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The Role of Laboratory Data and Screening Criteria on the WIPP Actinide Solubility Uncertainty Analysis

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Purpose

- The purpose of the actinide uncertainty analysis is to provide performance assessment (PA) with distributions of model prediction error to assign uncertainty to the actinide baseline solubility model. The prediction error distributions are probabilistically sampled by PA, and the sampled values are used as a multiplier for the WIPP baseline solubility models.

Methodology

- Literature Searches
- Literature Screening
- Simulate Laboratory Studies
- Compare Experimental and Simulated Results
- Derive Distributions

Literature Search and Screening

- Literature Search
 - Search Plus, Version 2.4, operated by Los Alamos National Laboratory (LANL)
 - Web of Science (including Science Citation Index and INSPEC),
 - DOE Energy Citations Database.
 - 100's of potential references identified

General Screening Criteria

- G1: Studies limited to January 1990 to November 2011
- G2: Peer reviewed journals, or government sponsored laboratory data only.
- G3: Include only results from solubility studies, exclude other studies such as corrosion, sorption etc.,
- G4: Include only results where water was the solvent
- G5: Include only studies at or close to atmospheric pressure and at temperatures close to those expected at WIPP
- G6: Include only results from studies of Th(IV); and Nd(III), Am(III), and Cm(III); because these are the elements used to parameterize the WIPP Th(IV) and Am(III) solubility models.

Literature Screening (continued)

- G7: Include only studies where the solubility controlling phase, solution dissolved elements, and species thermodynamic properties are included in the WIPP QA database.
- G8: Include only studies for which the composition of the aqueous solution is known.
- G9: Include only results with dissolved elements or species for which values of μ^0/RT and Pitzer ion-interaction parameters are in our models.
- G10: Include only studies where a complete description of the experimental procedure and method are provided.
- S1: For thorium, include results for experiments carried out at ionic strengths equal to, or greater than 3M (superseded by G11)

G11 Criteria

- Purpose: The G11 criterion was added to the CRA-2014 PA, Revision 1, to make the actinide solubility uncertainty analyses consistent with other WIPP specific models in PA. The G11 criterion include only results from experimental studies carried out under conditions at, or close to those predicted for WIPP disposal rooms.
 - Ionic Strength ≥ 3 m or M
 - $8.0 \leq \text{pCH} \leq 11.2$
 - $0.0 \leq \text{TIC} \leq 1.4 \times 10^{-2}$ M

Simulate Laboratory Experiments

- EQ3/6 version 8.0a and DATA0.FMT.R2 was used to simulate laboratory experiments.
- EQ3 was used to simulate simple equilibrium experiments.
- EQ6 was used to simulate more complex experiments, typically those that involved carbonate phase solubility controlling phases.
- pCH was the independent parameter that was either fixed, or matched in the simulation at the laboratory measured value, and the EQ3/6 calculated the dissolved concentration of the target actinide by using the same solubility controlling phase as the experiment.
- Excel macro GetEQData.xls was used to extract data from EQ3/6 output files.

Derive Cumulative Distributions

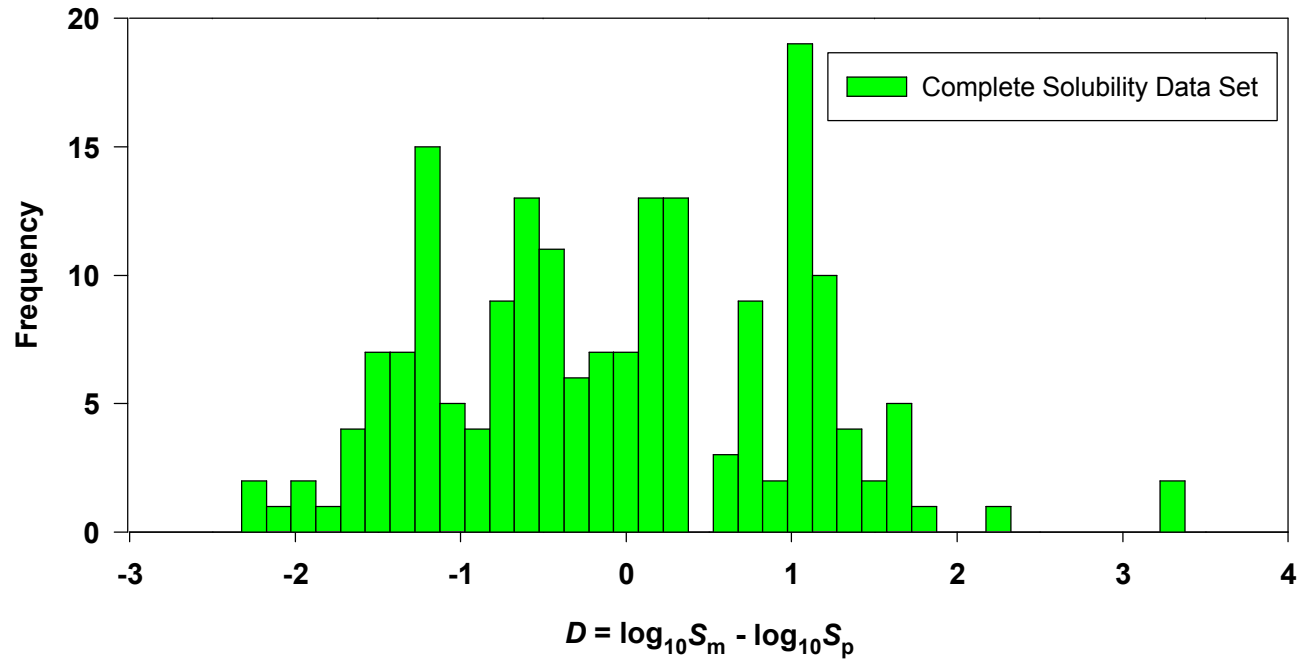
- Calculate the prediction error, i.e., the difference in the log of the measured and simulated data:

$$D = \log_{10} S_m - \log_{10} S_p$$

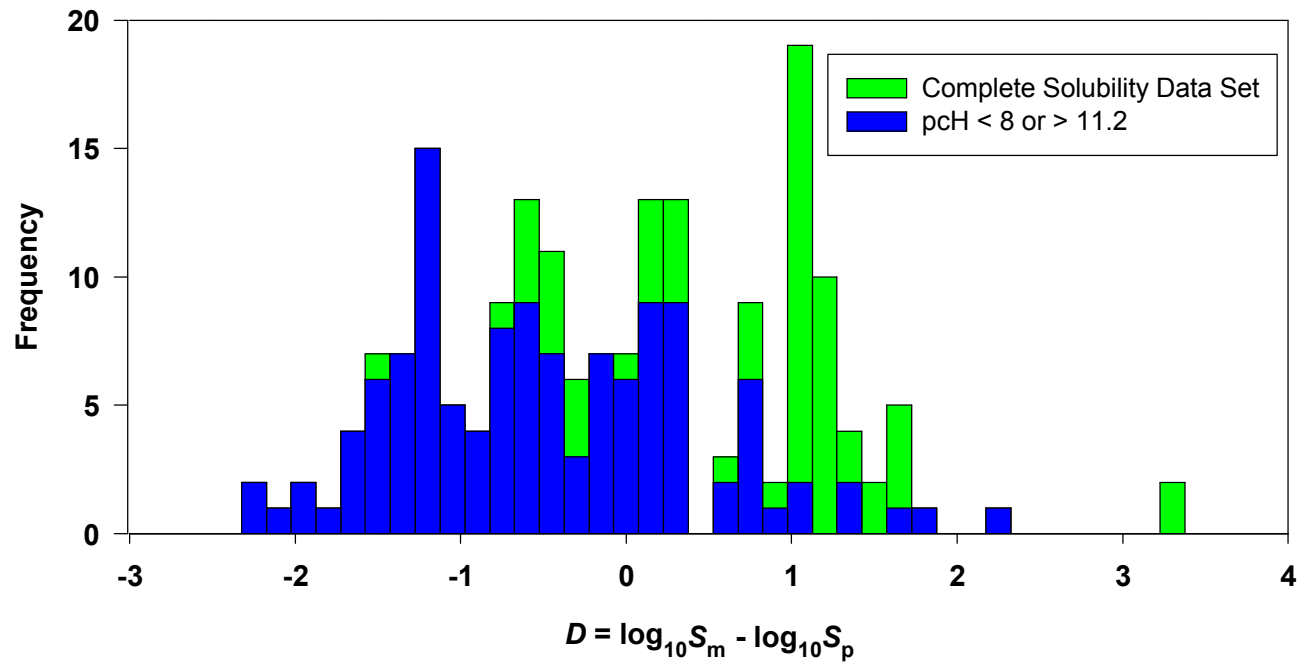
- D is the uncertainty metric
- Calculate the frequency and cumulative probability as a function of D
- Descriptive statistics
 - Mean, median, standard deviation, minimum, and maximum
- The cumulative probability and descriptive statistics are used by performance assessment to set the bounds on the baseline solubility model

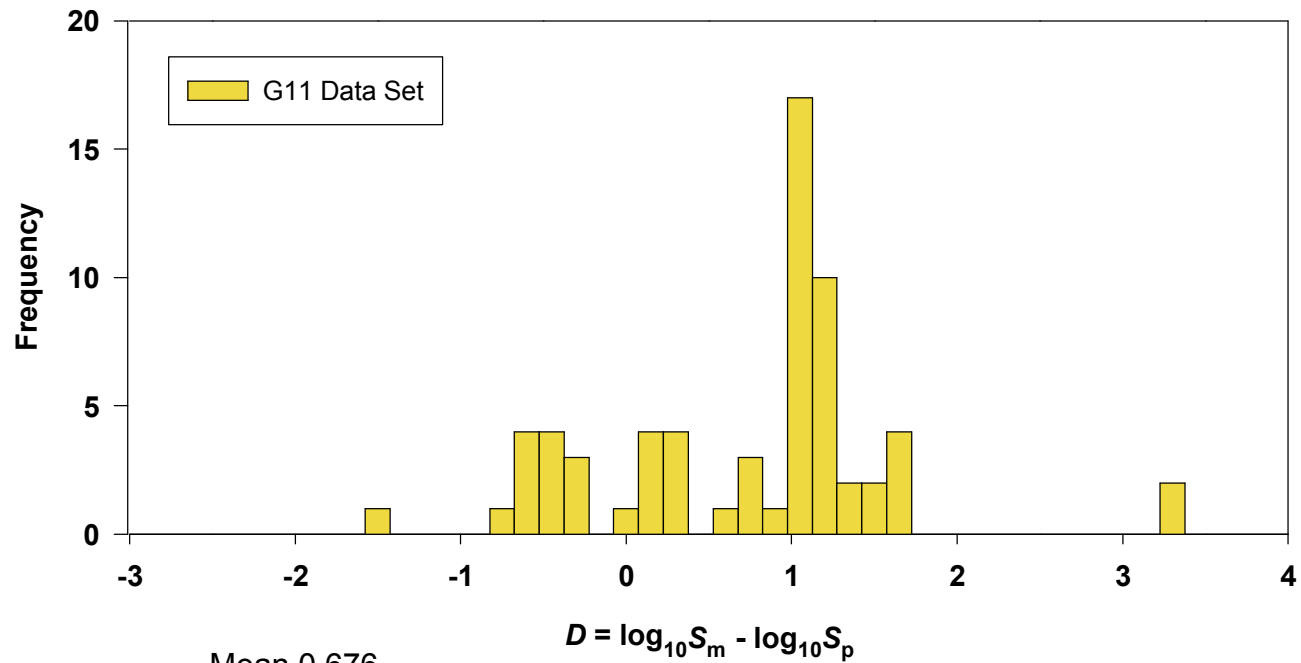
Thorium (IV) Uncertainty Analysis

	CRA-2004 PABC	CRA-2009 PABC	Complete Solubility Data Set	G11 Data Set
Maximum	2.40	3.30	3.19	3.19
Minimum	-1.80	-2.25	-2.28	-1.52
Mean	0.108	-0.346	-0.136	0.676
Median	0.075	-0.520	-0.189	1.029
Standard Dev.	0.837	0.995	1.061	0.869
<i>n</i>	45	140	185	64



Mean -0.136
Median -0.189
Std Dev 1.061
Minimum -2.28
Maximum 3.19
n = 185

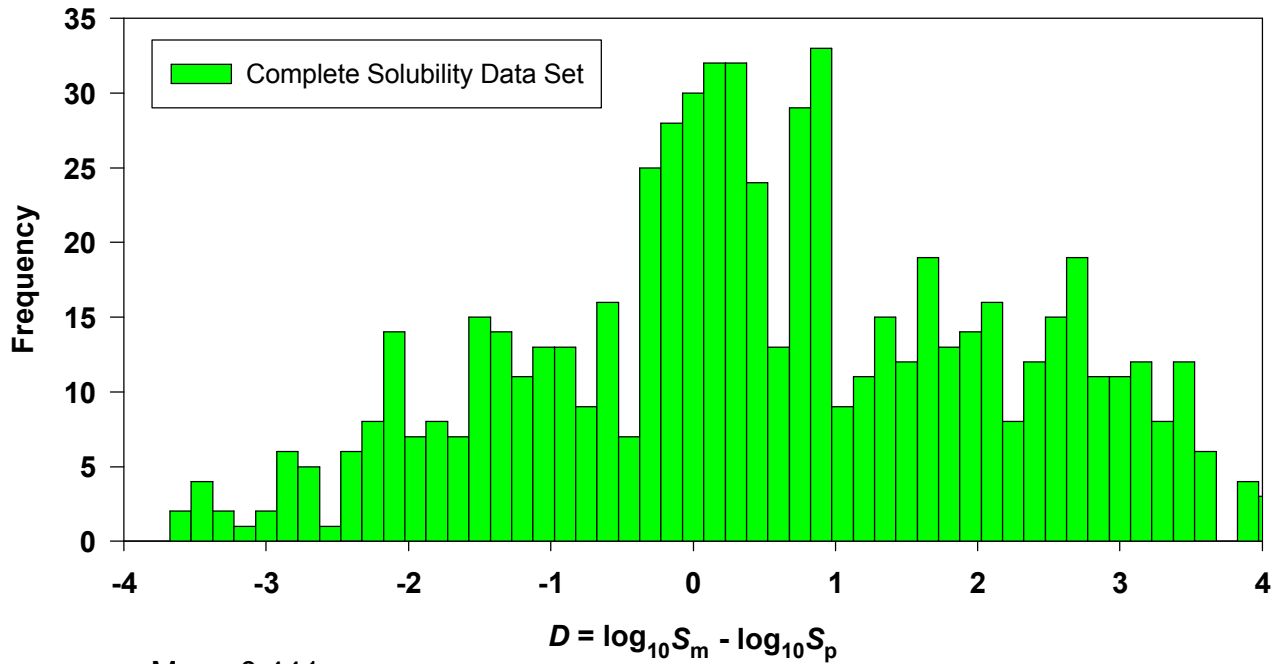




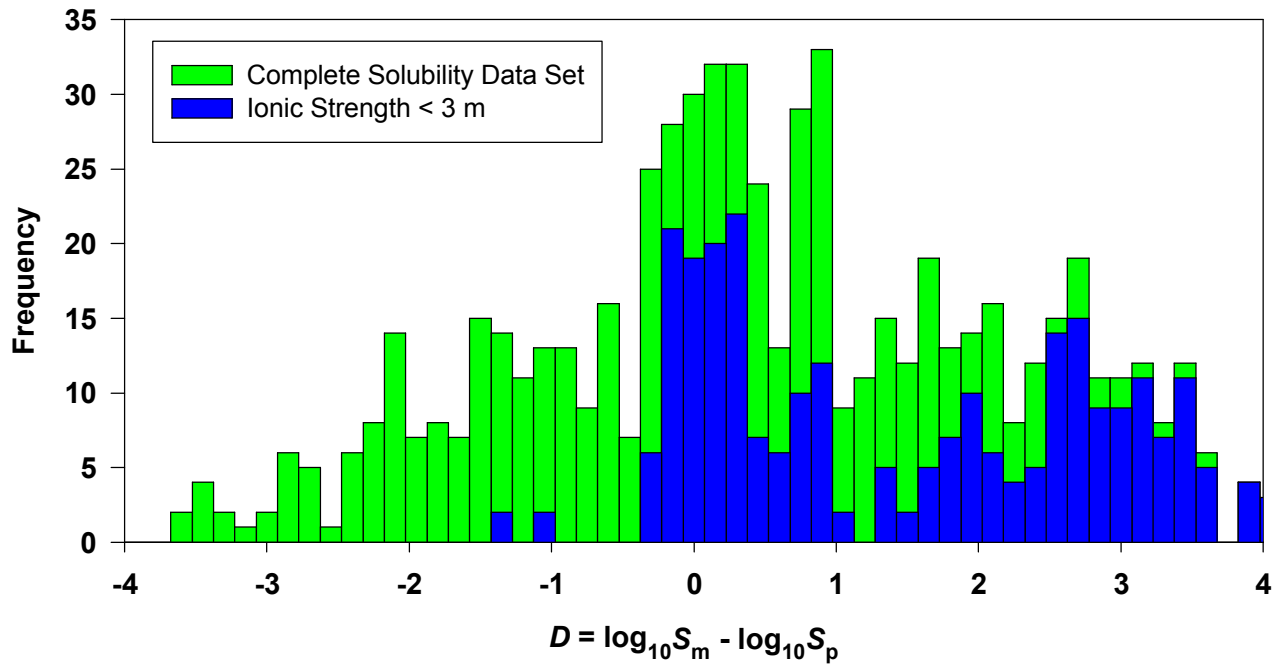
Mean 0.676
Median 1.029
Std. Dev. 0.869
Minimum -1.52
Maximum 3.19
n = 64

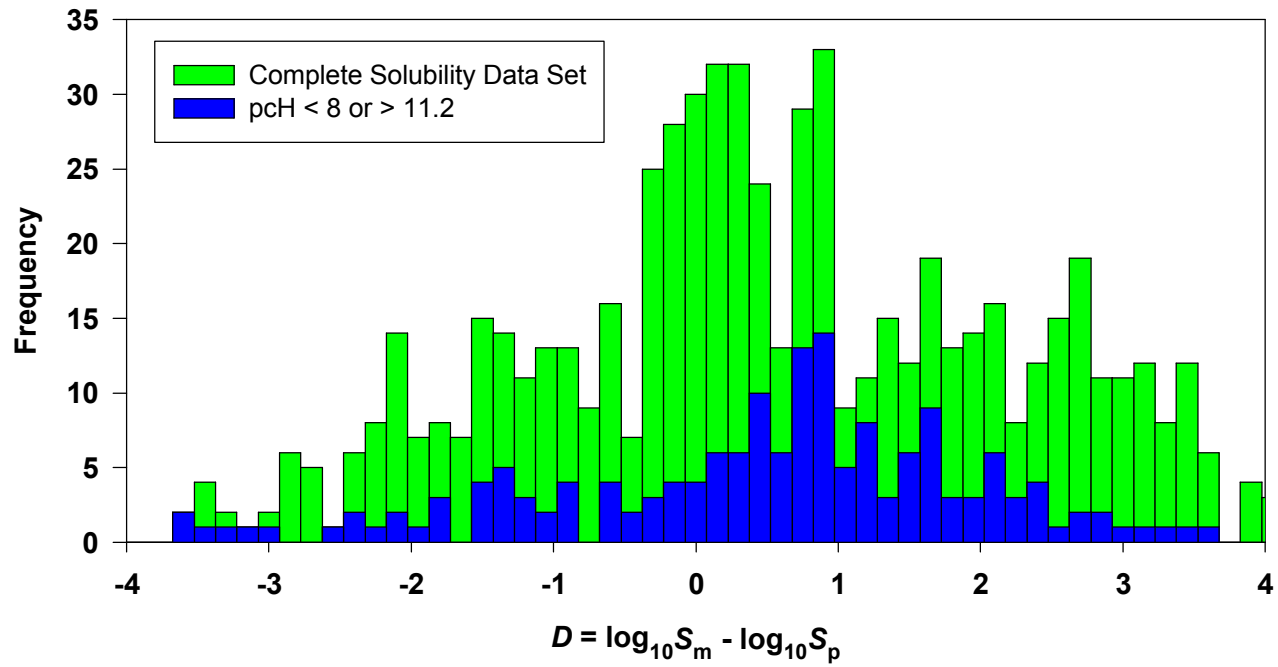
Americium (III) Uncertainty Analysis

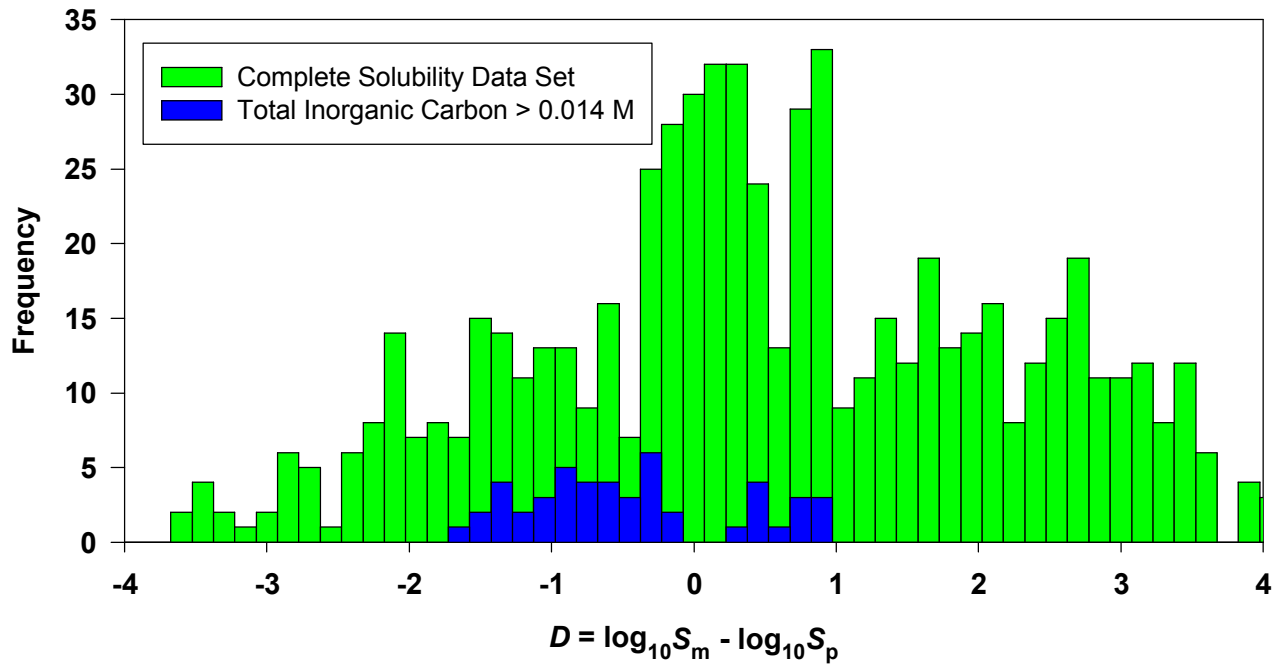
	CRA-2004 PABC	CRA-2009 PABC	Complete Solubility Data Set)	G11 Data Set
Maximum	2.85	2.70	3.95	2.97
Minimum	-3.00	-4.20	-3.67	-3.55
Mean	0.035	-0.142	0.441	-0.678
Median	-0.031	0.072	0.346	-0.866
Standard Dev.	0.900	1.17	1.67	1.57
<i>n</i>	243	441	647	172

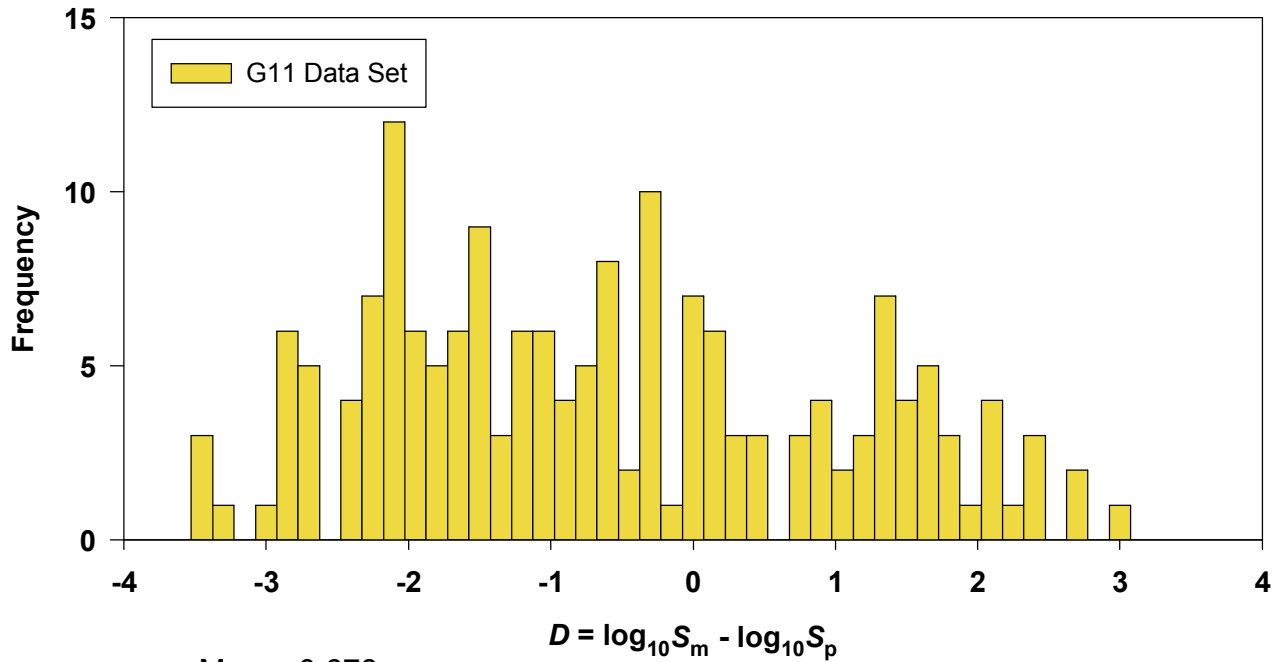


Mean 0.441
 Median 0.346
 Std Dev 1.67
 Minimum -3.67
 Maximum 3.95
 n = 647









Mean -0.678
Median -0.866
Std Dev 1.57
Minimum -3.55
Maximum 2.97
n = 172

Summary of Results

- The implementation of the G11 criterion
 - Th(IV) 64 of 185 experimental data points retained
 - Both the mean and median increased from negative values obtained for the CRA-2014 PA to positive values for CRA-2014 PA, Rev.1.
 - Am(III) 172 of 647 experimental data points retained
 - Both the mean and the median values decreased values from positive values obtained for the CRA-2014 PA to negative values for CRA-2014 PA, Rev.1.
 - The impact of implementing the G11 criterion on Performance Assessment is unknown.

Role of Laboratory Data

- Laboratory measured solubility data provides the “real” world bounds by which the WIPP baseline solubility uncertainty is determined.
- Measured laboratory data are the link between the theoretical geochemical model of the WIPP repository environment and independently measured values of actinide solubility.

Role of Screening Criteria

- To limit the data to studies that are adequately documented and represent chemical processes and systems that can be modeled with the WIPP QA-approved database and software.
- To consider experimental data that was obtained under experimental conditions at — or close to — the buffered values of pCH, ionic strength, and total inorganic carbon (TIC) content expected at WIPP.

References

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