setting, the elemental composition and minerals present at Shiprock similar to Riverton

Summary – Potential for alternative end states

- Hydrological and geochemical controls in arid environments (many mining and milling sites) work together to limit the size of groundwater plumes and can extend plume flushing times
- Incorporation of these concepts may support formulation of alternative end states
- At several DOE-LM sites, land and facilities have been transferred to the local community for transitioning to beneficial reuse and development.
- Some of the DOE-LM UMTRCA sites appear to be good candidates for brownfield reuse and to serve as a resource for future employment and ongoing benefit to stakeholders and Native American Nations.