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Sandia National Laboratories

SNL WIPP Geochemistry

Actinide Uncertainty Analysis:

Literature Screening Criteria Seminar

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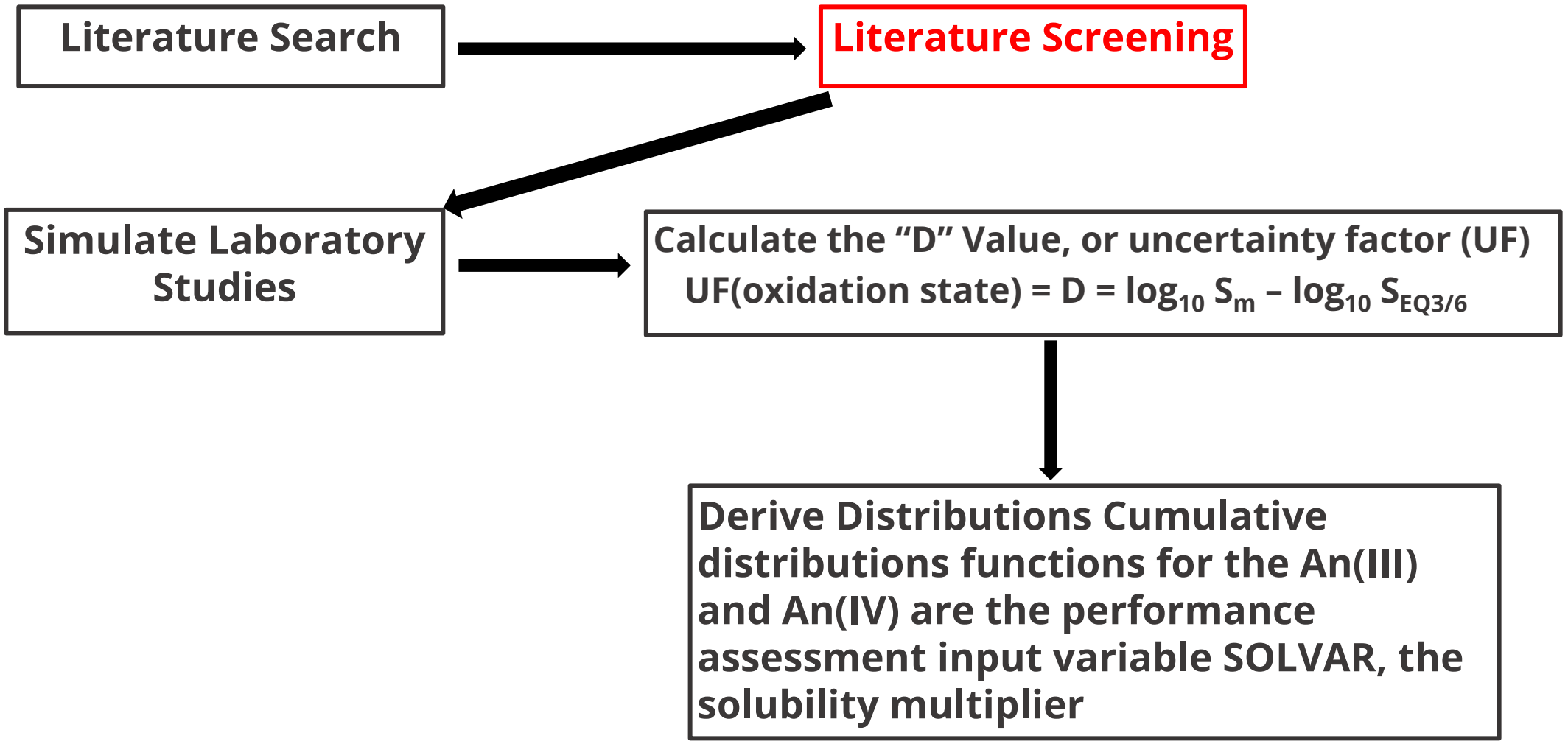
Meeting Purpose

The purpose of this June 30, 2021 seminar is to:

- Review the existing literature screening criteria for the WIPP actinide uncertainty analysis.
- Evaluate and consider changes to the literature screening criteria for the WIPP actinide uncertainty analysis.

It must be noted that changes to the screening criteria must be approved by the EPA at a future technical exchange.

Geochemistry-to-PA Actinide Solubility Process



Literature Screening Criteria

G1: Studies limited to the period beginning January 1990 to the closure date for the current recertification cycle.

G2. Include results from both papers published in peer-reviewed journals and unpublished reports (e.g., officially released reports from government laboratories such as Los Alamos National Laboratory, Lawrence Livermore National Laboratory, Pacific Northwest National Laboratory, etc.).

G3. Include only results from solubility studies. Exclude other studies that do not provide solubilities (e.g., studies of corrosion, leaching, sorption, or transport).

G4. Include only results from studies in which water was the solvent. Exclude studies in which other solvents were used (e.g., solids, molten metal or salts, or organic liquids).

G5. Include only results obtained from studies at pressures at or close to atmospheric, at temperatures at or close to those expected in the WIPP (i.e., 20°C - 30°C), and with post-test phase-separation methods similar to those used for the WIPP model, because these were the conditions and methods used to parameterize the Th(IV) and Am(III) speciation and solubility models for WIPP compliance-related PA calculations.

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. Exclude studies of U(IV), Np(IV), and Pu(IV); and Pu(III) because there could be systematic differences between the solubilities of these elements and those of their oxidation-state analogs used to parameterize the models; and because the difficulties inherent in maintaining these elements in these oxidation states could introduce experimental artifacts in the results obtained with these elements.

Literature Screening Criteria (continued)

G7. Include only results from studies with a characterized solubility-controlling solid for which solubility data are in the WIPP Th(IV) or Am(III) model (i.e., in the EPA-certified thermodynamic database), and in which the quantity of solid initially present was sufficient to prevent complete dissolution of this solid during the experiments.

G8. Include only results from studies with aqueous solutions of known composition. Exclude studies performed with groundwaters, sedimentary pore waters, and soil solutions that may contain unknown quantities of species that can be complexants or adsorbents (e.g., humic acids or other dissolved organic compounds, microbial colloids, or pseudocolloids).

G9. Include results from studies with dissolved elements or species that are present in WIPP brines. Exclude studies with dissolved elements or species that are absent in WIPP repository brines and for which our models do not include association/dissociation constants or Pitzer parameters.

G10. Include only results from studies for which the investigators provided a complete description of their experiments and the original solubilities. Exclude literature reviews and summaries, and studies in which the authors only provided average dissolved concentrations or solubility products, thus necessitating back-calculation of the solubilities.

G11. Include only results from experimental studies carried out under conditions at or close to those predicted for WIPP disposal rooms. Specifically, include only results from experiments in which:

Ionic Strength ≥ 3 m or M,

pcH = 8.0 ≤ pcH ≤ 11.2 , and

Total inorganic C 0.0 ≤ TIC ≤ 2 x 10-2 m or M

Proposed Enhancement to the Screening Criteria

General Screening Criteria – G1

- G1: Studies limited to the period beginning January 1990 to the closure date for the current recertification cycle.

SNL Comment:

Accepting studies from before 1990 will require setting a new start year, and determination of what constitutes acceptable analytical methods and experimental techniques for older studies.

General Screening Criteria – G2

- G2: Include results from both papers published in peer-reviewed journals and unpublished reports (e.g., officially released reports from government laboratories such as Los Alamos National Laboratory, Lawrence Livermore National Laboratory, Pacific Northwest National Laboratory, etc.).

General Screening Criteria – G3

- G3: Include only results from solubility studies. Exclude other studies that do not provide solubilities (e.g., studies of corrosion, leaching, sorption, or transport).

General Screening Criteria – G4

- G4: Include only results from studies in which water was the solvent. Exclude studies in which other solvents were used (e.g., solids, molten metal or salts, or organic liquids).

General Screening Criteria – G5

G5: Include only results obtained from studies at pressures at or close to atmospheric, at temperatures at or close to those expected in the WIPP (i.e., 20°C - 30°C), and with post-test phase-separation methods similar to those used for the WIPP model, because these were the conditions and methods used to parameterize the Th(IV) and Am(III) speciation and solubility models for WIPP compliance-related PA calculations.

SNL Comment:

The origin of the "post-test phase-separation" rule was the need to make equivalent comparisons between colloidal and non-colloidal Th(IV) used to parameterize the WIPP Th(IV) model. To enforce these equivalent comparisons, a decision was made to screen out studies that used experimental methods, such as ultra filtration and ultra centrifugation, both of which remove the colloidal material.

General Screening Criteria – G6

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. Exclude studies of U(IV), Np(IV), and Pu(IV); and Pu(III) because there could be systematic differences between the solubilities of these elements and those of their oxidation-state analogs used to parameterize the models; and because the difficulties inherent in maintaining these elements in these oxidation states could introduce experimental artifacts in the results obtained with these elements.

SNL Comment:

In the future if solubilities studies of U(IV), Np(IV), Pu(IV), and Pu(III) are to be screened in, LANL could assist us by providing criteria by which such studies can be considered valid given the concerns listed in the current G6. Note that the current G6 is an EPA approved criterion, and the DOE will have to defend any changes to it to the EPA.

General Screening Criteria – G7

G7: Include only results from studies with a characterized solubility-controlling solid for which solubility data is in the WIPP Th(IV) or Am(III) model (i.e., in the EPA-certified thermodynamic database), and in which the quantity of solid initially present was sufficient to prevent complete dissolution of this solid during the experiments (EPA, 2016).

Suggested Rewording:

G7: Include data from studies for which: (1) the solubility controlling phase is included in the WIPP An(III) or An(IV) model (i.e., in the EPA-certified thermodynamic database); (2) for undersaturation experiments the solubility controlling phase has been well characterized and the quantity of solid initially present was sufficient to prevent complete dissolution of this solid during the experiments; (3) for oversaturation experiments for which the solubility controlling phase(s) have been well characterized post-test.

General Screening Criteria – G8

G8: Include only results from studies with aqueous solutions of known composition. Exclude studies performed with groundwaters, sedimentary pore waters, and soil solutions that may contain unknown quantities of species that can be complexants or adsorbents (e.g., humic acids or other dissolved organic compounds, microbial colloids, or pseudocolloids).

General Screening Criteria – G9

G9: Include results from studies with dissolved elements or species that are present in WIPP brines. Exclude studies with dissolved elements or species that are absent in WIPP repository brines and for which our models do not include association/dissociation constants or Pitzer parameters (EPA, 2016).

Suggested Rewording:

G9: Include data from studies with dissolved elements or species that are present in WIPP brines, and are included in the WIPP An(III) or An(IV) solubility model(s). Include data from studies with dissolved elements or species not included in either WIPP brines or the WIPP An(III) or An(IV) solubility model(s) but for which the authors can demonstrate have no influence on the solubility of the actinide system being tested.

General Screening Criteria – G10

G10: Include only results from studies for which the investigators provided a complete description of their experiments and the original solubilities. Exclude literature reviews and summaries, and studies in which the authors only provided average dissolved concentrations or solubility products, thus necessitating back-calculation of the solubilities.

Suggested Rewording:

G10: Include data from studies which provide: (1) a complete description of the experimental and/or analytical procedures such that the experiment may be translated into an equivalent computer simulation; and (2) tabulated values of measured experimental quantities (i.e., concentrations, pH, ionic strength, etc.), in lieu of tabulated values graphic data displays, which can be digitized, may be accepted. Exclude literature reviews and summaries, and studies in which the authors only provided average dissolved concentrations or solubility products, thus necessitating back-calculation of the solubilities.

General Screening Criteria – G11

G11. Include only results from experimental studies carried out under conditions at or close to those predicted for WIPP disposal rooms. Specifically, include only results from experiments in which:

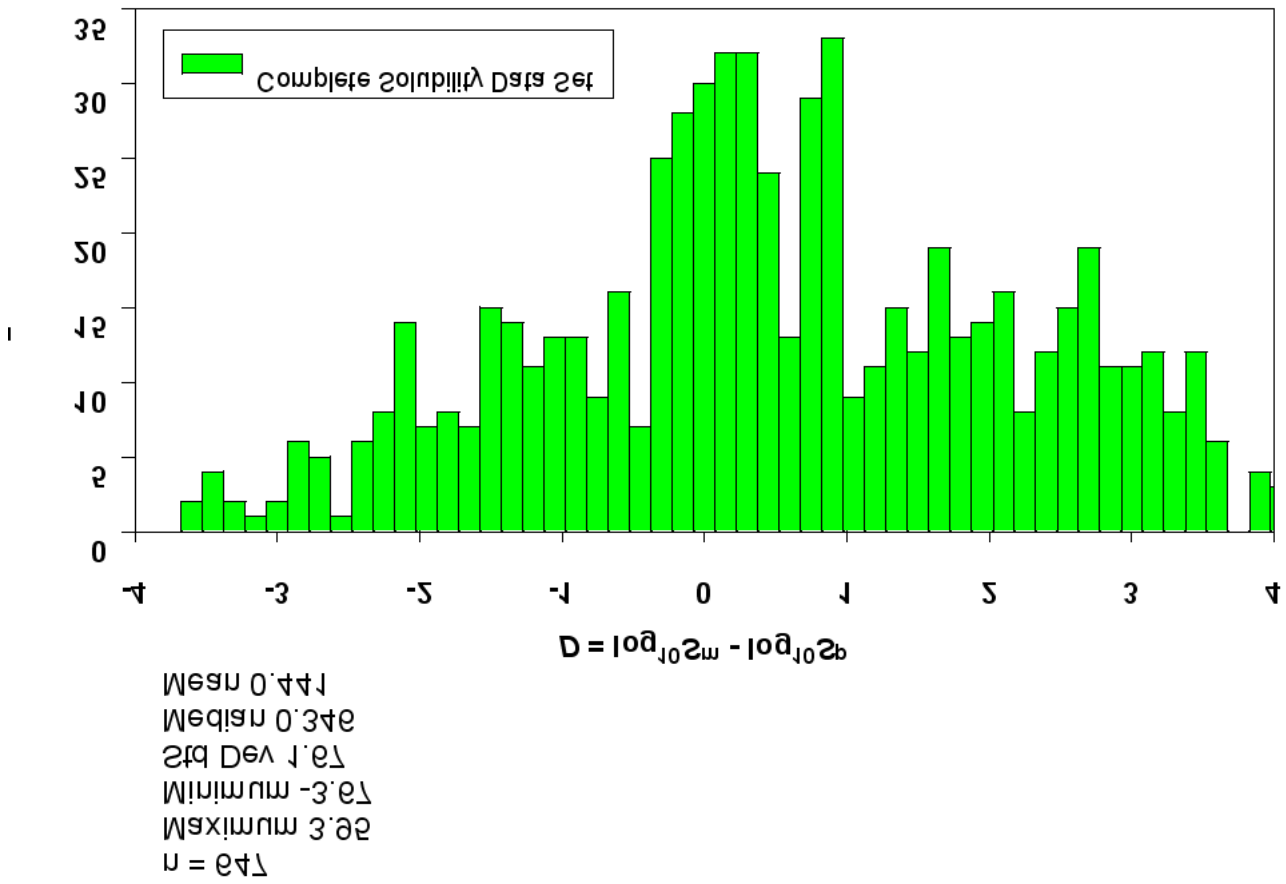
- Ionic Strength ≥ 3 m or M
- 8.0 ≤ pcH ≤ 11.2
- 0.0 ≤ TIC ≤ 2×10^{-2} m or M

SNL Comment:

Suggest setting maximum ionic strength at different values depending on the matrix solution. With NaCl solutions being the highest; $MgCl_2$ lower; and $CaCl_2$ the lowest ionic strength. This would be more representative of WIPP brines.

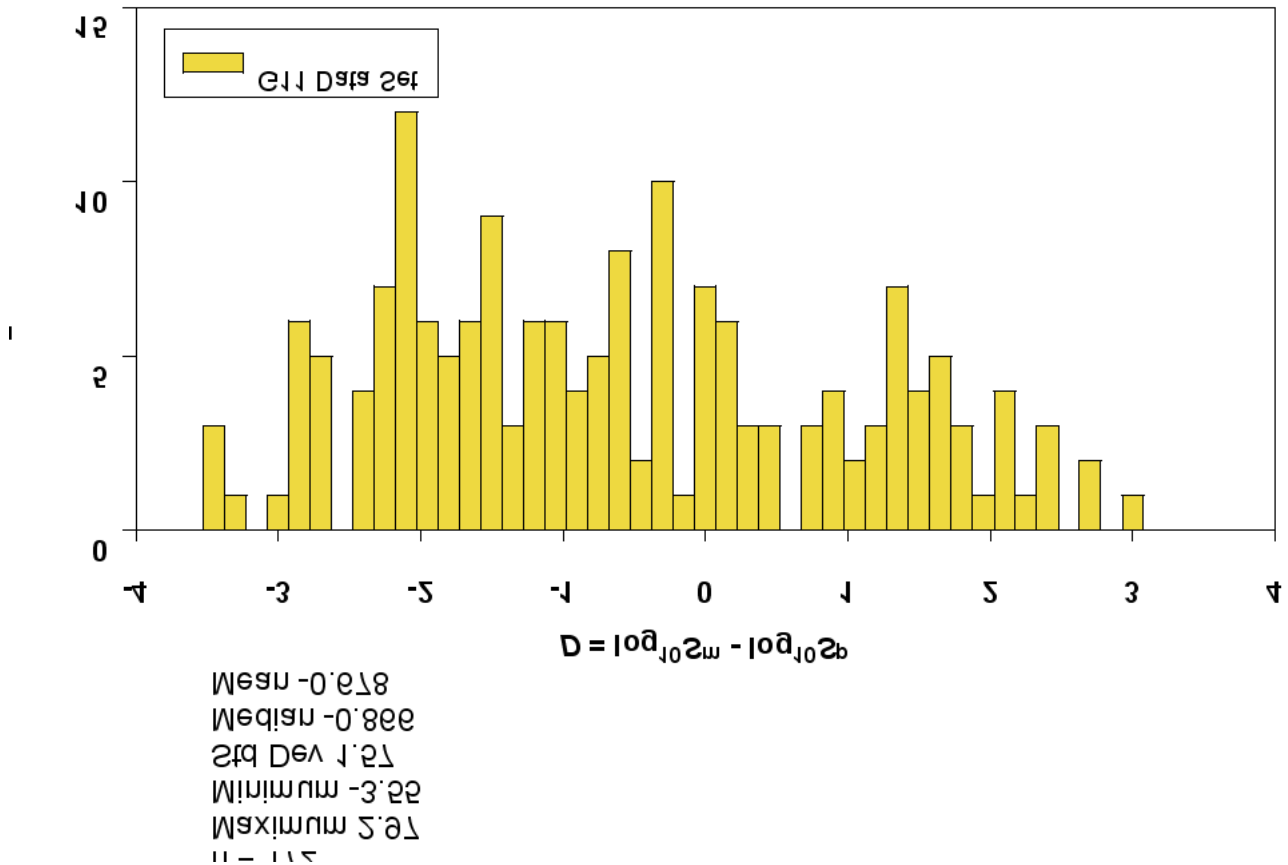
Current Application of General Screening Criteria – G11

Am(III) CRA-2014 Full Dataset



Current Application of General Screening Criteria – G11 (continued)

Am(III) CRA-2014 G11 criterion



New Suggested General Screening Criteria – G12

G12: Include only data from studies which have NOT been used to parameterize the WIPP An(III) or An(IV) models.

The End

References

EPA. 2016. “Reference and Data Selection for the CRA-2014 PABC Actinide Solubility Uncertainty Distributions”. U.S. Environmental Protection Agency. September 30, 2016. ERMS 567126.

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