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Subject: PA Implementation of Uncertainties Associated with Calculated Actinide Solubilities

INTRODUCTION

This write-up provides the following information: (1) a general description of how the uncertainty ranges and associated probability distributions for uncertain parameters are incorporated in WIPP performance assessment (PA); (2) a brief description of how the uncertainty range and probability distribution were established for actinide solubilities in 1996, and how they were used in the CCA PA, the 1997 PAVT, and the CRA-2004 PA; (3) a brief summary of how the range and distribution was re-established, and its proposed implementation in future PA calculations, starting with any verification calculations that might be required to support the EPA's recertification decision.

INCORPORATION OF RANGES AND DISTRIBUTIONS IN PA

This section describes the PA implementation of the uncertainty ranges and probability distributions associated with actinide solubilities calculated for the +III, +IV, and +V oxidation states (An(III), An(IV), or An(V)) - or estimated for the +VI oxidation state (An(VI)). This description uses the results of the comparison of measured and predicted An(III) solubilities (Xiong et al., 2004) as an example.

Construction of a Cumulative Distribution Function from a Histogram

The histogram in Figure 1 (see below), from Xiong et al. (2004), presents the results of 243 comparisons of An(III) solubilities measured in laboratory experiments and the solubilities predicted for the conditions used in each experiment by the An(III) thermodynamic speciation and solubility model implemented in the code Fracture-Matrix Transport (FMT).

The overall interval depicted in Figure 1, [-3.15, 3.15], is divided into 42 intervals of equal widths. (Each interval is 0.15 units wide.) Xiong et al. (2004) referred to these intervals as "bins." "Bin -3" refers to the interval containing the values greater than -3.15 and less than or equal to -3.00; "Bin -2.85" contains the values that are greater than -3.00 and less than or equal to -2.85, and so on. The values of "D" (the difference between logarithms of measured and predicted actinide solubilities) were sorted so that each value of "D" resides in one bin. This histogram is a plot of the bin values versus the number of "D" values that occur within the bin. The horizontal axis represents the measurement scale, and the boundaries of the bins are marked. The vertical axis represents the frequency.

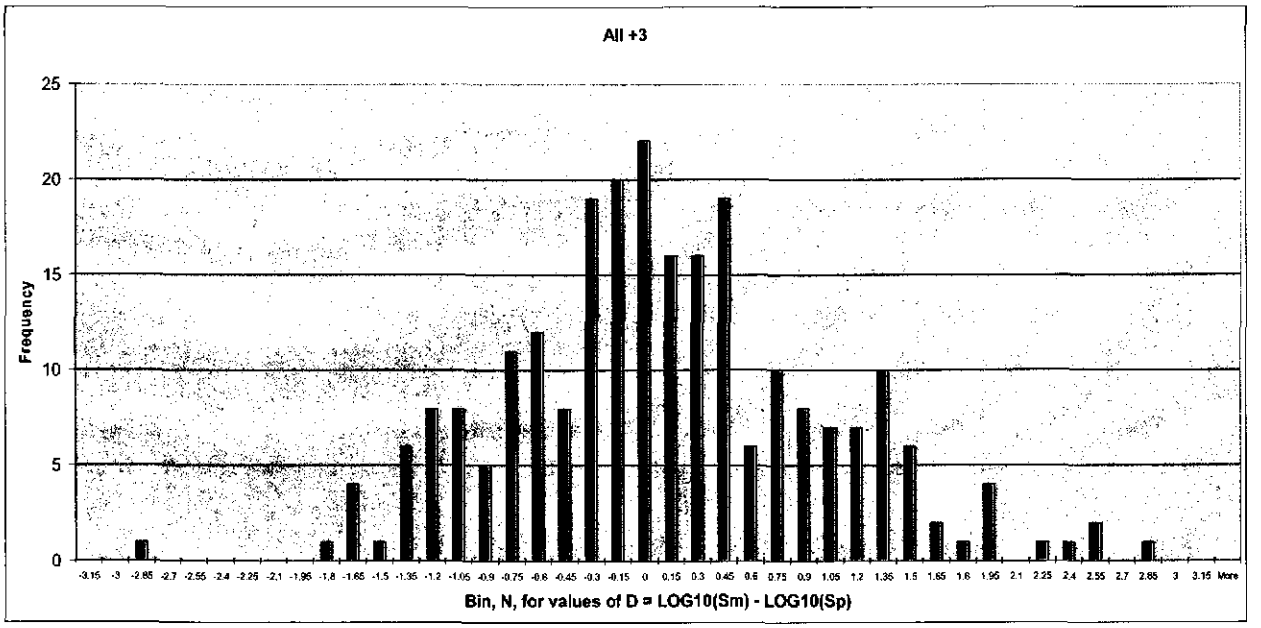

Figure 1. Histogram of An(III) comparisons by Xiong et al. (2004). A total of 243 measured and predicted solubilities were compared.

Using the histogram in Figure 1, Xiong et al. (2004) developed a cumulative distribution function (CDF) for the bins described above (see Figure 2 below). The CDF is a function that indicates the probability that a value of "D" is less than or equal to a specified bin value. For example, the CDF in Figure 2 equals 0 for Bin -3 since there are no values of "D" less than or equal to -3.00. Thus, the probability that "D" is less than or equal to -3.00 is 0. The histogram in Figure 1 confirms this observation. There is one occurrence of "D" in Bin -2.85. As there are a total of 243 different occurrences of "D," the probability that "D" is less than or equal to -2.85 is 1/243 or 0.00412. Bins -2.7 through -1.95 all have zero frequency. Thus, the value of CDF is 0.00412 for these bins as well. There are two occurrences when "D" is less than or equal to -1.80, so the probability that "D" is less than or equal to -1.80 is 2/243 or 0.00823. Thus, CDF is 0.00823 at -1.80. Similarly, the probability that "D" is less than or equal to -1.65 is 6/243, or 0.0247, and the CDF is 0.0247 at -1.65. This process is repeated to compute the probability for the rest of the bins. Ultimately, the probability that "D" is less than or equal to +2.85 is 1.00 since all values of "D" are less than or equal to +2.85

How Latin Hypercube Sampling Uses a CDF

The PA code Latin Hypercube Sampling (LHS) divides the probability axis into another set of equal-size bins. The size of these bins is determined by the number of realizations. For the case of 100 realizations, the probability bin size is 0.01. In a given bin on the probability axis, LHS randomly selects a probability value within this bin. LHS then selects the value of "D" that corresponds to this randomly selected probability values. LHS repeats this procedure for each of the given 100 probability bins. This process, which has been used for numerous other parameters that are sampled for WIPP PA, ensures that LHS samples from the complete range of values of "D."

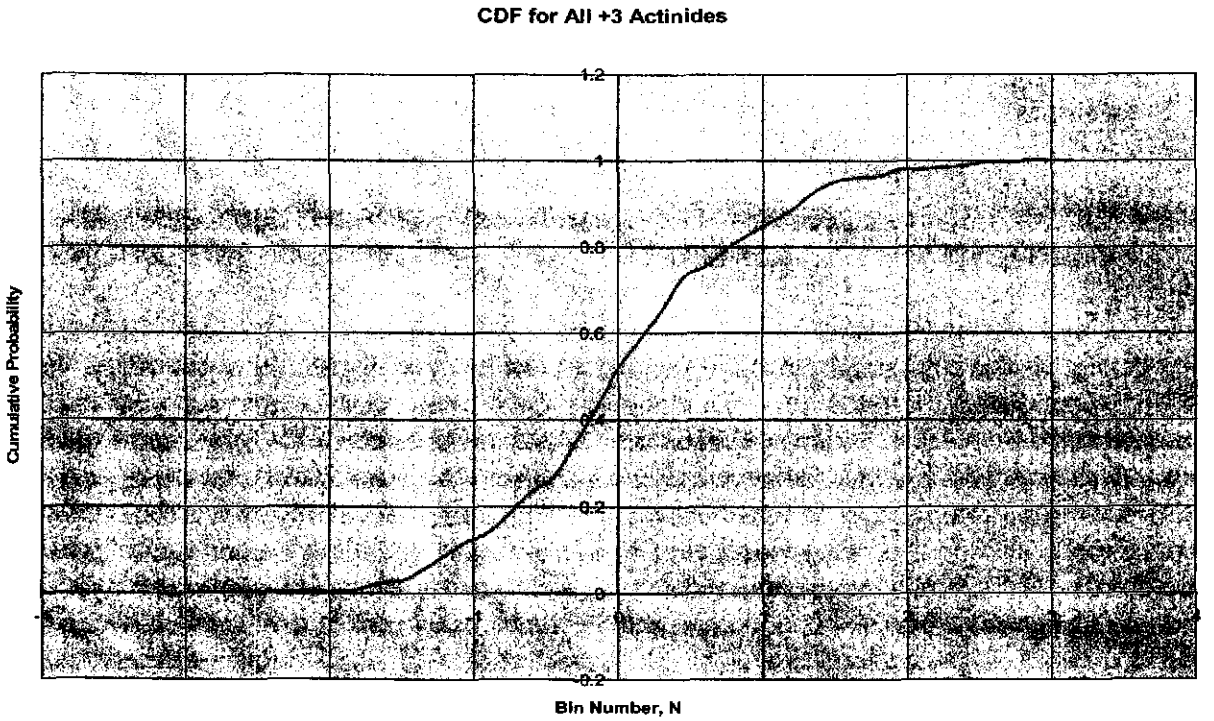

Figure 2. Plot of the CDF for the An(III) comparison of Xiong et al. (2004).

LHS uses the tabulated data from which a CDF is for sampling. CDF's are included only to illustrate the tabulated data.

HOW THE RANGE AND DISTRIBUTION WAS ESTABLISHED IN 1996, AND HOW IT WAS USED IN THE CCA PA, THE 1997 PAVT, AND THE CRA-20004 PA

Bynum (1996a, 1996b, 1996c) established the uncertainty range and probability distribution used for the CCA PA. This range and distribution were also used for the 1997 PAVT and the CRA-2004 PA. Bynum established one overall range and distribution that included comparisons for both the +III and the +V actinides. Bynum did not include any An(IV) comparisons in his range and distribution. Bynum's histogram and the CDF derived from his histogram are presented in U.S. DOE (1996, Appendix SOTERM, Figure SOTERM-6 and Figure SOTERM-7, respectively). The CDF shown in CCA Figure SOTERM-7 was derived from the histogram shown in CCA Figure SOTERM-6 in a manner similar to that described above for the recent results of Xiong et al. (2004), *except that*

the CDF in CCA Figure SOTERM-7 was simplified by limiting it to nine points. This simplification omitted some of the fine structure of the curve that would have been produced by including all of the bins in the histogram, but still captured the essence of Bynum's results.

For the CCA PA and the 1997 PAVT, the simplified distribution shown in CCA Figure SOTERM-7 was applied to each of the following ten sampled parameters: three U solubilities (two oxidation states - U(IV) and U(VI) - in the Salado brine, and one oxidation state - U(VI) - in the Castile brine), four for Pu (two oxidation states - Pu(III) and Pu(IV) - × both brines), two for Am (one oxidation state - Am(III) - × both brines), and one for Th (one oxidation state - Th(IV) - in the Salado brine). In the CRA-2004 PA, the simplified distribution used for the CCA PA and the 1997 PAVT was applied to 12 sampled parameters: four for uranium (two oxidation states × two brines), four for plutonium (same as above), two for americium (same as above), and two for thorium (one oxidation state × two brines).

HOW THE RANGES AND DISTRIBUTIONS WERE RE-ESTABLISHED, AND HOW WE PROPOSE TO USE THEM IN FUTURE CALCULATIONS

Xiong et al. (2004) re-established ranges and probability distributions associated with actinide solubilities calculated for future PA calculations. They established separate ranges and probability distributions for the +III, +IV, and +V actinides, and an overall range for all three of these oxidation states. Their analysis included 243 An(III) comparisons, 159 An (IV) comparisons, and 136 An(V) comparisons, for a total of 538 comparisons. Their histogram and CDF for the An(III) comparison are shown in Figures 1 and 2 above, respectively, and in Xiong et al. (2004, Figure 1 and Figure 2). Their results for the An(IV) and An(V) comparisons and their overall results are shown in Xiong et al. (2004, Figures 3 through 8). They also included the tabulated results used to plot the CDF's (Xiong et al., 2005, Tables 2 through 5). In all four cases ((An(III)), An(IV), An(V), and overall results), they constructed their CDF's from their histograms exactly as described above (i.e., no simplifications were used).

We propose that the results of Xiong et al. (2004) be used in future PA calculations as follows: (1) use the CDF from the An(III) comparison (Xiong et al., 2004, Table 2) for the calculated An(III); (2) use the CDF from the An(IV) comparison (Xiong et al., 2004, Table 3) for the calculated An(IV); (3) use the CDF from the overall results of the An(III), An(IV), and An(V) comparisons (Xiong et al., 2004, Table 5) for the estimated An(VI) solubility. (LHS uses the tabulated data for sampling. The figures in the report are only for illustration.) Therefore,

- Table 2 of Xiong et al. (2004) will be input into the Performance Assessment Parameter Database (PAPDB) with the proposed material name SOLMOD3 and property name SOLVAR;
- Table 3 of Xiong et al. (2004) will be input into the PAPDB with the proposed material name SOLMOD4 and property name SOLVAR;
- Table 5 of Xiong et al. (2004) will be input into the PAPDB with the proposed material name SOLMOD6 and property name SOLVAR.

We propose to sample each of the three distributions once per vector in all three 100-vector replicates.

We propose *not* to use the CDF from the An(V) comparison (Xiong et al., 2004, Table 4) for the calculated An(V) at this time because Np is the only actinide that will speciate in the +V oxidation

state under the conditions expected in the WIPP, and because there will not be sufficient Np present in the WIPP to affect the long-term performance of the repository whatever its solubility. (The solubility of the +V oxidation state has never been sampled by LHS.)

We propose to define the variables sampled in the previous CRA-2004 PA with these three sampled values. Thus, the 12 sampled variables that pertained to solubilities in the CRA-2004 PA would be replaced by these new sampled variables as follows. (Note that the nomenclature used is the Material Name/Property Name.)

- For the +III actinides, Pu(III) and Am(III):
  - SOLAM3/SOLSIM will be set to the value of SOLMOD3/SOLVAR;
  - SOLAM3/SOLCIM will be set to the value of SOLMOD3/SOLVAR;
  - SOLPU3/SOLSIM will be set to the value of SOLMOD3/SOLVAR;
  - SOLPU3/SOLCIM will be set to the value of SOLMOD3/SOLVAR;
- For An(IV), Th(IV), U(IV), and Pu(IV):
  - SOLPU4/SOLSIM will be set to the value of SOLMOD4/SOLVAR;
  - SOLPU4/SOLCIM will be set to the value of SOLMOD4/SOLVAR;
  - SOLU4/SOLSIM will be set to the value of SOLMOD4/SOLVAR;
  - SOLU4/SOLCIM will be set to the value of SOLMOD4/SOLVAR;
  - SOLTH4/SOLSIM will be set to the value of SOLMOD4/SOLVAR;
  - SOLTH4/SOLCIM will be set to the value of SOLMOD4/SOLVAR;
- For An(VI), U(VI):
  - SOLU6/SOLSIM will be set to the value of SOLMOD6/SOLVAR;
  - SOLU6/SOLCIM will be set to the value of SOLMOD6/SOLVAR.

The solubility calculated by FMT for each brine (Salado or Castile) will be multiplied by the antilogarithm of the appropriate sampled value. For example, for the +III actinides (Pu(III) and Am(III)), the solubility calculated by FMT will be multiplied by the antilog of the sampled value of SOLMOD3/SOLVAR. (Note that "SOLMOD3/SOLVAR" refers to the Material Name and the Property Name.)

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