



SRR-CWDA-2016-00112

October 20th, 2016

Interagency Steering Committee on P&RA
CoP Annual Technical Exchange Meeting



EVALUATION & INTERPRETATION OF PROPERTIES FOR EMPLACED SALTSTONE

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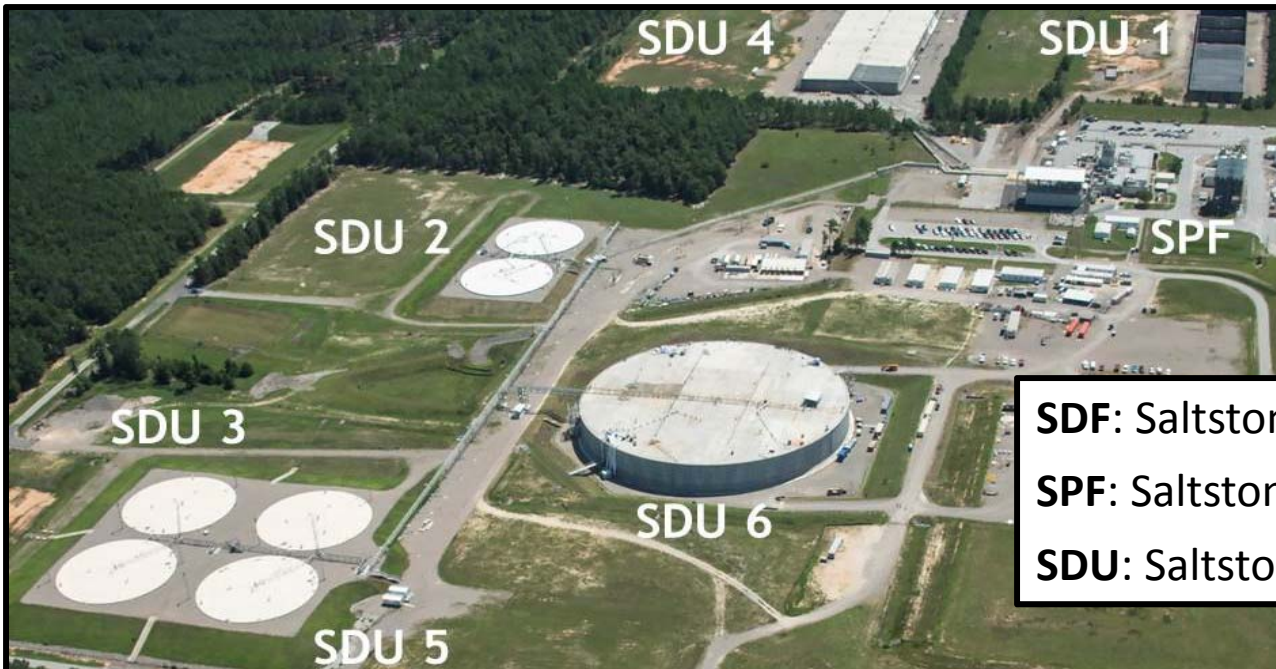
Overview

We do the right thing.

- **Background:**
 - Saltstone Disposal Facility (SDF) & Performance Assessment (PA)
- **Objectives for Extracting SDU-emplaced Saltstone**
 - Laboratory vs. field comparison
- **Preparation for Extracting SDU-emplaced Saltstone**
 - Multiple mock-ups, optimization, operator training, etc.
- **Core Extraction and Analysis**
 - Physical properties (density and porosity)
 - Key PA-related properties (K_{sat} and K_d)
- **PA Impacts**
 - Comparison of data for actual saltstone compared to PA modeling values

SDF Background

We do the right thing.

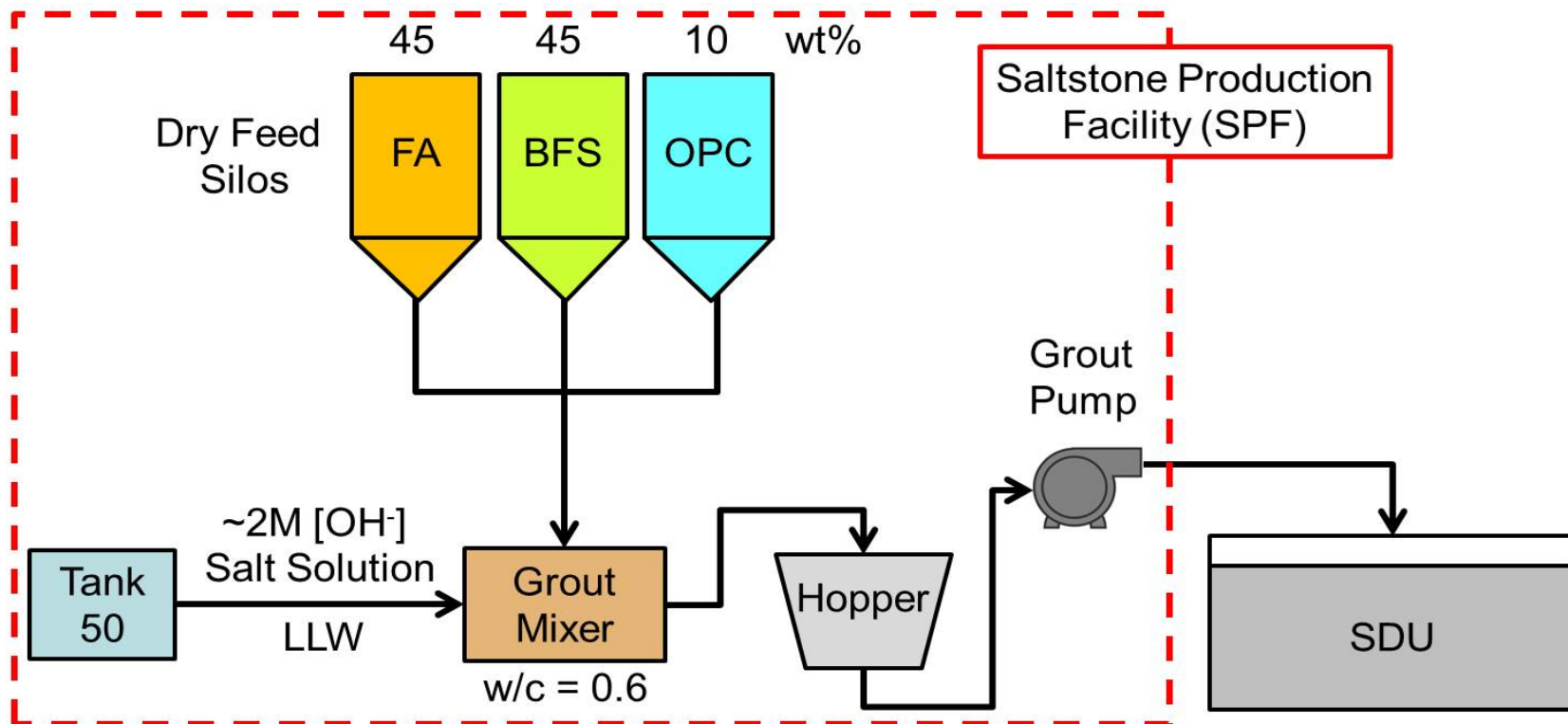


SDF: Saltstone Disposal Facility
SPF: Saltstone Production Facility
SDU: Saltstone Disposal Unit

- Liquid waste treatment at SRS will generate ≈ 100 million gallons of low-level salt solution containing $<0.1\%$ of total SRS radionuclide inventory.
- Salt solution combined with cementitious materials in the SPF to form saltstone grout \Rightarrow emplaced in permanent SDUs.
- The PA models fate and transport of constituents in SDUs to determine the long-term potential for future interactions with the environment and public.

Saltstone Processing

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- Time from mixing to emplacement \approx 5 min. depending on SDU-transfer distance (\approx 2000 ft.).
- Approximately 65,000 gallons of grout can be processed in 8-hour shift period.

Objectives

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- Most PA-modeled saltstone properties are derived from testing lab-prepared simulant samples ⇒ but are lab samples representative of field samples?

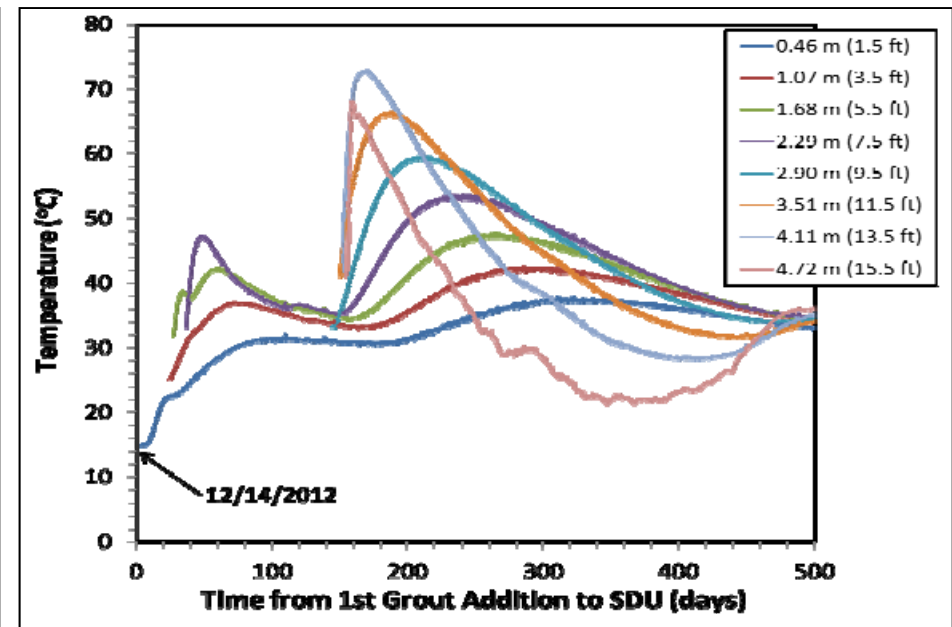
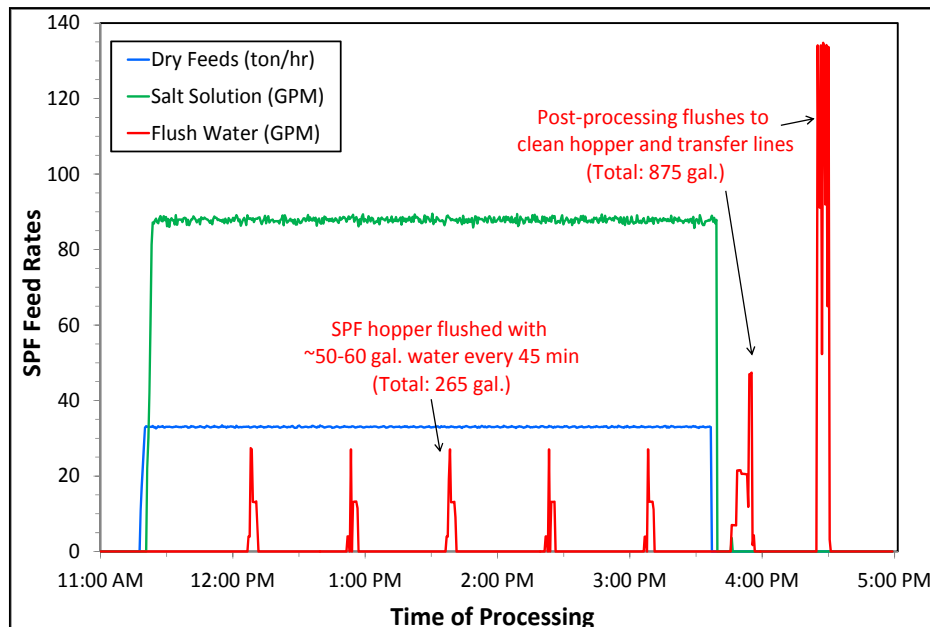
PARAMETER	LAB-PREPARED	FIELD-PROCESSED
<i>Dry Feed & Liquid Ratios</i>	<ul style="list-style-type: none"> Components accurately measured 	<ul style="list-style-type: none"> Bulk process – potential for processing variations: <ul style="list-style-type: none"> ⇒ Avg. dry feed rate ≈30 ton/hr (± 1%) ⇒ Avg. salt soln. feed rate ≈90 GPM (± 2%) ⇒ Periodic system flushing with water
<i>Mixing</i>	<ul style="list-style-type: none"> Small-scale lab batch mixing (≈1 gal.) 	<ul style="list-style-type: none"> Large-scale continuous mixer (≈135 GPM)
<i>Delivery</i>	<ul style="list-style-type: none"> Gently poured into curing molds 	<ul style="list-style-type: none"> Transferred through ≈2000 ft. pipe from SPF to SDU using peristaltic pump Free-falls into SDU and spreads to extremities
<i>Curing Environment</i>	<ul style="list-style-type: none"> Environmental chamber with controlled temperature and RH 	<ul style="list-style-type: none"> Curing temperature profile dependent on location in SDU
<i>Storage Environment</i>	<ul style="list-style-type: none"> Samples often maintained in sealed containers and/or inert, humidity-controlled environments 	<ul style="list-style-type: none"> Samples maintained in variable, field-emplaced environment

- Strategy is to extract emplaced saltstone from an SDU to compare to lab-processed samples.

SPF Processing & SDU Curing

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- SPF system flushes and variable SDU curing temperatures are potentially significant disparities between lab and field processing.



Laboratory vs Field Strategy

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- On August 12th, 2013 \approx 33,000 gallons of saltstone (3-inch lift) emplaced into SDU 2A at \approx 16-foot SDU elevation.

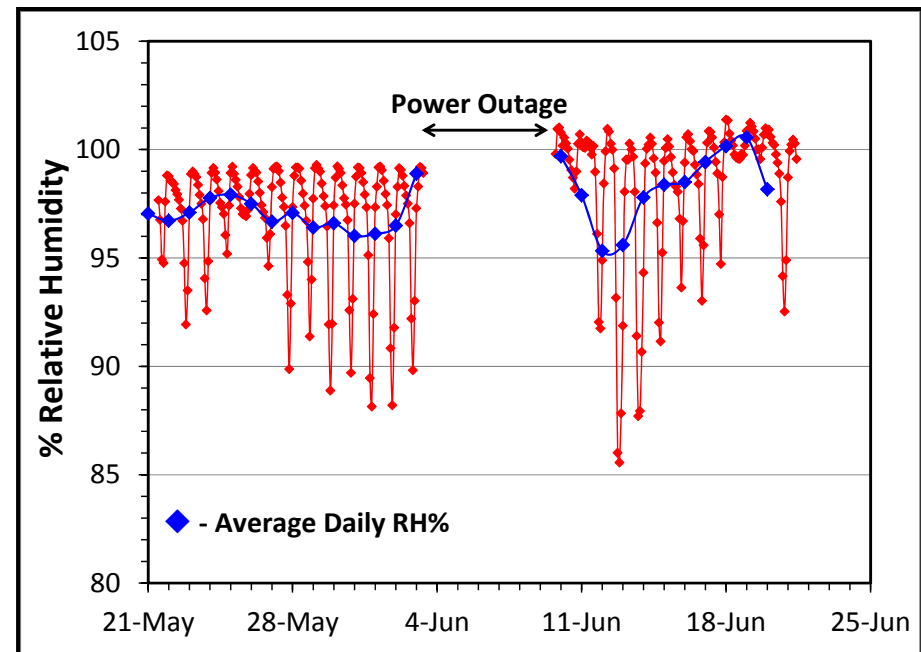
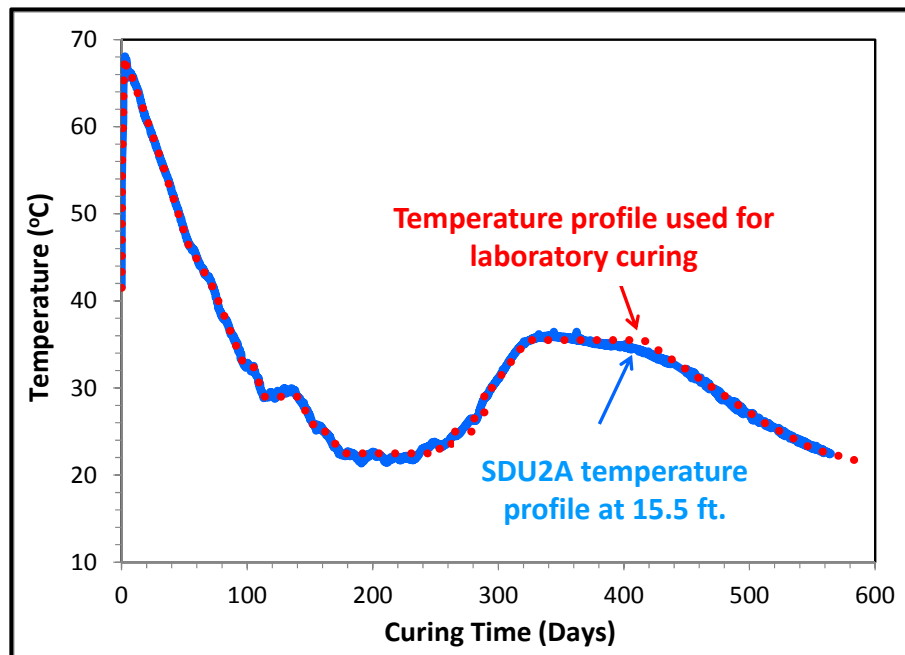


- Intent was to ultimately extract saltstone samples from around the 16-foot elevation and conduct property comparison with lab-processed samples.

Lab-Processed Samples

We do the right thing.

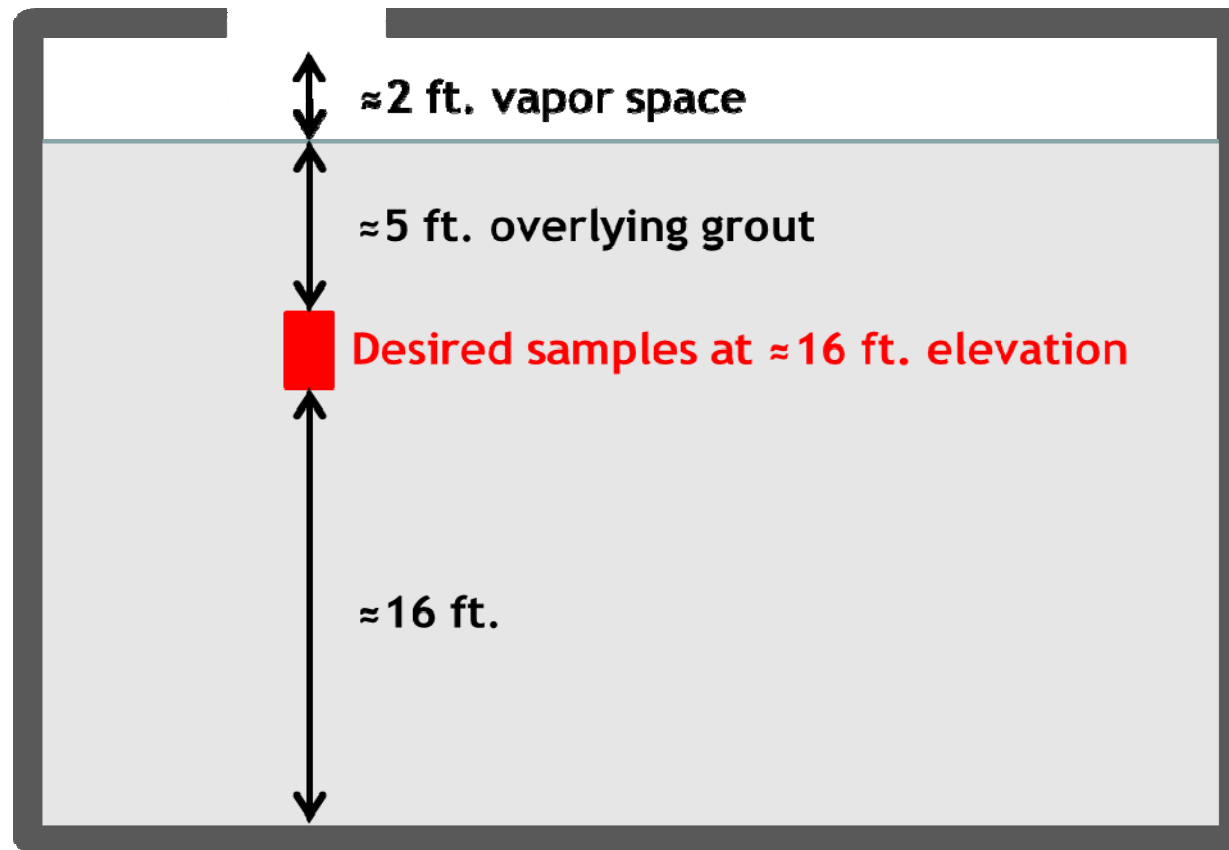
- Lab samples cured according to SDU2A measured environment.
- Thermowell temperature measurement at ≈ 16 -foot elevation in SDU2A.
- Relative humidity measured in SDU2A vapor space (used average RH 95%).



Planning for Sample Extraction

We do the right thing.

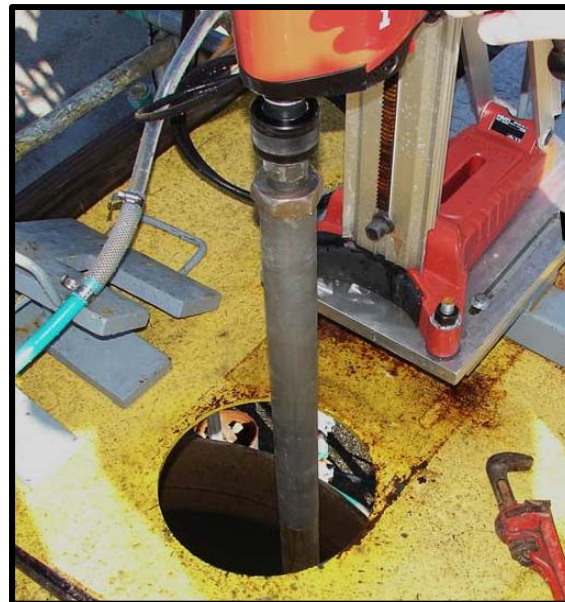
- 18 months dedicated to developing and optimizing techniques to drill and extract saltstone cores from SDU2A.



- **Three separate mock-ups were used to:**
 - Evaluate wet and dry core-drilling techniques
 - Test off-the-shelf and in-house fabricated drilling equipment options (motors, drill bits, etc.)
 - Simulate SDU configuration drilling via a camera port through 2-feet of vapor space and 5-feet of overlying grout
 - Overcome technical issues that compromised sample integrity: drill bit skidding, drill binding, sample fracture, etc.
 - Develop a technique and equipment to retrieve the sample from the core hole once drilling process complete
 - Simulate operator environment on SDU roof ⇒ no line of sight (reliant on video monitor), required to develop sense of touch
 - Allow operators to train in rad protection suits
 - Post-extraction sample inerting, storage, and transport.

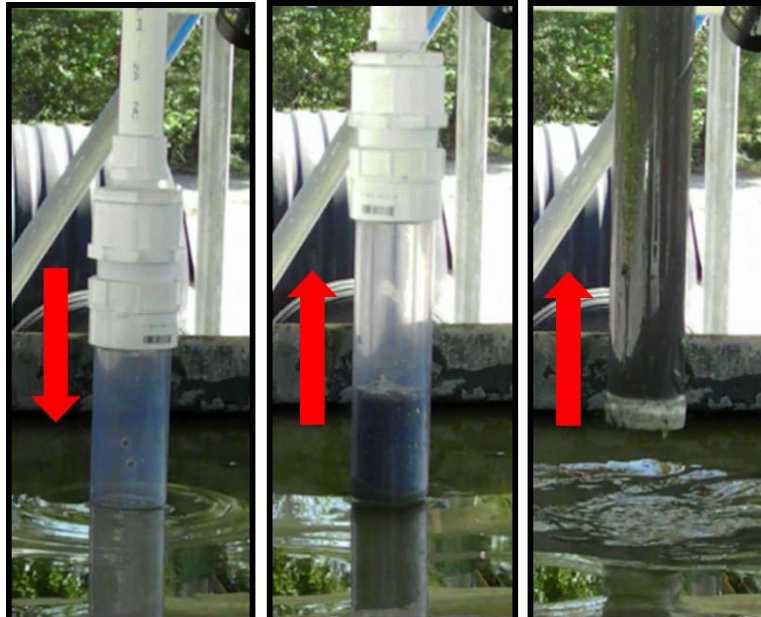
Mock-Up Core Drilling

We do the right thing.



Mock-Up Core Extraction

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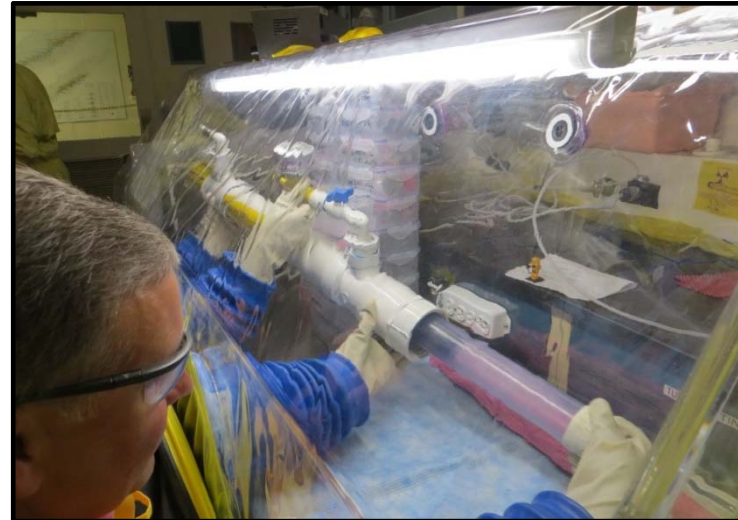
SDU2A Drilling & Extraction

We do the right thing.



Laboratory Storage

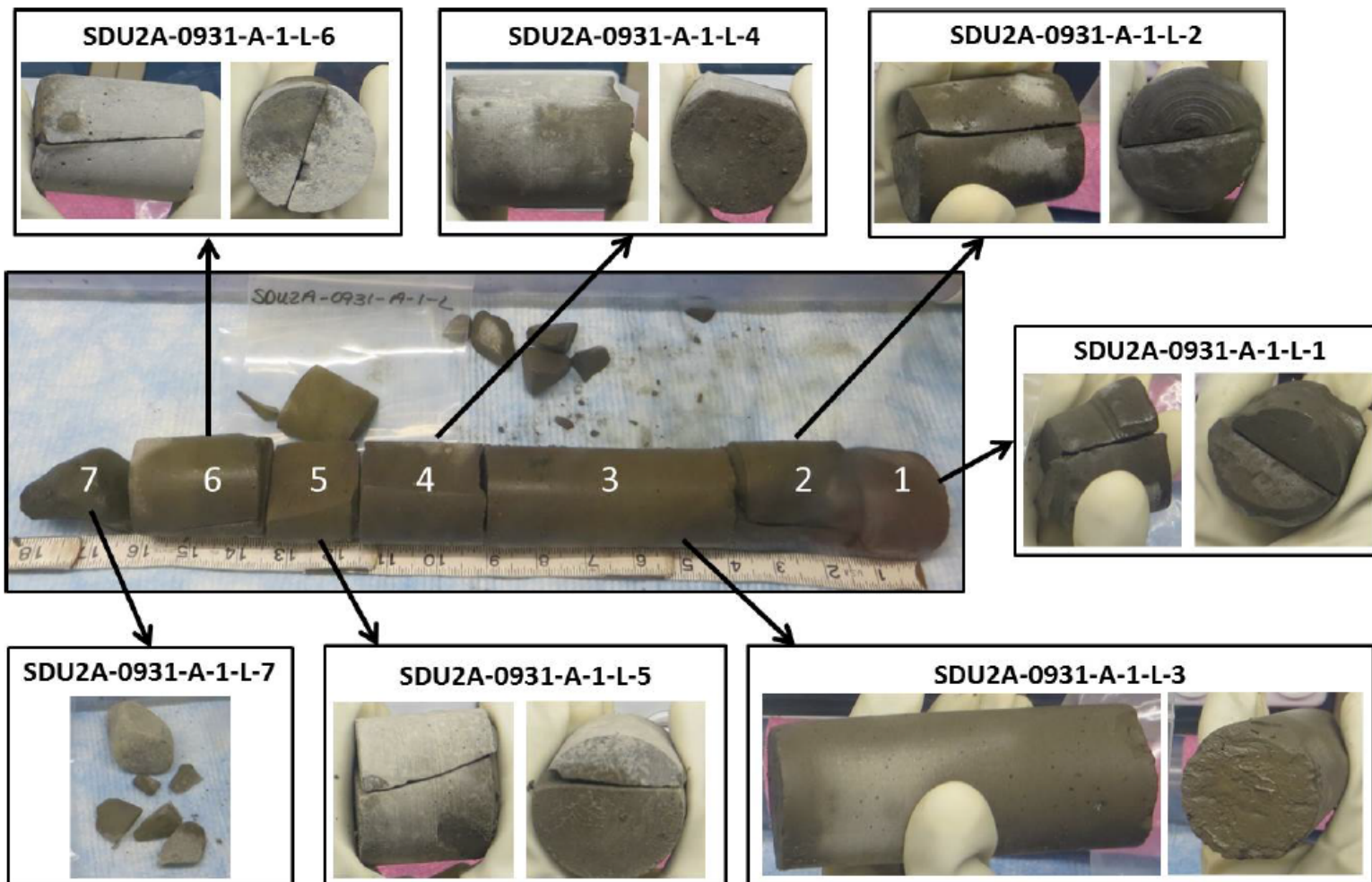
We do the right thing.



Core Appearance

We do the right thing.

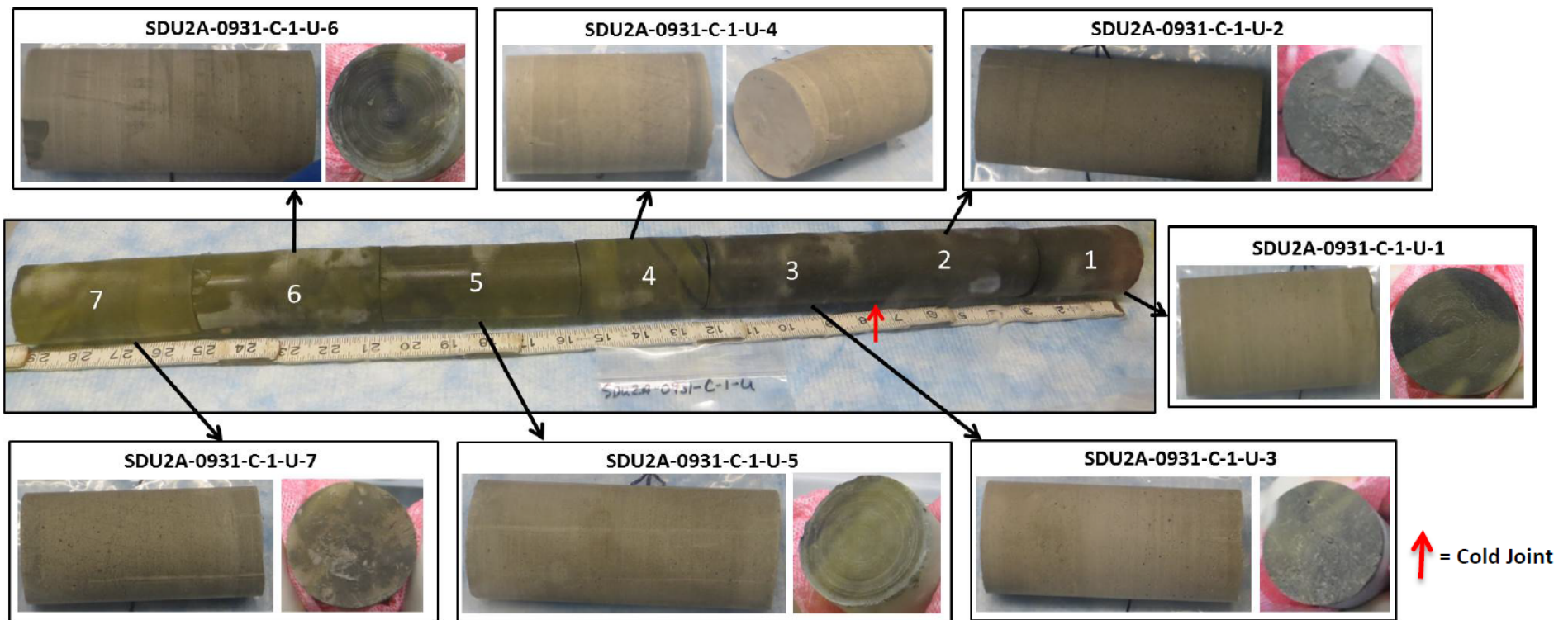
Sample ID: SDU2A-0931-A-1-L (Lower Core Portion Extracted from Port A – Drill Position 1 on May 6, 2015)



Core Appearance

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Sample ID: SDU2A-0931-C-1-U (Upper Core Portion Extracted from Port C – Drill Position 1 on April 28, 2015)



Sample Analyses

We do the right thing.

PROPERTY		RATIONALE
<i>Density / Porosity</i>		<ul style="list-style-type: none"> • Determine if variability of saltstone field- processing, transfer, and environment impact the physical properties. • How do changes in physical properties impact other performance indicators?
K_{sat}		<ul style="list-style-type: none"> • Saturated hydraulic conductivity of saltstone impacts: <ul style="list-style-type: none"> ⇒ groundwater ingress into saltstone, and ⇒ leaching of radionuclides/contaminants to the SDF environment
^{99}Tc	<i>Solubility (Anoxic)</i>	<ul style="list-style-type: none"> • ^{99}Tc designated as contributor to long-term, radiological dose to public. • PA assumes that under anoxic conditions ^{99}Tc is controlled by the solubility of reduced ^{99}Tc species.
	<i>K_d (Oxic)</i>	<ul style="list-style-type: none"> • In oxic environments ^{99}Tc is considered oxidized (and highly soluble) and leaching is controlled by adsorption.
^{90}Sr & ^{129}I <i>K_d (Oxic / Anoxic)</i>		<ul style="list-style-type: none"> • ^{90}Sr and ^{129}I designated as contributors to radiological dose to public. • In oxic/anoxic environments ^{90}Sr and ^{129}I leaching controlled by adsorption.

Physical Properties

Density, Porosity, & Water Content

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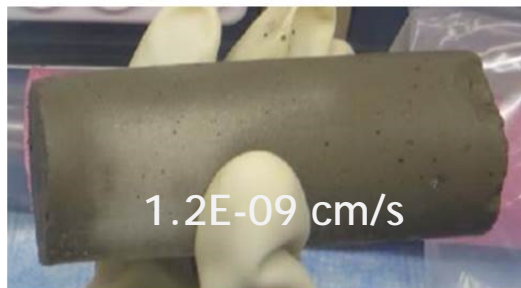
Sample Type	Bulk Density (g/cm ³)	Total Porosity (vol. %)	Open Porosity (vol. %)	Water Content (wt. %)
SDU2A (18 discrete sub-samples)	1.71 – 1.76 $\bar{x} = 1.73$	63.6 – 68.8 $\bar{x} = 65.8$	41.9 – 46.6 $\bar{x} = 43.7$	29.0 – 32.1 $\bar{x} = 30.3$
Lab-Prepared Samples (9 discrete sub-samples)	1.75 – 1.77 $\bar{x} = 1.76$	58.3 – 60.9 $\bar{x} = 59.8$	40.1 – 42.1 $\bar{x} = 40.8$	29.9 – 30.1 $\bar{x} = 30.4$

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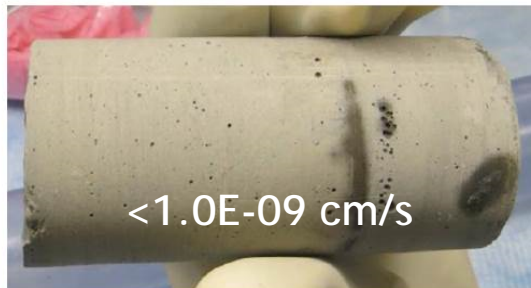
Saturated Hydraulic Conductivity (K_{sat})

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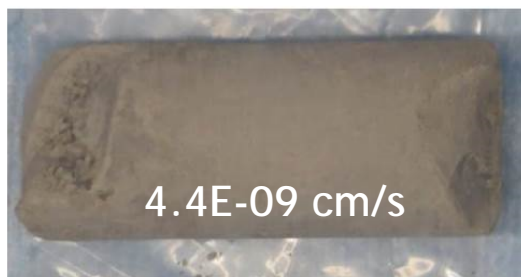
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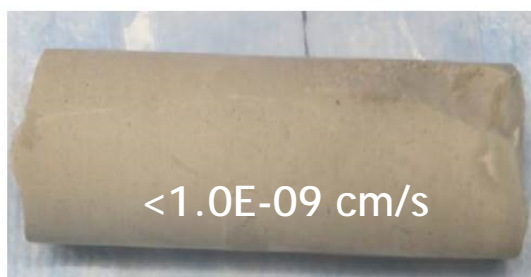
SDU2A-0931-A-2-L-2



SDU2A-0931-B-1-L-2



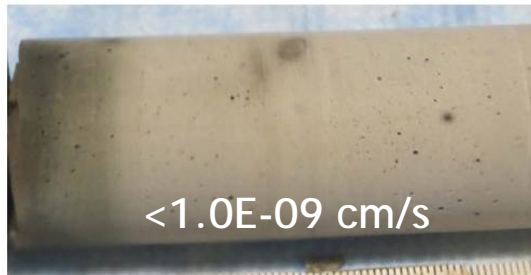
SDU2A-0931-C-1-L-2



SDU2A-0931-C-2-L-1



SDU2A-0931-C-2-L-1



- Despite higher open porosity for SDU cores impact to K_{sat} is insignificant.
- All lab-prepared samples indicated $K_{sat} < 1.0E-09$ cm/s.
- Lab = demolded samples without surface defects.
- PA-utilized $K_{sat} = 6.4E-09$ cm/s.

Radionuclide Leaching

We do the right thing.

- Tests involved mixing 1 gram of ground SDU2A core with 10 mL leachate.
- One set of leachates contained 8 ppm O₂ (oxic); other set boiled to remove dissolved O₂ (DO) (anoxic).
- Six discrete measurements for each environment.
- Samples equilibrated for 7 days and leachates analyzed for ⁹⁹Tc, ⁹⁰Sr, ¹²⁹I, E_h and pH .
- PA transport model:
 - ⁹⁹Tc assumed to be in reduced, sparingly soluble Tc(IV) form in anoxic, high pH environment ⇒ leaching behavior solubility controlled.
 - In oxic environment Tc(VI) assumed to be transformed to highly soluble Tc(VII) (pertechnetate) ⇒ leaching behavior controlled by adsorption to saltstone particle surfaces (liquid-solid partitioning K_d).
 - ⁹⁰Sr and ¹²⁹I leaching controlled by adsorption in both oxic and anoxic environments (liquid-solid partitioning K_d).

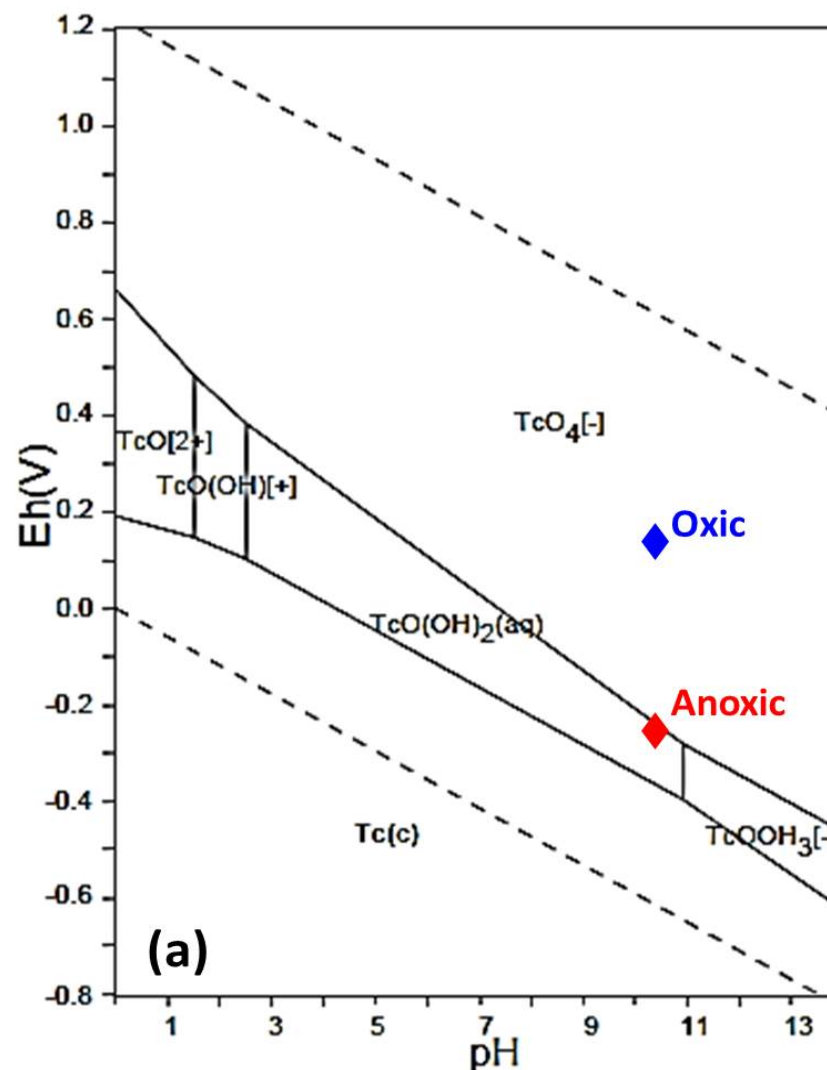
^{99}Tc Solubility (Anoxic)

Predicted Tc Species

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Anoxic	
pH	E_h (mV)
10.4 to 10.6 $\bar{x} = 10.5$	-186 to -240 $\bar{x} = -217$

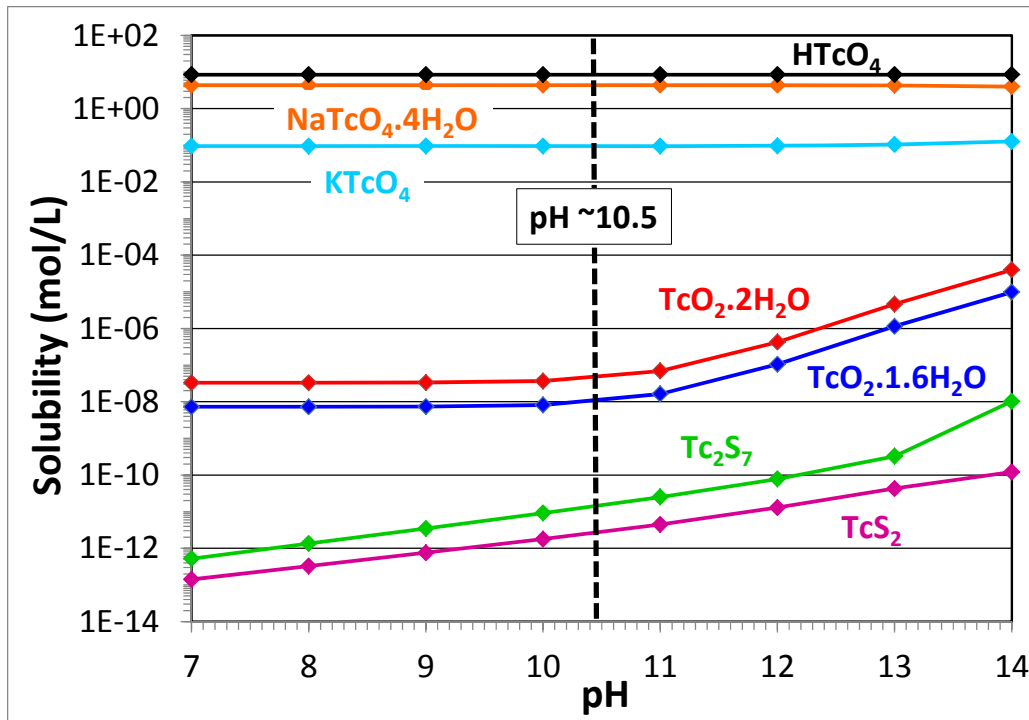
Oxic	
pH	E_h (mV)
10.6 to 10.8 $\bar{x} = 10.7$	+144 to +178 $\bar{x} = +155$



⁹⁹Tc Solubility (Anoxic)

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Sample ID	⁹⁹ Tc Conc. (mol/L)
SDU2A Cores(6 samples)	7.0E-09 to 3.9E-08 $\bar{x} = 2.2E-08$
SDF PA Model	1.0E-08



- ⁹⁹Tc concentrations in leachate (anoxic) ≈ 2E-08 mol/L.
- Agrees with predicted solubility of reduced TcO₂·xH₂O species at measured pH.
- Suggests leaching behavior is controlled by reduced Tc-species solubility as modeled in PA.
- Measured ⁹⁹Tc solubility 2x that utilized for PA model – impact?.

Liquid-Solid Partitioning (K_d)

Desorption (Reverse) K_d

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Sample ID	Reverse K_d (mL/g)				
	⁹⁹ Tc (Oxic)	⁹⁰ Sr (Anoxic)	⁹⁰ Sr (Oxic)	¹²⁹ I (Anoxic)	¹²⁹ I (Oxic)
SDU2A Samples	25 to 32 $\bar{x} = 28$	36 to 70 $\bar{x} = 55$	79 to 176 $\bar{x} = 112$	-5 to +4 $\bar{x} = 0.3$	-5 to +2 $\bar{x} = -2$
SDF PA Model Input	0.5	15	15	9	15

- K_d for ⁹⁹Tc and ⁹⁰Sr > assumed PA value.
- For ¹²⁹I lowest feasible K_d is zero.
- Unfeasible negative K_d for ¹²⁹I may be partially associated with low ¹²⁹I concentration (≈ 5 pCi/g) and analytical uncertainty of radiochemical analyses.

PA Transport Model Impacts

Collective Impacts

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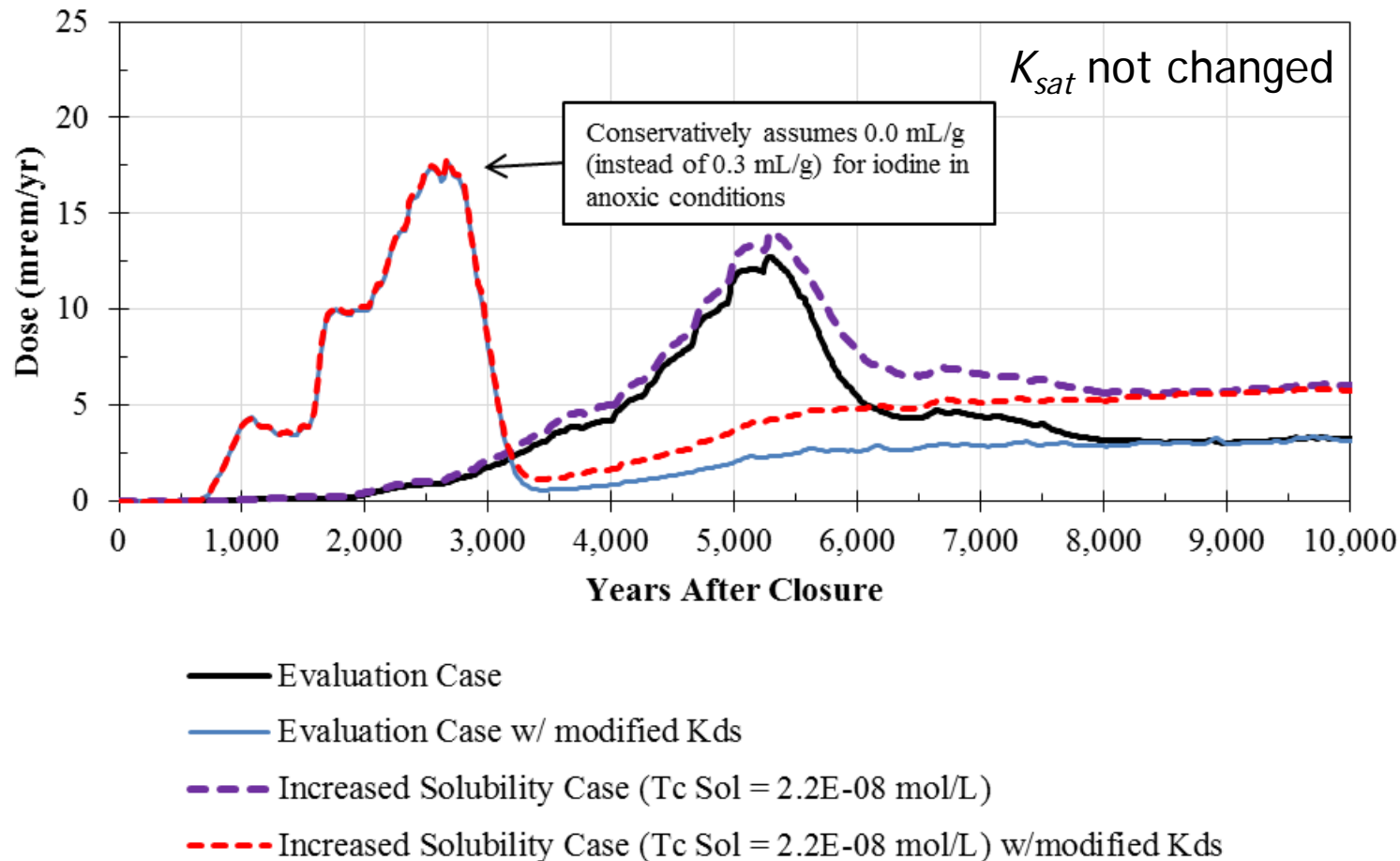
- **With the exception of the modified I K_d s, all of the measured K_d values improve the expected results (or have negligible impacts)**
 - The adverse impacts of the modified I K_d s are expected to be partially mitigated by the lower K_{sat} .
 - Due to the analytical approach used to estimate the I K_d s, there is some uncertainty (i.e., the negative values from the reverse K_d analysis are not realistic)
 - Assuming a value of 0 mL/g is intended to bound this uncertainty

Note: A lower cementitious degradation rate for saltstone within the first 3,000 years after closure would significantly limit the release of ¹²⁹I.

Collective Impacts

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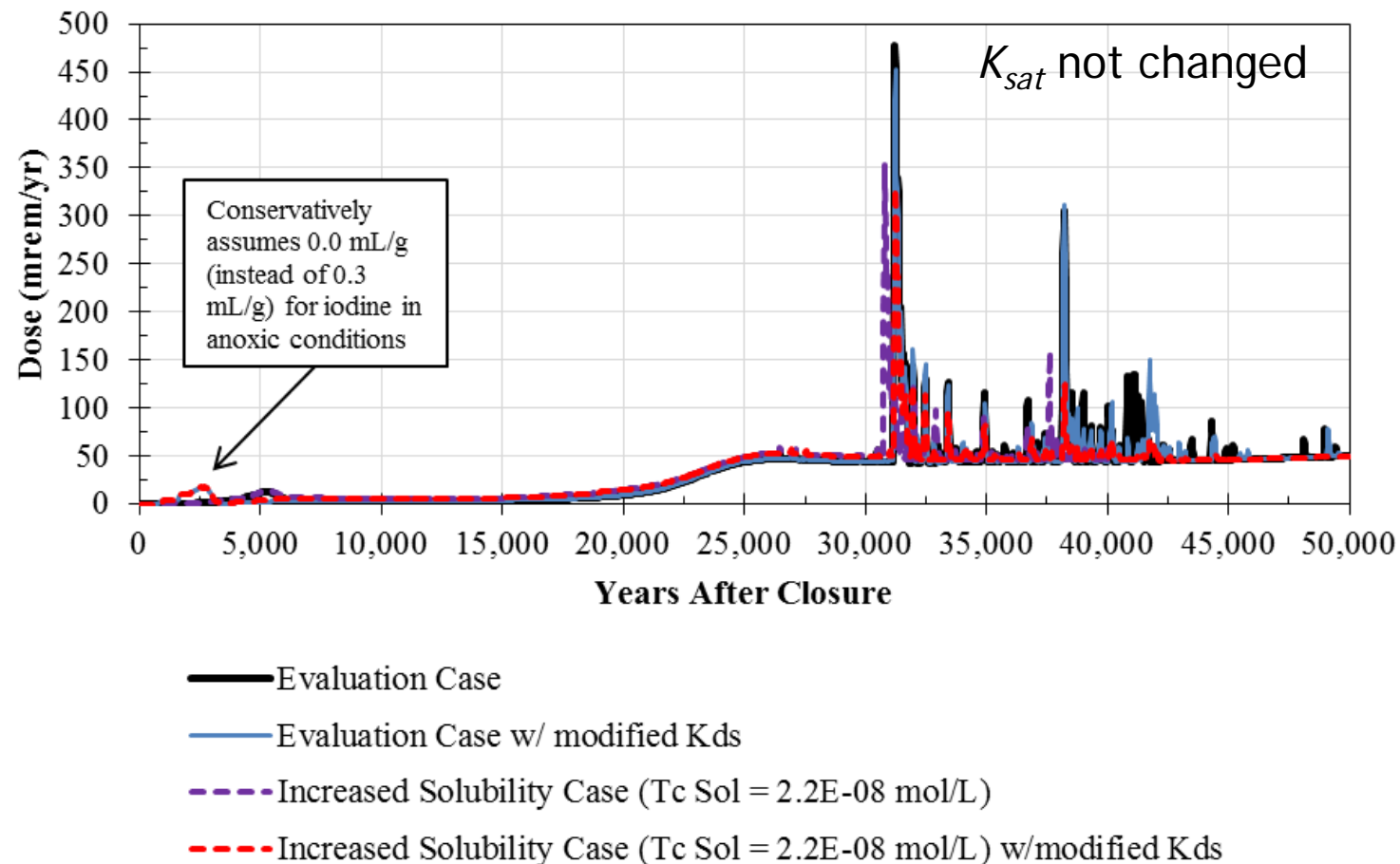
- Incorporating the results of the SDU 2A Core Sample Analysis gives a higher and earlier ^{129}I dose peak.



Collective Impacts

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- Incorporating the results of the SDU 2A Core Sample Analysis gives a lower ^{99}Tc dose peak.



SDU2A Core Analysis Summary

We do the right thing.

- Differences in SDU2A core sample densities and porosities reflect anticipated variability associated with field processing. Variations insignificant with respect to key saltstone properties.
- Higher K_{sat} which indicated equivalent performance to lab-processed samples.
- Under anoxic conditions the leaching behavior of ^{99}Tc is controlled by the solubility of reduced $\text{TcO}_2 \cdot x\text{H}_2\text{O}$ phases as modeled in the SDF PA transport model.
- Measured ^{99}Tc solubility of $2.2\text{E}-08$ mol/L compared to $1.0\text{E}-08$ mol/L assumed in PA transport model.
- ^{90}Sr and ^{99}Tc K_d higher than modeled in SDF PA transport model.
- ^{129}I $K_d < 1$ ml/g and potentially zero; lower than currently assumed in PA transport model.
- PA impacts \Rightarrow earlier and higher ^{129}I peak dose
 \Rightarrow lower ^{99}Tc peak dose.

Acknowledgements

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