



# Developments into Understanding Spent Nuclear Fuel Canister Surface Environment

*International High-Level Radioactive Waste Management Conference, Phoenix, AZ*

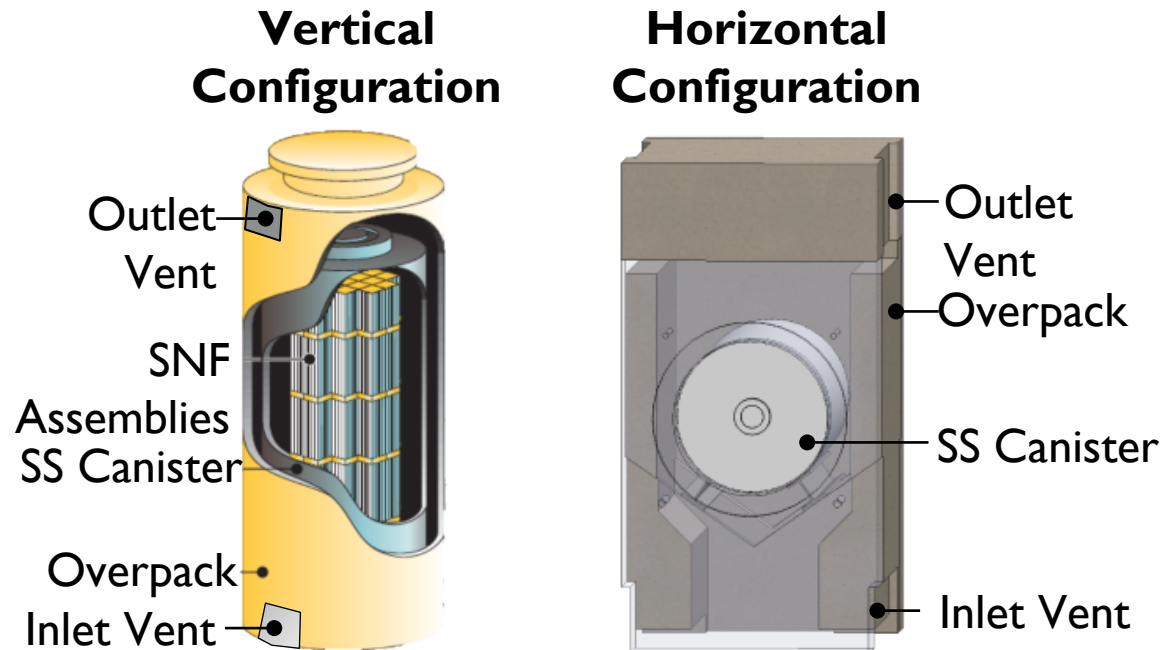
November 15<sup>th</sup>, 2022 | Andrew Knight

Ryan Katona, Makeila Maguire, Brendan Nation, Rebecca Schaller, and Charles Bryan

Sandia National Laboratories

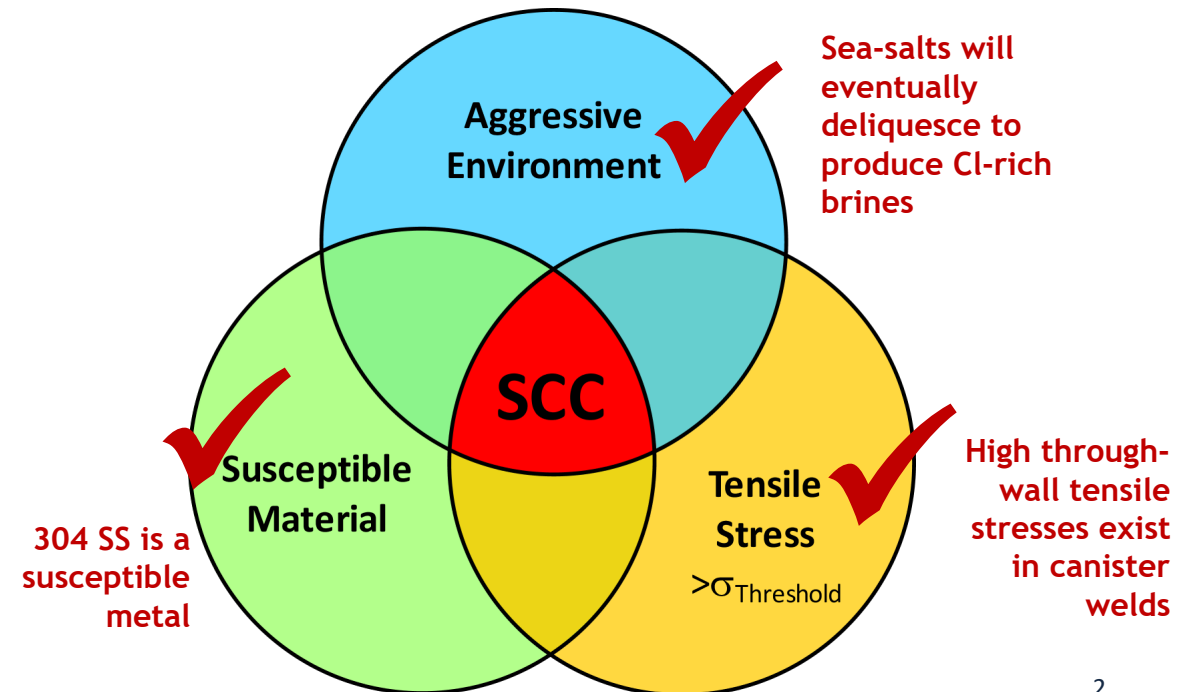
## Vertical and Horizontal Systems

- Welded stainless steel canister shielded with concrete or concrete/steel enclosure. Passively cooled by advective air flow
  - Ambient air contains dust that will accumulate on canister
  - Dust contains salts that will eventually deliquesce in humid air

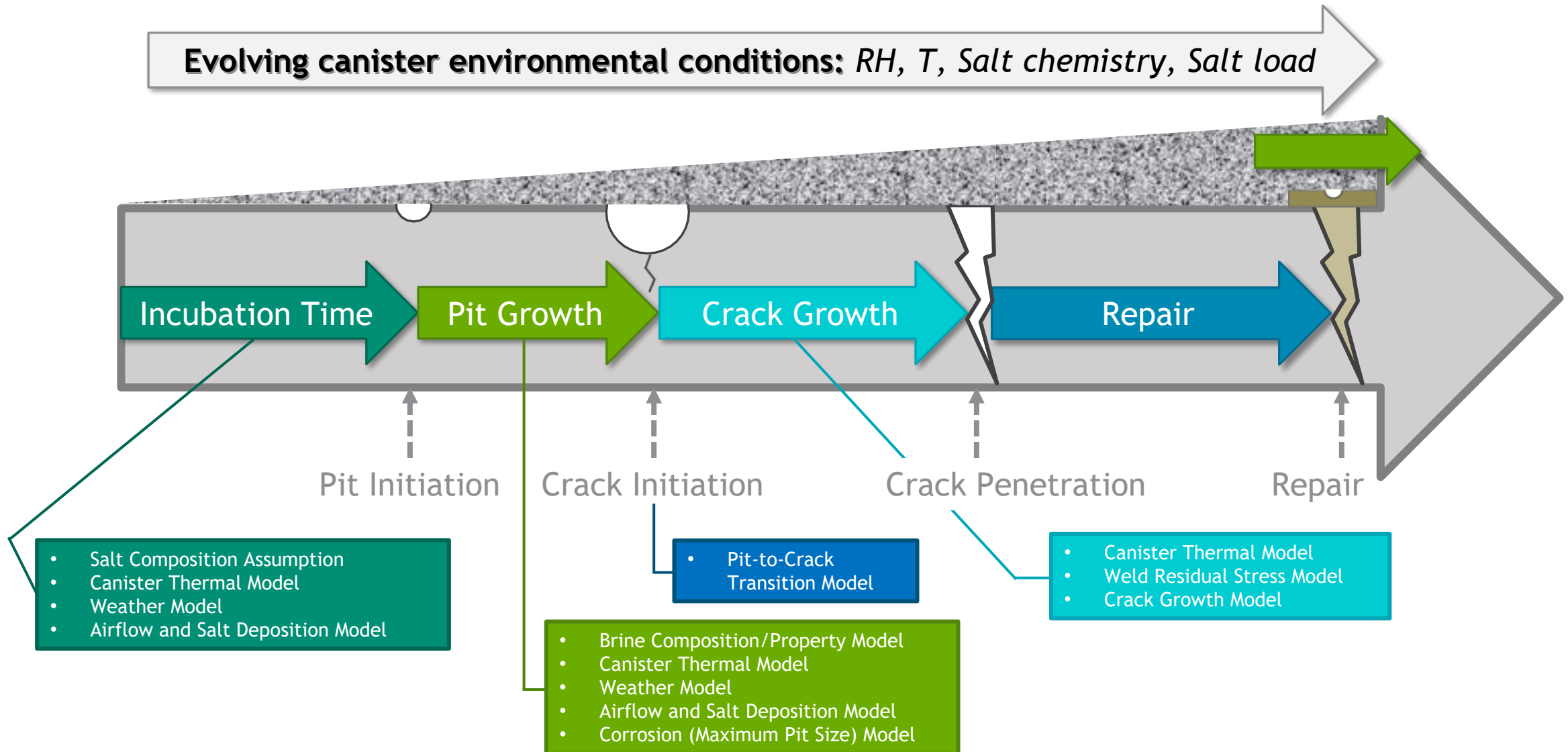


## Resulting concentrated brines may cause localized corrosion on the canister

- With sufficient stress, corrosion may evolve into stress corrosion cracks (SCCs)
  - Canister welds produce high residual stresses
  - SCCs initiating in weld regions could penetrate through the canister wall



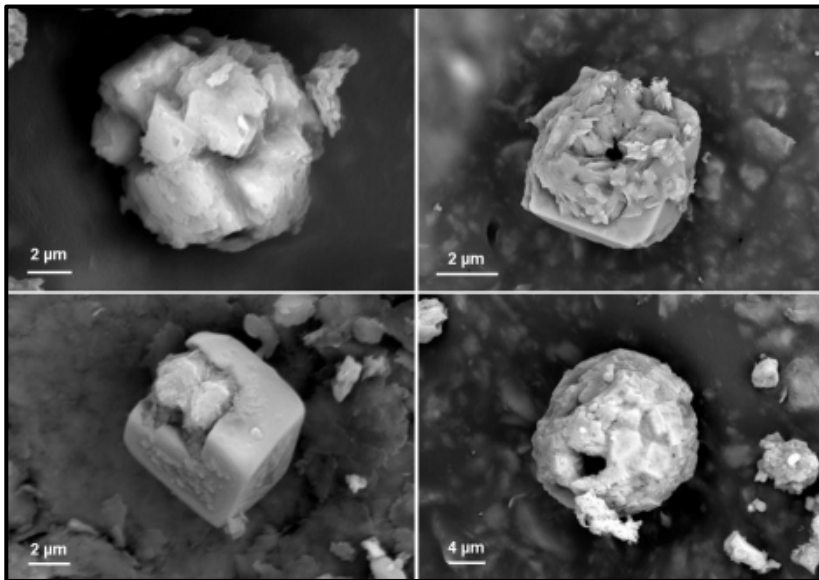
# Canister Surface Environment Underpins SNL SCC Model



**We aim to improve our understanding of the canister surface environment to reduce uncertainties in the SCC model by reducing conservatisms in assumptions**

## Dust/Salt Composition

Evaluate how different salt chemistries at IFISIs impact brine properties and corrosion susceptibility



Sea-salt aggregates from the surface of a Diablo Canyon storage canister (Bryan, C. 2014)

## Presence of Inert Dust

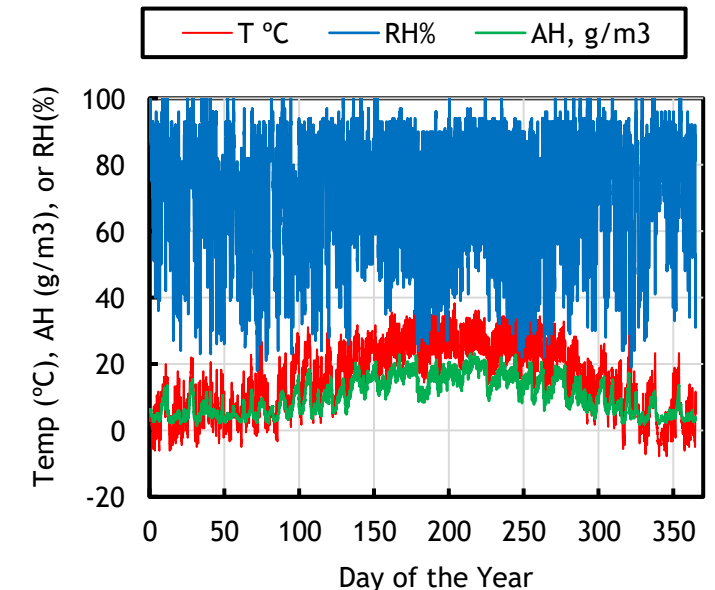
Determine how the presence of inert dust (e.g. minerals, organic materials) impact brine behavior and corrosion



Dust on a storage canister surface, Calvert Cliffs ISFSI (EPRI 2014)

## Diurnal Cycling

Employ diurnal or seasonal T and RH changes in testing to assess its impact to develop corrosive environment



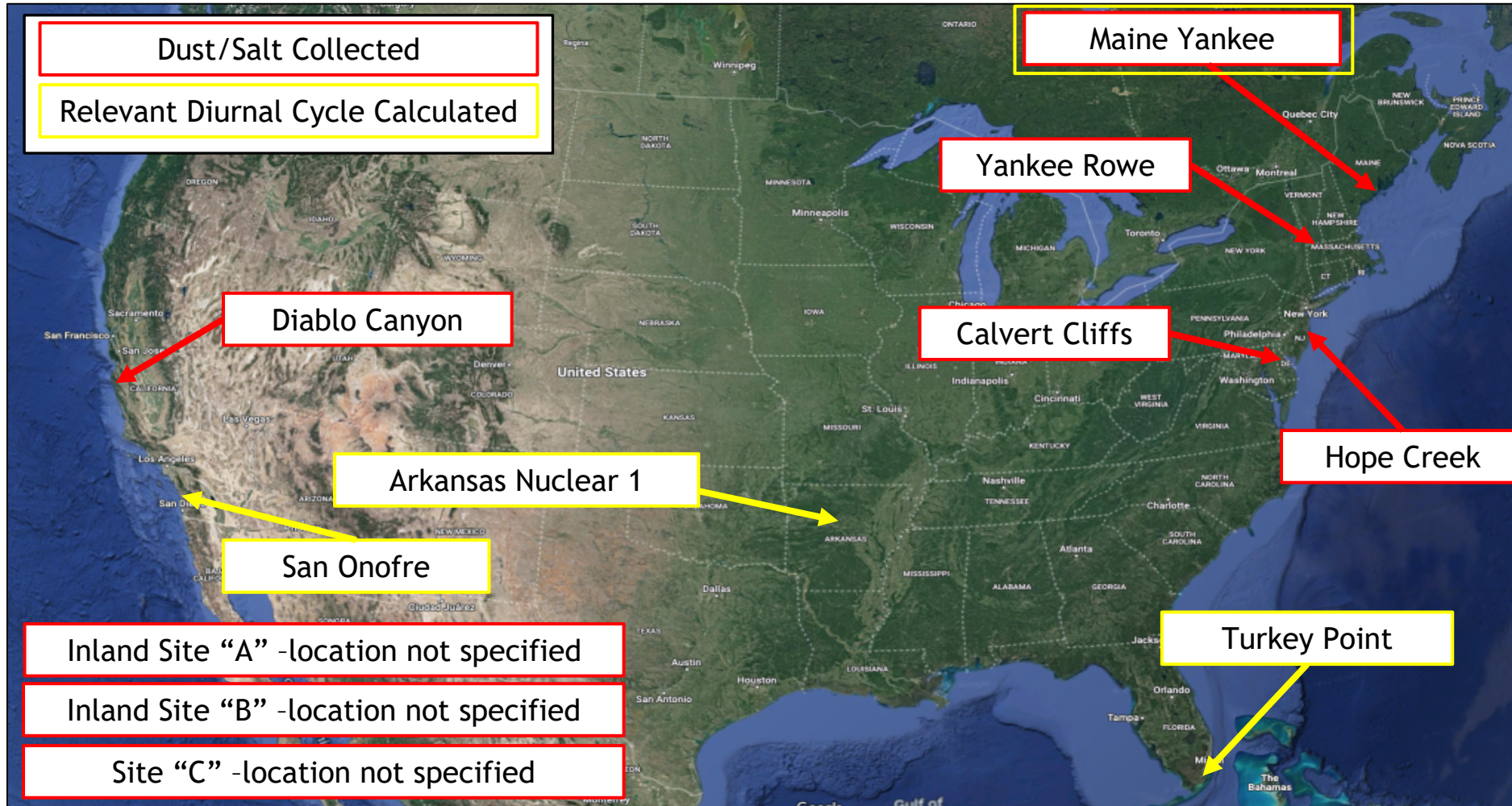
Weather Data from Arkansas Nuclear 1 (Schaller et al. (2021))



# ISFSI Site Sampling 2014 to Date



Dust samples collected from 8 ISFSIs, and relevant diurnal cycles from weather data were calculated for 5 ISFSIs. Site-specific data are used to better understand factors that influence surface environment



## The canister configuration, sampled area, and sampling methods varied from site-to-site.

- Sampling efficiency has not been validated for any method

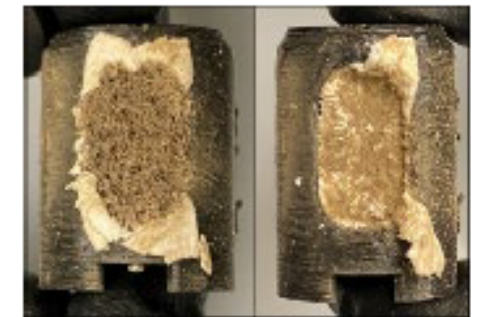
ISFSI Site	Year	Canister Type	#	Sampling Methods	Sampling Locations	Approx. Age of Canister when sampled
Calvert Cliffs <sup>1,2</sup>	2013, 2015	Horizontal	2	Scotch-Brite® + paper filter backing with vacuum-operated hand probe	Middle and 10 o'clock position	19 years
Hope Creek <sup>3,4</sup>	2014	Vertical	2	Scotch-Brite® and Salt-smart® pads	Top and side of vertical canister	10 years
Diablo Canyon <sup>3,5</sup>	2014	Vertical	2	Scotch-Brite® and Salt-smart® pads	Top and side of vertical canister	2-4 years
Maine Yankee <sup>2,6,7</sup>	2016, 2018, 2019	Vertical	4-6	Scotch-Brite® with robot, wet sponges, and witness coupons	Top of vertical Canister, inlet/outlet vents, shield plug <sup>b</sup>	10 - 20 years
Inland Site A <sup>8</sup>	2020	Horizontal	1	Scotch-Brite® + quartz filter backing with vacuum robot	2, 4, 10, and 12 o'clock, HSM rails	20 years
Inland Site B <sup>9</sup>	2020	Horizontal	1	Scotch-Brite® + quartz filterbacking with vacuum robot	2, 4, 8, 10, and 12 o'clock, welds, and HSM rails	20 years
Site C <sup>10</sup>	2022	Horizontal	1	Scotch-Brite® + paper filter backing with vacuum-operated hand probe	2, 10, 11:30 o'clock at both 24 and 39 inch transects	29 years
Yankee Rowe <sup>11</sup>	2022	Vertical	1	Hand sampled with large Whatman filter	North, South, East, and West Outlet Vents	---
Arkansas Nuclear <sup>17</sup>				No Dust Collected		
Turkey Point <sup>7</sup>				No Dust Collected		
San Onofre <sup>7</sup>				No Dust Collected		



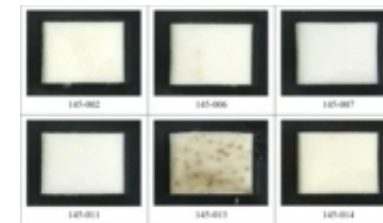
Vacuum robot used for sampling at Inland Site A and Inland Site B



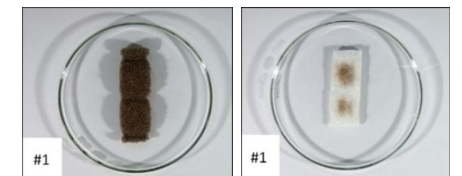
Sample from Yankee Rowe



Sample from Inland Site B



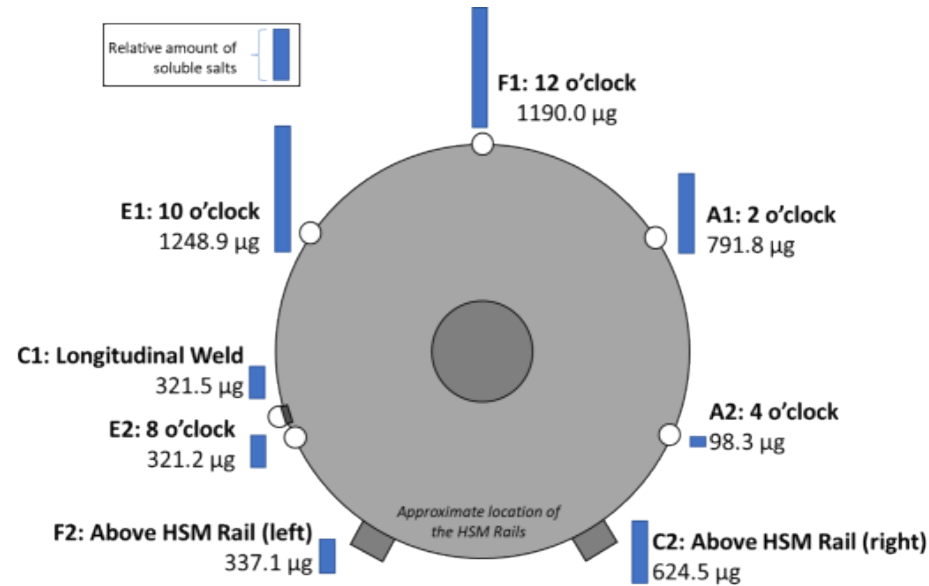
Salt-smart samples from Hope Creek



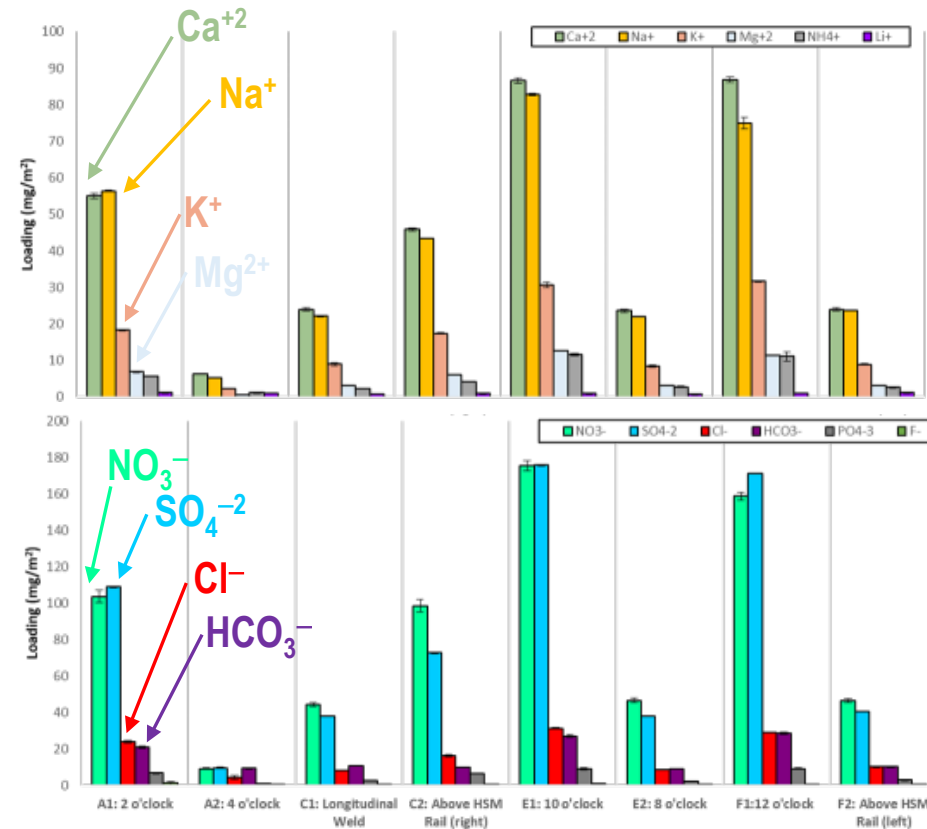
Sample from Calvert Cliffs<sup>6</sup>

<sup>1</sup>EPRI (2014); <sup>2</sup>Bryan, C. and Schindelholz, E (2017); <sup>3</sup>Bryan, C. and Enos, D. (2014); <sup>4</sup>Bryan, C. and Enos, D. (2015); <sup>5</sup>EPRI (2016); <sup>6</sup>Schaller, R. et al. (2019); <sup>7</sup>Schaller et al. (2020); <sup>8</sup>Bryan, C. and Knight, A. (2020); <sup>9</sup>Knight, A. and Bryan, C. (2020); <sup>10</sup>Bryan, C. et al. (2022); <sup>11</sup>Schaller et al. (2022)

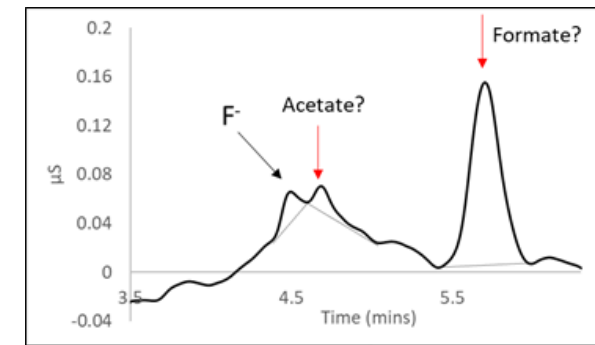
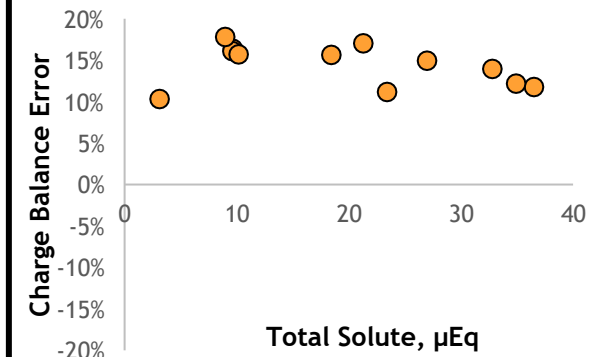
Soluble salts were leached and analyzed by ion chromatograph and dissolved inorganic carbon analyzer to determine the salt composition and loading



Total Soluble Salts as a function of canister location at Site B



Cation and anion composition in each sample collected from Site B



Charge balance error and unidentified peaks in the IC



# Determining Canister Relevant Brine Composition for Testing

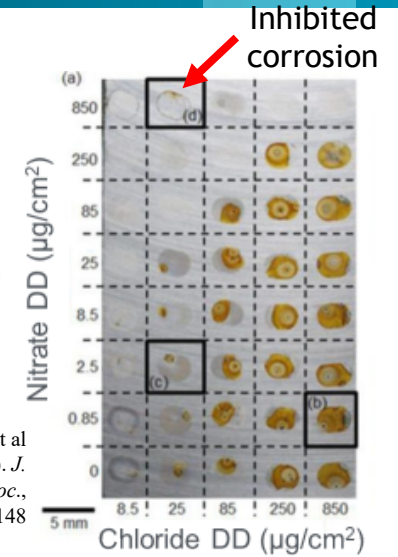


## Site specific deliquescence behavior of observed brine compositions

- Salt composition collected from 6 ISFSI sites
  - 3 east coast near-marine sites (Calvert Cliffs, Maine Yankee, and Hope Creek) – mix of sea salts and continental salts
  - 1 west coast near-marine site (Diablo Canyon) – primarily sea salts
  - 2 inland sites (“Site A” and “Site B”) – primarily continental salts

## Deliquescence behavior and salt chemistry will impact the corrosion properties of the brine

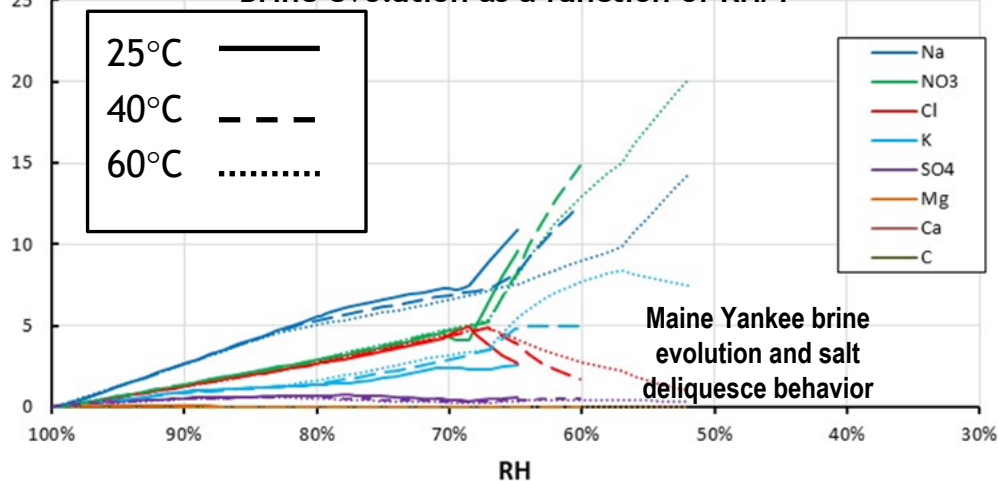
- Evidence suggests that nitrates *may* inhibit corrosion, therefore the  $\text{NO}_3^-/\text{Cl}^-$  is being evaluated (see Rebecca Schaller's paper)



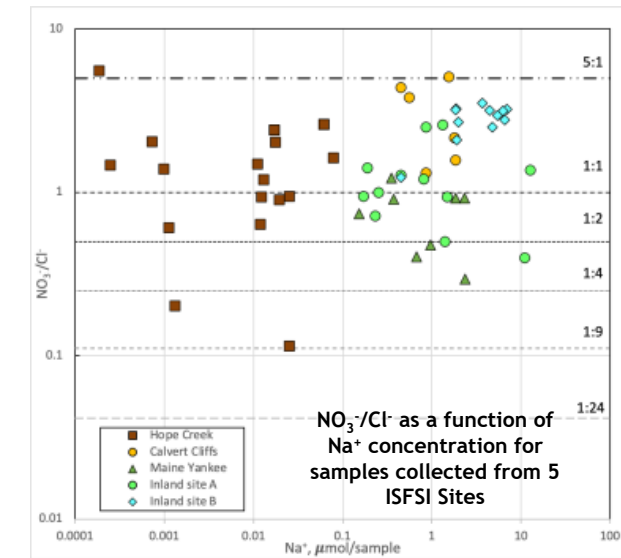
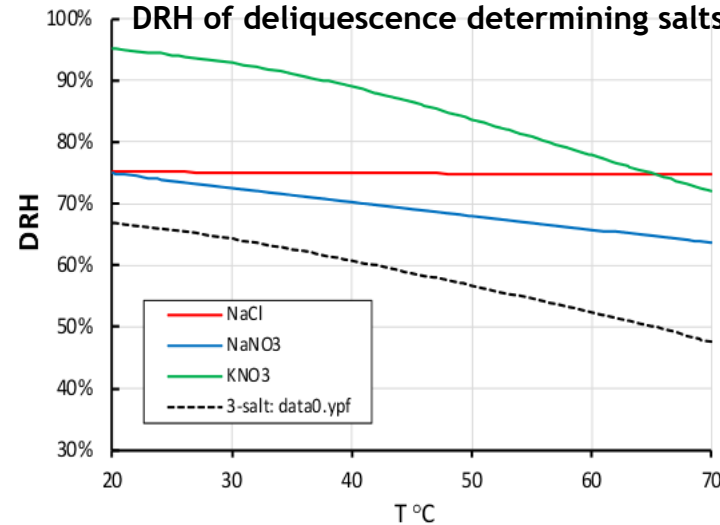
Cook, A. J. et al  
(2017). *J. Electrochem. Soc.*,  
164(4), C148

304L plate with mixed  
droplets of  $\text{MgCl}_2 + \text{Mg}(\text{NO}_3)_2$ .

Brine evolution as a function of RH/T



DRH of deliquescence determining salts

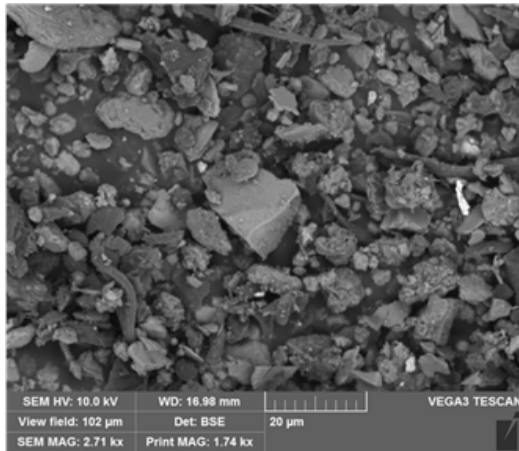




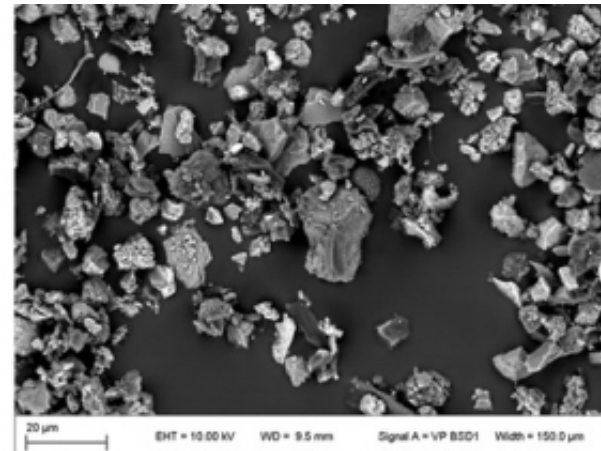
# Dust/Salt Morphology and Mineralogy Seen at ISFSIs



Evaluated particle size distributions at 6 ISFSI sites (4 coastal, 2 inland) to estimate particle size distributions via SEM image analysis

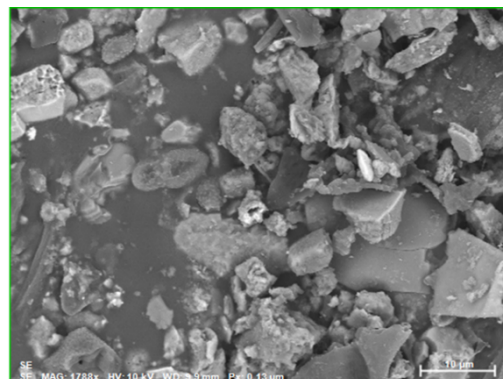


Site B



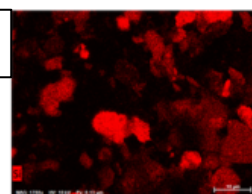
Site A

EDS analysis aided in mineralogical identification

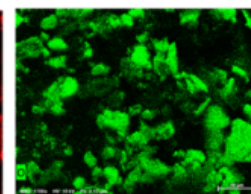


Site B

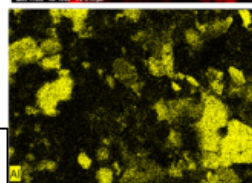
Si



O

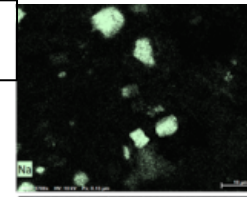


Al

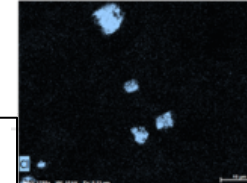


Detrital minerals consisted of aluminosilicates

Na

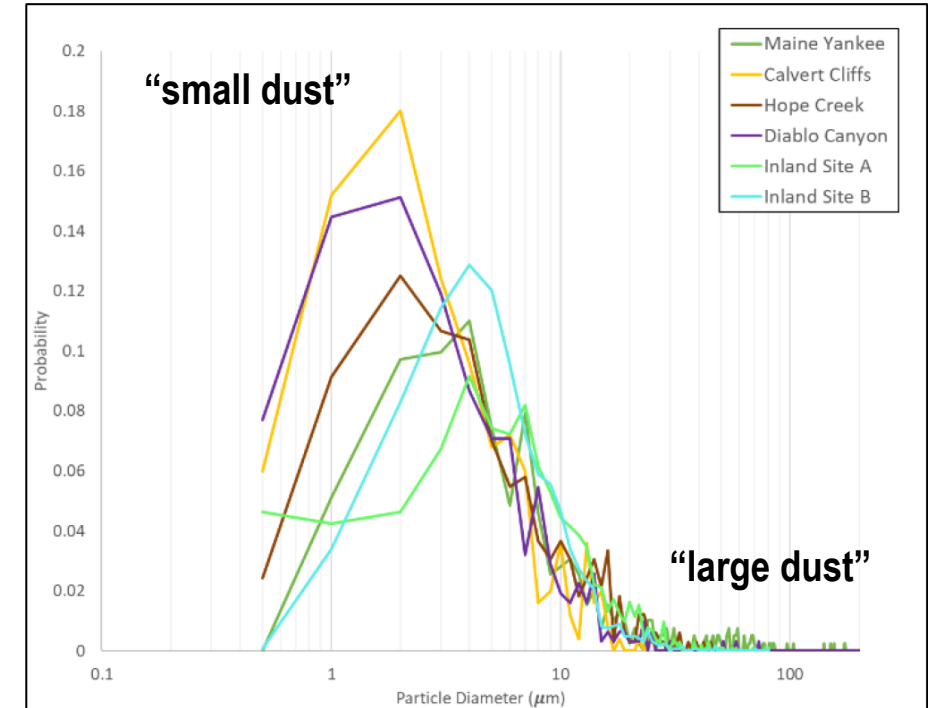


Cl



Soluble salts (NaCl)

Particle Size distribution at 6 ISFSI Sites



## What role does inert dust (e.g. minerals, organic materials) play on brine properties and corrosion?

- Capillary processes: potential formation of a continuous brine (small dust particles); potential for crevicing (large dust particles)
- Deposited 74  $\mu\text{m}$ ,  $\leq 10 \mu\text{m}$ , and mixed sized silica onto corrosion coupons for atmospheric exposure (See Rebecca Schaller's paper)

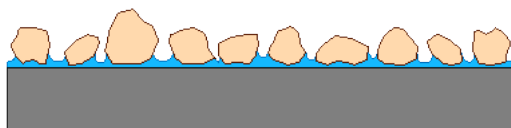


Dust deposition at the aerosol facility

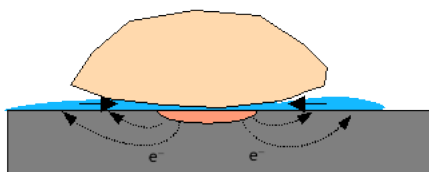
No dust—discrete deliquesced brine droplets



Small dust grains — continuous brine film



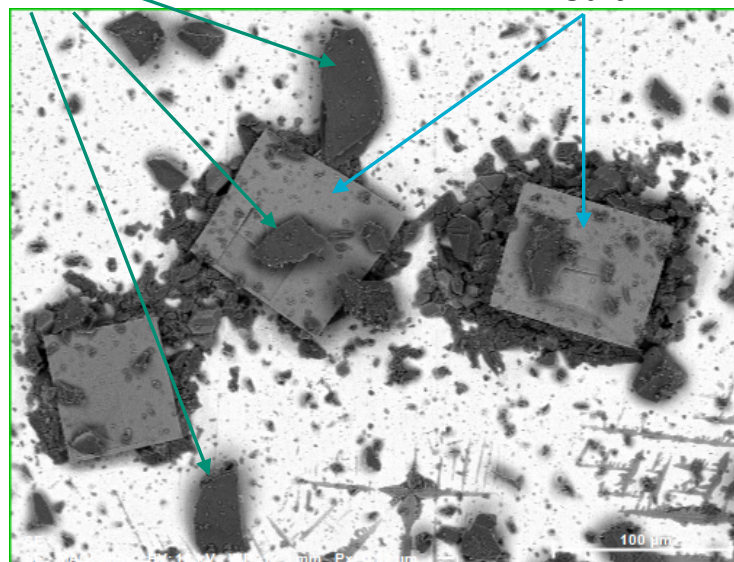
Large dust grains — crevicing?



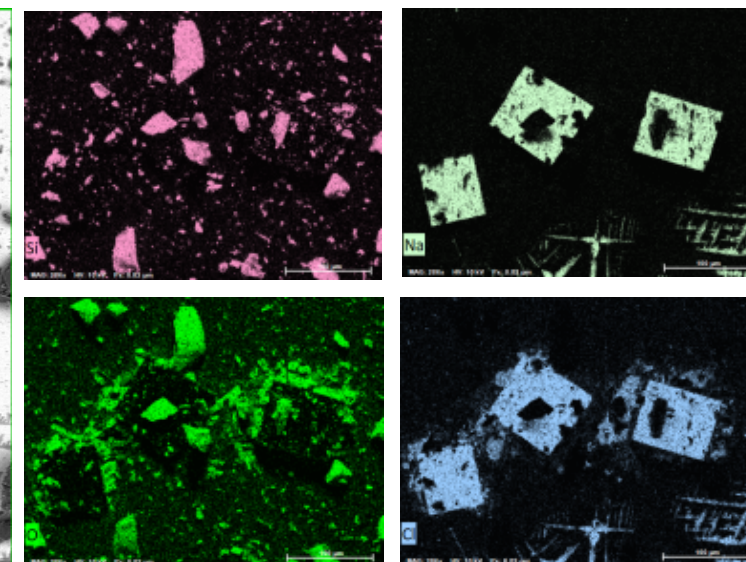
Possible influence dust can have on corrosion

Dust

Salt



Dust deposited onto ASW salt printed coupons

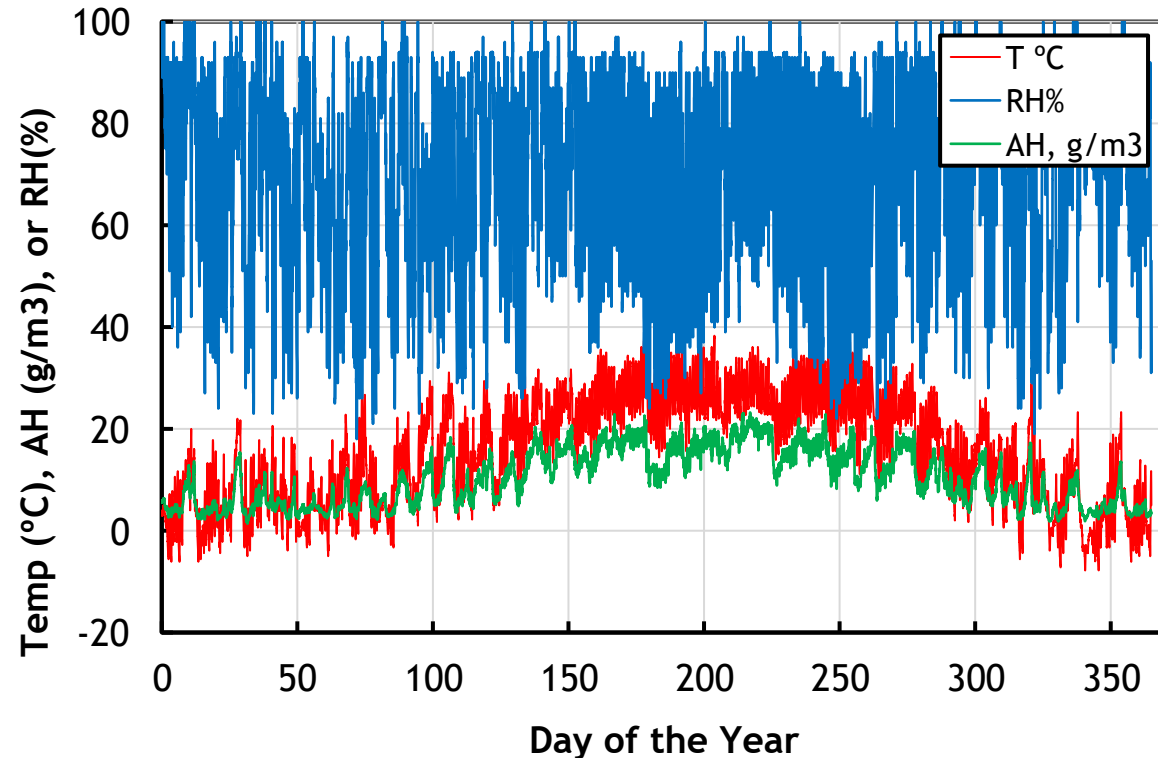




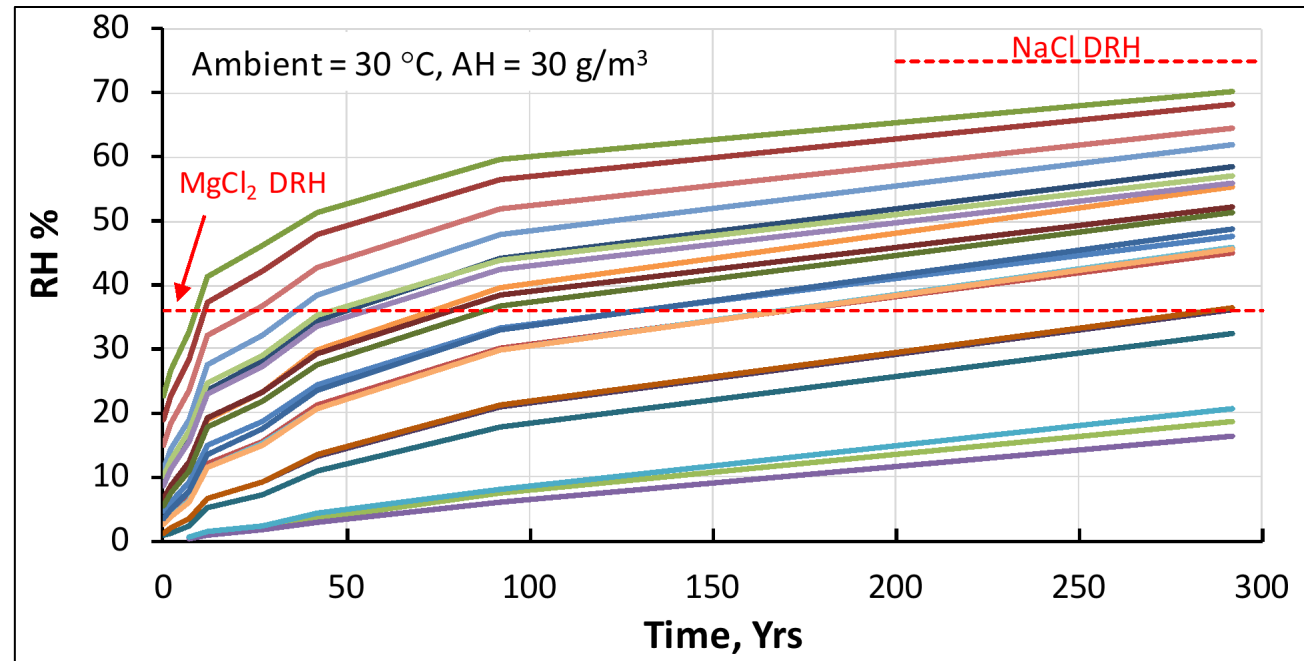
Using local weather data (T, AH, RH), realistic diurnal cycles can be simulated at various  $\Delta T$  using water equation of state<sup>1</sup> (Wagner and Pruß (2002)).

- We know that the canister surface temperature fluctuates with changes in ambient T, while maintaining a temperature delta (as a function of the canister heat load and time)
- As the canister cools, the RH can increase to a point where salts will deliquesce.

Ambient weather data from Arkansas Nuclear 1



RH as a function of time and location on the canister





# Developing a Canister Relevant Cyclic Environment



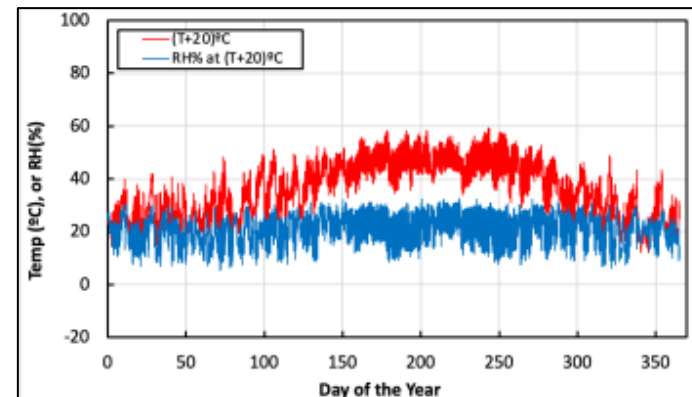
## What are relevant diurnal cycles?

- $\Delta T$  varies over time as SNF cools, so infinite number of cycles exist.
- Focus on RH ranges where large changes in brine volume/composition occur
  - $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  (bischofite) deliquescence ( $\sim 35\%$  RH) – potential brine deliquescence/dryout can occur within a few decades (worst case)
  - NaCl (halite) dissolution ( $\sim 74\%$  RH) - associated with large changes in brine volume and composition – but may not occur for  $>300$  years

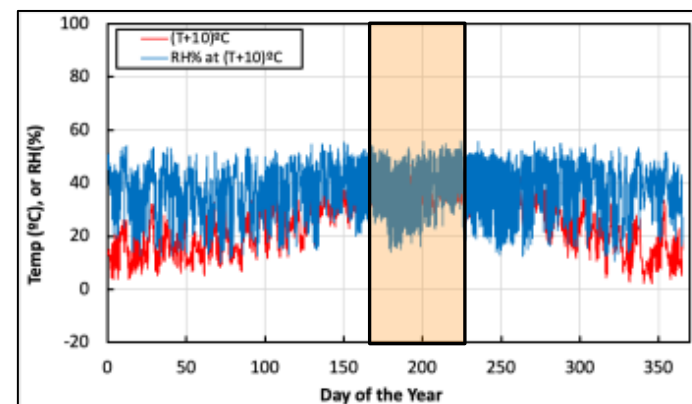
## Best fit to fluctuate about the $\text{DRH}_{\text{bischofite}}$

- Arkansas Nuclear 1 weather data with a  $\Delta T = 10^\circ\text{C}$ , 12-step daily (24 hour) cycle fit to canister surface T-RH data

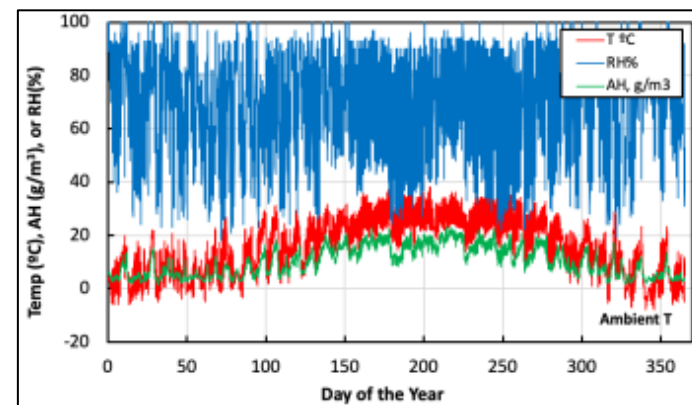
Canister surface + 20 °C above ambient



Canister surface + 10 °C above ambient



Canister surface at ambient T

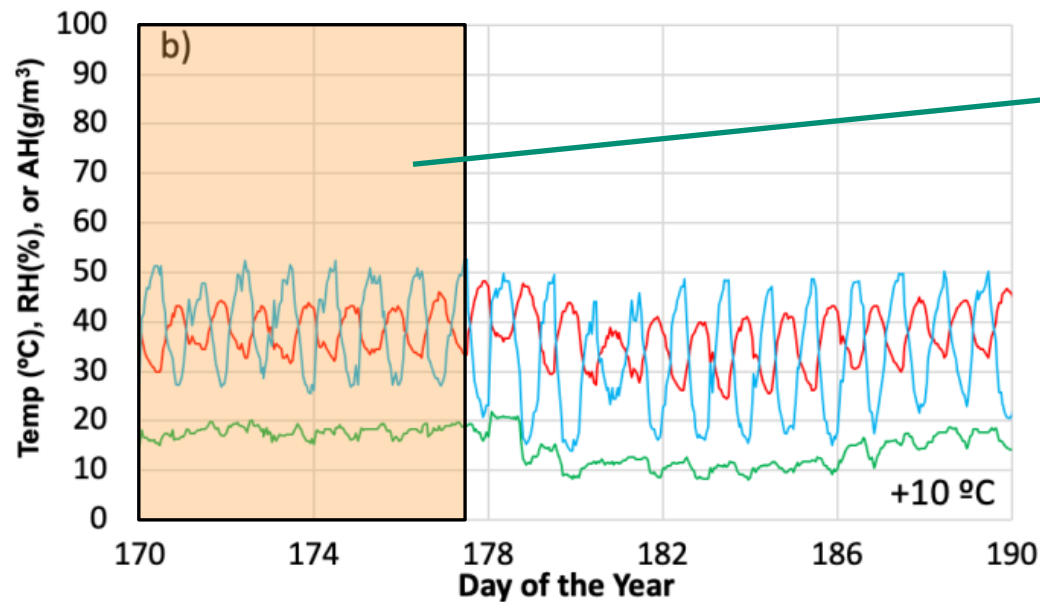


Canister surface cooling as radioactive decay occurs over time

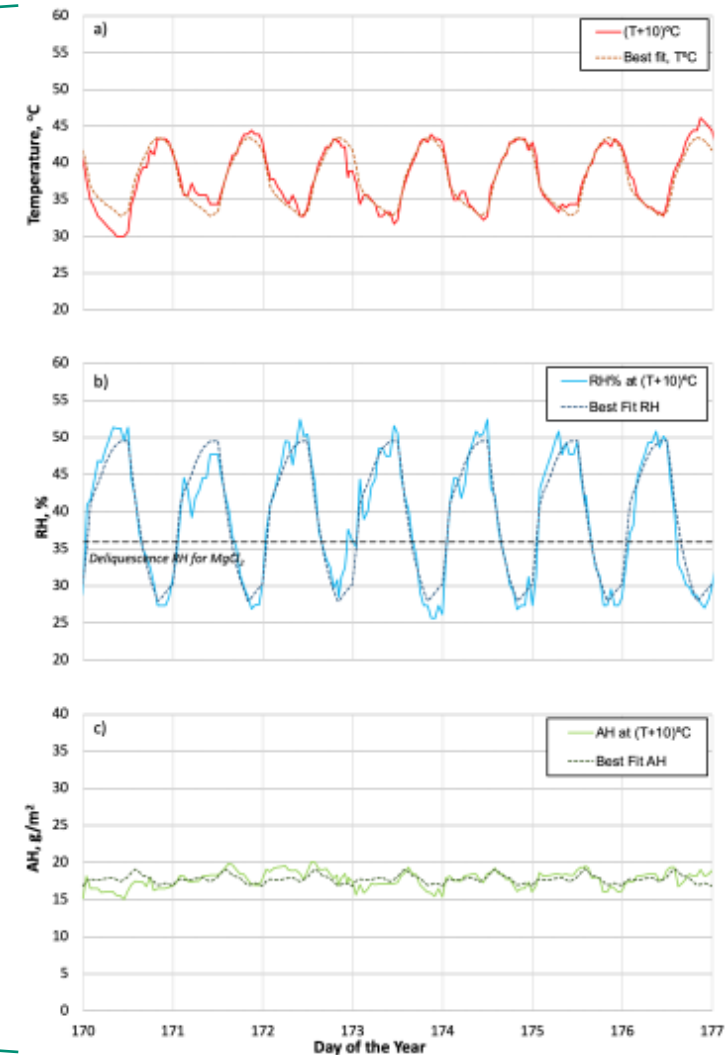
# Evaluating Cyclic Conditions



A experimental cycle modeled off of Arkansas Nuclear 1 weather data and a 10°C temperature delta is being used for a long-term corrosion experiments (See *Rebecca Schaller's paper*)



T, AH, and RH assuming a 10°C surface temperature delta



Calculated experimental cycle shown with the actual T, RH, and AH



Site specific data allows for informed testing of relevant factors that may impact localized corrosion and potential for SCC

- Brine chemistry, specifically the  $\text{NO}_3^-/\text{Cl}^-$  ratio
- Presence of inert dust of varying particle sizes
- Diurnal and seasonal cycling, specially when the cycles cross specific deliquescence RHs regularly

Corrosion testing to evaluate the impact of each of these specific conditions is underway (See Rebecca Schaller's paper "Accelerated Relevant Atmospheric Corrosion Testing of Austenitic Stainless Steel for SNF Storage Environments")

SNL plans to continue to obtain dust collected from ISFSIs across the US (2 more this FY) and will continue to explore relevant factors that may impact the risk of SCC on SNF canisters.





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We would also like to thank the collaborating ISFSI sites and those involved in the sample collection





Questions?