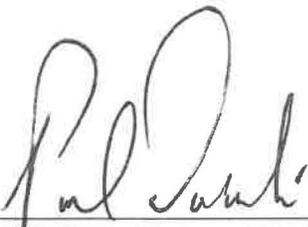
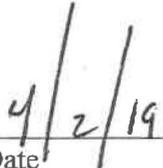
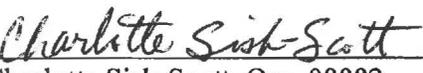
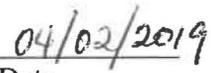
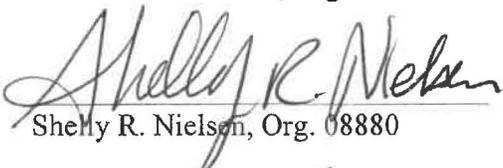
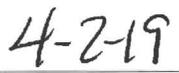
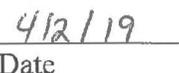


Sandia National Laboratories  
Waste Isolation Pilot Plant

## Uncertainty Analysis of Actinide Solubilities for CRA 2019

Work Carried Out under Task 6 of the Analysis Plan for WIPP Near-Field  
Geochemical Process Modeling, AP 153, Rev. 1.  
To be included in the AP-153 records package

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## 1 INTRODUCTION

The actinide solubility uncertainty analysis provides distributions of the solubility multiplier SOLVAR for the oxidation state III and oxidation state IV model material SOLMOD3 and SOLMOD4 in the WIPP PA Parameter Database for the CRA-2019 PA. These distributions were created by applying the WIPP geochemical model to peer-reviewed independent laboratory experimental data that meet project specific criteria, and by calculating the difference between the measured data and the data simulated using the WIPP model. Multiple studies for both the An(III) and the An(IV) model were included in this uncertainty analysis, and were used to build the oxidation-state specific uncertainty distributions.

This analysis report documents the creation of the uncertainty distributions for CRA 2019. Documentation includes the project specific screening criteria, discussions of which studies were screened out and which were included, the EQ3/6 model setup for the included studies, presentation of the An(III) and An(IV) uncertainty distributions, and a discussion of the results. The final uncertainty distributions will be transmitted to PA in the form of an official memo that will be issued after this report is submitted to records.

We do not include U(VI) in our analysis because a thermodynamic speciation-and-solubility model has not been developed for U(VI). Instead, the EPA specified (U.S. EPA, 2006) that an estimate of  $1 \times 10^{-3}$  M be used for the solubility of U(VI) in Generic Weep Brine (GWB) and Energy Research and Development Administration (WIPP Well) 6 (ERDA-6).

Additionally, we do not include Np(V) in our analysis because PA does not sample the uncertainties in the solubilities of Np in view of the fact that releases of this element do not affect the long-term performance of the WIPP (Brush and Garner, 2005).

The EQ3/6 software package, Version 8.0a, (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010; Xiong, 2011b) is used for this, and all WIPP related geochemical modeling exercises. An updated Pitzer thermodynamic database (DB) DATA0.FM4 (Domski 2019) is being implemented for the first time in the CRA 2019 PA uncertainty analysis. This same database was used for the CRA 2019 PA baseline solubility model (Domski and Sisk-Scott 2019).

Wolery (2008), Wolery et al. (2010) and Xiong (2011b) completed the qualification of Version 8.0a of EQ3/6 according to Sandia National Laboratories' (SNL's) WIPP quality assurance (QA) procedures for WIPP compliance-related actinide-solubility calculations.

We conduct our analysis under Task 6 of AP-153, Rev. 1, the current analysis plan (AP) for WIPP near-field geochemical process modeling (Brush et al., 2012, Subsection 4.4). This analysis was performed using the same process described as used through the submission of CRA-2014, and therefore is a deviation from AP-153, Rev. 1.

Table 1 defines the abbreviations, acronyms, initialisms, etc., used in this report.

**Table 1. Abbreviations, Acronyms, Initialisms, etc.**

Abbreviation, Acronym, or Initialism	Definition
Am, Am(III)	americium, americium in the +III oxidation state
am	amorphous
anhydrite	CaSO <sub>4</sub> (cr)
AP	analysis plan
aq	aqueous (dissolved in an aqueous solution)
atm	atmosphere(s)
C, c	carbon, crystalline
Ca, Ca <sup>2+</sup>	calcium, calcium ion
CAP	(SNL/WIPP QA) corrective action plan
CAR	(SNL/WIPP QA) corrective action request
CDF	cumulative distribution function
citrate	(CH <sub>2</sub> COO) <sub>2</sub> C(OH)(COO) <sup>3-</sup> or (CH <sub>2</sub> CO <sub>2</sub> ) <sub>2</sub> C(OH)(CO <sub>2</sub> ) <sup>3-</sup>
Cl, Cl <sup>-</sup>	chloride, chloride ion
ClO <sub>4</sub> , ClO <sub>4</sub> <sup>-</sup>	perchlorate, perchlorate ion
Cm, Cm(III)	curium, curium in the +III oxidation state
CMS	(Sandia/WIPP software) Configuration Management System
CO <sub>3</sub> , CO <sub>3</sub> <sup>2-</sup>	carbonate, carbonate ion
col	colloidal (suspended in an aqueous or other solution)
CPR	cellulosic, plastic and rubber (materials)
cr	crystalline

Table 1 continued on next page

**Table 1. Abbreviations, Acronyms, Initialisms, etc. (Cont.)**

Abbreviation, Acronym, or Initialism	Definition
CRA-2004	the first WIPP Compliance Recertification Application, submitted to the EPA in March 2004
CRA-2009	the second WIPP Compliance Recertification Application, submitted to the EPA in March 2009
CRA-2014	the third WIPP Compliance Recertification Application, submitted to the EPA in March 2014
D	the difference(s) between logs (base 10) of the measured and predicted actinide solubilities ( $S_m$ and $S_p$ )
EPA	(U.S.) Environmental Protection Agency
EQ3/6	a geochemical software package for speciation and solubility calculations (EQ3NR) and reaction-path calculations (EQ6), Version 8.0a
ERDA-6	Energy Research and Development Administration (WIPP Well) 6, a synthetic brine representative of fluids in Castile brine reservoirs
$f_{CO_2}$	fugacity (similar to the partial pressure) of $CO_2$
Fm.	Formation
FMT	Fracture-Matrix Transport, a geochemical speciation and solubility code
G-Seep	a naturally-occurring brine formerly collected from G Drift in the WIPP underground workings and used for laboratory and modeling studies
GWB	Generic Weep Brine, a synthetic brine representative of intergranular Salado brines
H, $H^+$	hydrogen, hydrogen ion
$HCO_3$ , $HCO_3^-$	bicarbonate, bicarbonate ion
$H_2O$	water
I	ionic strength, defined by $I = \frac{1}{2} \times \sum_i (M_i \times z_i^2)$ , in which $M_i$ and $z_i$ are the molarity and charge of species $i$

Table 1 continued on next page

**Table 1. Abbreviations, Acronyms, Initialisms, etc. (Cont.)**

Abbreviation, Acronym, or Initialism	Definition
I/O	(EQ3/6) input and output files
K, K <sup>+</sup>	potassium, potassium ion
LIB	Library
log	the common logarithm or logarithm (base 10)
M	molar, mol•dm <sup>-3</sup>
m	molal, mol•kg <sup>-1</sup>
mcr	microcrystalline
Mg, Mg <sup>2+</sup>	magnesium, magnesium ion
mM	millimolar
N	bin number (see Section 2 for explanation)
Na, Na <sup>+</sup>	sodium, sodium ion
NaTr	sodium trifluoromethanesulfonate (also abbreviated as sodium triflate)
Nd, Nd(III)	neodymium, neodymium in the +III oxidation state
NO <sub>3</sub> , NO <sub>3</sub> <sup>-</sup>	nitrate, nitrate ion
Np, Np(IV), Np(V)	neptunium, neptunium in the +IV oxidation state , neptunium in the +V oxidation state
O	oxygen
OH, OH <sup>-</sup>	hydroxide, hydroxide ion
P	probability
PA	performance assessment
PABC	(WIPP) Performance Assessment Baseline Calculations, carried out in 2005 and 2009
pCH	the negative log (base 10) of the molar concentration of H <sup>+</sup>

Table 1 continued on next page

**Table 1. Abbreviations, Acronyms, Initialisms, etc. (Cont.)**

Abbreviation, Acronym, or Initialism	Definition
$p_{\text{CO}_2}$	partial pressure (similar to the fugacity) of $\text{CO}_2$
pmH	the negative log (base 10) of the molal concentration of $\text{H}^+$
ppm	parts per million
pH	the negative log (base 10) of the activity of $\text{H}^+$
Pu, Pu(III), Pu(IV)	plutonium, plutonium in the +III oxidation state, plutonium in the +VI oxidation state
QA	quality assurance
R	gas constant
Rev.	Revision
$S_m$	measured solubility
$S_p$	predicted solubility
SNL	Sandia National Laboratories
$\text{SO}_4, \text{SO}_4^{2-}$	sulfate, sulfate ion
T	temperature
Th, Th(IV)	thorium, thorium in the +IV oxidation state
TIC	total inorganic C (the sum of the dissolved species of inorganic C)
U, U(IV), U(VI)	uranium, uranium in the +IV oxidation state, uranium in the +VI oxidation state
$\mu\text{m}$	micrometer
whewellite	$\text{CaC}_2\text{O}_4 \cdot \text{H}_2\text{O}$
WIPP	Waste Isolation Pilot Plant
XRD	X-ray diffraction
Zr, Zr(IV)	zirconium, zirconium in the +IV oxidation state
$\mu^0, \mu^0/RT$	standard chemical potential, dimensionless standard chemical potential

## 2 METHODS

The method employed in the actinide uncertainty analysis has gone largely unchanged since the CCA PA. The core of the method is the creation of frequency distributions based on the difference between the base 10 logarithm of measured laboratory solubility data and solubility data simulated with the WIPP model. These differences are compiled and sorted for a number of studies and the cumulative probability is calculated. These cumulative distribution functions (CDFs) are sampled by the PA code and used as multiplier for the baseline solubility values for performance assessment. While the core of the method has remained constant since the CCA PA various aspects of the method have evolved and been refined over the years. The evolution of the actinide uncertainty analysis is summarized in Table 2.

**Table 2. Evolution of the Actinide Uncertainty Analysis.**

PA Event	Pertinent Information	Code/Database	References
CCA PA PAVT CRA-2004 PA	Established the frequency distribution for the differences of log (base 10) of measured solubilities and logarithms of the values predicted for comparable conditions as the standard.  Combined single distribution for An(III), An(IV) and An(V). Based on Np(V) and Am(III) data.  CRA-2004 PA used the same distributions as CCA and PAVT.	FMT/CHEMDAT	Bynum (1996a, 1996b, 1996c)
CRA-2004 PABC	Separate distributions for An(III), An(IV) and An(V) <sup>A</sup> .  Screening criteria informally implemented for laboratory studies.	FMT/CHEMDAT	Xiong et al., (2004), Xiong et al., (2005)
CRA-2009 PA	Re-used the CRA-2004 PABC distributions.	FMT/CHEMDAT	U.S. DOE (2009)
CRA-2009 PABC	Screening criteria formally documented for laboratory studies.	FMT/CHEMDAT	Xiong et al., (2009) Xiong et al. (2011)
CRA-2014 PA	Migrated to EQ3/6 included all of the previous lab studies and new studies.  Implemented the G11 screening criteria, which effectively	EQ3/6 / DATA0.FM1	Xiong (2011a) Brush and Domski(2013a, 2013b)

PA Event	Pertinent Information	Code/Database	References
	eliminated special criterion S1.		
SEN4	Major update to DATA0.FM1 and re-screening of lab studies based of EPA completeness comments.	EQ3/6 / DATA0.FM2  DATA0.FM1 for Rev. 1	Domski (2015) Xiong and Domski (2016)
CRA-2019	Major update to DATA0.FM1, not incorporating DATA0.FM2 changes. Slight modifications to the screening criteria.	EQ3/6 / DATA0.FM4	Domski (2019)

A – The An(V) distribution has never been implemented by PA because Np(V) does not affect long-term performance.

The current uncertainty analysis follows this procedure:

1. Identify potential laboratory solubility studies using internet search tools.
2. Screen laboratory studies by applying our screening criteria.
3. Create EQ3/6 input files to emulate as closely as possible the laboratory studies screened in from Step 2.
4. Execute the EQ3/6 inputs using the DATA0.FM4 Pitzer thermodynamic database and post process the outputs using the Excel macro GetEQdata.xlsx, i.e., extract the pH and An(III) or An(IV) concentrations.
5. Calculate, D, the log base 10 differences between the measured data and the simulated data.
6. Combine the D values for all oxidation state specific studies, sort the D values, and calculate the cumulative probability.
7. Document all of the above steps in an analysis report (this report).
8. Provide a memo to PA which includes the distributions of the solubility multiplier SOLVAR for the oxidation state III and oxidation state IV model material SOLMOD3 and SOLMOD 4, respectively, in the WIPP PA Parameter Database for the CRA-2019 PA.

We use ten general criteria that were informally implemented by Xiong et al. (2005), and formally documented by Xiong et al. (2009 and 2011) for CRA-2004 PABC and CRA-2009 PABC, respectively, for including results of experimentally measured solubilities in their comparisons of measured Th(IV), Nd(III), Am(III), and Cm(III) solubilities and predicted Th(IV) and Am(III) solubilities. The general criteria were:

- G1. Include only results from experimental studies published from January 1, 1990, through 2011. Note, the CRA-2014 was limited to January 1990 to November 2011, and at EPA's direction no additional literature search was done for the CRA-2019.
- 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., 20-30 °C), and with posttest phase-separation methods similar to those used for the WIPP, 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.

G7. Include only results from studies with a characterized solubility-controlling solid for which the value of the dimensionless standard chemical potential ( $\mu^0/RT$ ) or thermodynamic equilibrium constant ( $\log K^0$ ) 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.

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 (EPA, 2016).

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.

An eleventh criterion, G11, was implemented by Brush and Domski (2013b):

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: (1)  $I \geq 3$  m or M, (2)  $\text{pcH} = 8.0\text{--}11.2$ , and (3) total inorganic C (TIC) =  $0 - 2 \times 10^{-2}$  M. (In WIPP brines after equilibration with the important solids in the repository, the TIC concentration is essentially the sum of the concentrations of the dissolved species  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$  and is closely related thermodynamically to  $f_{\text{CO}_2}$  or  $P_{\text{CO}_2}$ .)

For the CRA 2019 uncertainty analysis no new laboratory studies were considered, therefore the screening criteria were not implemented. This was directed by the EPA (Peake, 2018) that we only use the studies used in the SEN 4 uncertainty analysis (Domski and Xiong 2016), with the exception of Khalili (1994) which was eliminated from consideration.

EQ3/6 input files created for SEN4 to predict the solubilities of Th(IV), Nd(III), and Am(III) for the selected laboratory studies were used for the CRA-2019 uncertainty analysis. All of the laboratory studies with the exception of one, Rao et al., (1999) were simulated with the EQ3NR code. In the EQ3NR runs, the pH was specified; the composition of the laboratory supporting solution was used as input, either in molar or molal units; and the radionuclide composition was calculated based on equilibrium with the solubility-controlling phase in the experiment. In instances in which the data are reported in molarity, it was necessary to input the following EQ3 input parameters: total dissolved solids (tdspl), solution density (rho), and the aqueous-phase scale factor (scamas). These parameters are calculated based on the fluid composition in the MS-Excel spreadsheet "Conc\_density\_calcs\_brines.xls." This spreadsheet will be submitted to the SNL/WIPP Records Center.

In the case of the Rao et al. (1999) runs it was necessary to use both EQ3NR and EQ6 to simulate these experiments. In this case, the experiments were conducted with a carbon dioxide ( $\text{CO}_2$ ) atmosphere, and they were titrated to specific values of pH at which the solubility measurements were made.

For those experiments that measured the pcH, a two-step approach was used to match the measured pcH. EQ3NR does not accept pcH as input though it does provide pcH as an output, which allows using an initial guess at the pcH in an initial EQ3/6 run, and then calculating a differential to calculate a new initial pH which will provide the correct output pcH. To arrive at the measured value of pcH, an initial EQ3/6 run was performed in which the input pH was referred to as  $\text{pH}_{\text{init}}$ . This resulted in an output value of pcH referred to as  $\text{pcH}_{\text{init}}$ . The difference between  $\text{pcH}_{\text{init}}$  and the measured experimental value  $\text{pcH}_{\text{exp}}$  was then calculated and used to calculate a new corrected input  $\text{pH}_{\text{correct}}$ , and a second EQ3/6 run was performed to obtain a simulated pcH equal to  $\text{pcH}_{\text{exp}}$ .

$$pH_{correct} = pH_{init} + (pcH_{exp} - pcH_{init})$$

Each of the EQ3/6 files for each of the laboratory studies were executed, and the resulting EQ3NR output files (\*.3o), or EQ6 output files (\*.6o) were post processed using the Excel macro GetEQData\_v101f.xls (included in the records package). The results for the An(III) and An(IV) are compiled in the spreadsheets *An(III)\_CRA-2019\_Uncertainty.xlsx* and *AN(IV)\_CRA-2019\_Uncertainty.xlsx*, respectively, both of which are included in the records package. Each of the laboratory studies are saved on separate tabs which are clearly labeled. These two spreadsheets also contain the probability distributions and CDF and histogram plots.

The frequency distributions were generated of the differences ( $D$ ) between the logs of the measured and predicted actinide solubilities ( $S_m$  and  $S_p$ , respectively), and displayed as histograms in Microsoft Excel spreadsheets using Excel's frequency function in this commercial spreadsheet software. One other deviation was implemented. AP153, Rev. 1 (Brush et al., 2012) specifies that histograms of the data will be produced by binning the data and implies that the data will be used in this form. However, the Latin Hypercube Sampling (LHS) code when implementing a distribution function uses the cumulative-distribution function (CDF) rather than probability density functions (PDF) such as the histograms shown in the figures (see Subsection 3.1 and 3.2 below). The CDF must be constructed using the unbinned data so that those data can be submitted directly to PA for entry into their parameter database.

Negative values of  $D$  indicate that the Th(IV) or Am(III) model implemented in EQ3/6 predicted a solubility greater than the corresponding measured value (overprediction of solubility by the model); positive values of  $D$  indicate that the model predicted a solubility less than the measured value (underprediction). The Excel files "*An(III)\_CRA-2019\_Uncertainty.xlsx*" and "*AN(IV)\_CRA-2019\_Uncertainty.xlsx*" contain spreadsheets with  $S_m$  or  $S_p$  values (or logs thereof), frequency distributions, and histograms for this analysis. The variable  $D$  is calculated via:

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

Where  $S_m$  is the measured solubility, and  $S_p$  is the predicted solubility.

We will submit electronic copies of the files "*An(III)\_CRA-2019\_Uncertainty.xlsx*" and "*AN(IV)\_CRA-2019\_Uncertainty.xlsx*" and all of the other spreadsheets to the archived file CRA-2019PA.ZIP, which is stored at "/nfs/data/CVSLIB/WIPP\_EXTERNAL/ap153\_task6".

In this analysis, we use the DATA0.FM4 to perform calculations to construct the uncertainty distributions based on the data sets selected by EPA. We follow the rounding rules of Gamsjäger et al. (2012) for the numbers in the CDFs generated in this analysis. The same methods and criteria in Brush and Domski (2013b) are used in this analysis.

Table 3 provides the location and run-control information for the thermodynamic database, Excel spreadsheets, and EQ3/6 I/O files for the calculations for the original analysis.

**Table 3. Run-Control Information for the Calculations Carried Out for the Original Analysis.**

Description or Title of File(s)	Location of File(s)
EQ3/6 Database DATA0.FM4	In zip file Uncertainty_CRA-2019PA.ZIP, /nfs/data/CVSLIB/WIPP_EXTERNAL/ap153_task6
Excel macro GetEQData_v101e.xls and GetEQData_v101f.xls	In zip file Uncertainty_CRA-2019PA.ZIP, /nfs/data/CVSLIB/WIPP_EXTERNAL/ap153_task6
EQ3/6 I/O files and Excel spreadsheets with input and extracted data	In zip file Uncertainty_CRA-2019PA.ZIP, /nfs/data/CVSLIB/WIPP_EXTERNAL/ap153_task6
Peake (2018) Letter to Mike Brown	In zip file Uncertainty_CRA-2019PA.ZIP, /nfs/data/CVSLIB/WIPP_EXTERNAL/ap153_task6

### 3 RESULTS

This section provides the results of the CRA 2019 PA actinide solubility uncertainty analysis for Th(IV) (Subsection 3.1) and Am(III) (Subsection 3.2). Only the laboratory studies used in the CRA 2019 PA uncertainty analysis will be discussed in sections 3.1 and 3.2. Xiong and Domski (2016) provide detailed descriptions and explanations of the published papers and unpublished reports identified and evaluated previously by Xiong et al. (2005) and/or Xiong et al. (2011), and the papers and reports evaluated by Brush and Domski (2013a). These lists will not be repeated in this analysis report.

#### 3.1 Th(IV) Solubility Model

The results of the Th(IV) uncertainty analysis are provided in this section.

The EPA selected the following data sets for construction of An(IV) solubility uncertainty distribution (U.S. EPA, 2016):

- Inclusion of 18 samples in 3 M NaCl,  $8.0 \leq \text{pcH} \leq 11.2$  from Felmy et al. (1991)
- Inclusion of 5 ultracentrifuged samples in 5 M NaCl and 5 ultracentrifuged samples in 2.5 M MgCl<sub>2</sub> from Altmaier et al. (2004)
- Inclusion of 12 samples in 4 M NaCl with 0.02 M TIC from Altmaier et al. (2006)
- Inclusion of 4 samples in 5.26 M CaCl<sub>2</sub> with  $\text{pcH} < 10$  from Altmaier et al. (2008)

Table 4 provides detailed descriptions of the laboratory studies used in the CRA 2019 uncertainty analysis, and discussions of whether these studies met or violated the selection criteria presented in Section 2.

**Table 4. Final Evaluations of Lab Studies for Comparisons of Measured and Predicted Th(IV) Solubilities.**

Reference	Solution(s) and Ionic Strength(s)	pH, pCH, or pmH	Solubility-Controlling Solid	Evaluation and EQ3/6 Run Numbers for Results Included in This Analysis
Altmaier et al. (2004)	(A) 0.5 M NaCl (B) 5 M NaCl (C) 0.25 M MgCl <sub>2</sub> (I = 0.75 M) (D) 2.5 M MgCl <sub>2</sub> (I = 7.5 M) (E) 4.5 M MgCl <sub>2</sub> (I = 13.5 M)	(A) pCH = 10.4 & 11-13.5 (B) pCH = 10.8 (C) pCH = 9.0 (D) pCH = 8.9 (E) pCH = 8.8	ThO <sub>2</sub> (cr) & ThO <sub>n</sub> (OH) <sub>4-2n</sub> ·xH <sub>2</sub> O(am), which they referred to as “Th(OH) <sub>4</sub> (am) or ThO <sub>2</sub> ·xH <sub>2</sub> O(am)”	<p>Not considered by Xiong et al. (2005) because this paper came to their attention after completion of their analysis. Six uncentrifuged results (no replicates) obtained at I ≥ 3 M included by Xiong et al. (2011) and Brush and Domski (2013a): 01ALT04.3i – 06ALT04.3i. All 6 results satisfied criterion G11 in Brush and Domski (2013b) and they also included them in their analysis. In the analysis performed by Xiong and Domski (2015), they also adopt these results for that analysis. Ultracentrifuged results excluded by Xiong et al. (2011) and Brush and Domski (2013a) because the posttest phase-separation method was different than those employed in the studies used to parameterize the Th(IV) model and appeared to support a different model (see text). These results are also excluded in Xiong and Domski (2015).</p> <p>In this report, we include ultracentrifuged samples, five in 5 M NaCl and five in 2.5 M MgCl<sub>2</sub>, exclude all of uncentrifuged results, per EPA’s instruction, for our analysis.</p>
Altmaier et al. (2006)	NaHCO <sub>3</sub> -Na <sub>2</sub> CO <sub>3</sub> -NaCl with I = 0.1-4 M.	pCH = 8–11	Th(OH) <sub>4</sub> (am) or ThO <sub>2</sub> ·xH <sub>2</sub> O(am)	<p>Most results excluded by Xiong et al. (2011) and from this analysis because I &lt; 3 M. All other results excluded because the Th(OH)(CO<sub>3</sub>)<sub>4</sub><sup>5-</sup> complex found to be important by Altmaier et al. (2006) is not in the WIPP Th(IV) model. Twelve results (no duplicates) obtained at I ≥ 3 M modeled but excluded by Xiong et al. (2011) and Brush and Domski (2013a, 2013b). These results were also excluded in the analysis performed by Xiong and Domski (2015).</p> <p>In this report, we include 12 samples in ~4 M NaCl with 0.02 M TIC, per EPA’s instruction, for our analysis.</p>

Table 4 continued on next page

**Table 4. Final Evaluations of Lab Studies for Comparisons of Measured and Predicted Th(IV) Solubilities (Cont.)**

Reference	Solution(s) and Ionic Strength(s)	pH, pCH, or pmH	Solubility-Controlling Solid	Evaluation and EQ3/6 Run Numbers for Results Included in This Analysis
Altmaier et al. (2008)	(A) 0.2–5.26 m CaCl <sub>2</sub> (I = 0.6–15.78 m) (experiments with Th(IV)) (B) 2.40 m Ca(ClO <sub>4</sub> ) <sub>2</sub> (I = 7.20 m) (Th(IV))	pmH = 9.3–12.1 (all Th(IV))	Solutions with CaCl <sub>2</sub> < 1 M (entire pH range) or CaCl <sub>2</sub> > 1 M (pH < 11.5): Th(OH) <sub>4</sub> (am) or ThO <sub>2</sub> ·xH <sub>2</sub> O(am). Solutions with CaCl <sub>2</sub> > 1 M (pH > 11.5): unidentified, possibly a Ca-Th solid(cr).	Most results excluded by Xiong et al. (2011) and Brush and Domski (2013a, 2013b) because I < 3 M. Some results excluded because of ultracentrifugation. All other results excluded because the Ca <sub>4</sub> (Th(OH) <sub>8</sub> ) <sup>4+</sup> complex found to be important by Altmaier et al. (2008) is not in the WIPP Th(IV) model. These results were also excluded in the analysis performed by Xiong and Domski (2015).  In this report, we include 4 samples in 5.26 M CaCl <sub>2</sub> with pCH < 10, per EPA's instruction, for our analysis.
Felmy et al. (1991)	(A) 0.1 M NaClO <sub>4</sub> (B) 0.6 M NaCl (C) 1.2 M NaCl (D) 3.0 M NaCl (E) 0.6 M KCl	(A) pCH = 4.9–5.9 (B) pCH = 2.91–10.24 (C) pCH = 2.83–10.19 (D) pCH = 3.82–10.56 (E) pCH = 3.75–7.95	Th(IV) hydrous oxide (all)	Forty-five results (no duplicates) obtained at I = 3 M included in Xiong et al. (2005, 2011) and Brush and Domski (2013a): 01FEL91.3i – 46FEL91.3i. I = 3 M results obtained at pCH ≤ 3.6 excluded by Xiong et al. (2005, 2011) and Brush and Domski (2013a) because the solids dissolved before saturation was attained. After application of criterion G11, Brush and Domski (2013b) excluded 18 results (no replicates) in their analysis. EQ3 files: 15FEL91.3i, 23FEL91.3i – 31FEL91.3i, 39FEL91.3i – 46FEL91.3i, File 16FEL91.3i not included because experimental pH ≤ 3.6. Xiong and Domski (2015) included 18 results from Brush and Domski (2013b) for their analysis.  In this report, we include 18 samples in 3 M NaCl, 8.0 ≤ pCH ≤ 11.2, per EPA's instruction, for our analysis.

Figure 1 provides the histogram and CDF for our comparisons of experimentally measured and predicted Th(IV) solubilities for CRA 2019 PA. The current comparison includes a total of 44 measured and predicted solubilities. The current distribution peaks at Bin -0.5 with a frequency of 8 comparisons, with a secondary peak at Bin 0.85 with 7 comparisons. Table 5 summarizes the statistical parameters of the current frequency distribution for CRA 2019 PA.

Inspection of Figure 1 shows that the WIPP Th(IV) model, the EQ3/6 code, and the DB Data0.FM4 overpredicted 23 ( $D < 0$ ) of the 44 measured solubilities included in the current analysis and underpredicted 21 ( $D > 0$ ). The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Th(IV) solubilities are -0.0114 and -0.090, respectively (Table 5), and the mean and median values of the sampled uncertainty factor are  $10^{0.0114} = 0.974$  and  $10^{-0.090} = 0.813$ . (The WIPP PA codes use actinide solubilities that are the products of the baseline solubilities and the sampled uncertainty factors).

The cumulative distribution function (CDF) for Th(IV) for CRA 2019 PA is tabulated in Table 6. Table 6 provides values of cumulative distribution function (CDF) for Th(IV) in response to EPA selected data sets for CRA 2019 PA. PA will use this CDF to define the solubility multiplier SOLVAR for the oxidation-state IV model, material SOLMOD4 in the WIPP PA Parameter Database in response to EPA selected data sets for CRA 2019 PA.

Appendix A provides a breakdown of D values by author and study and the descriptive statistics for each data set.

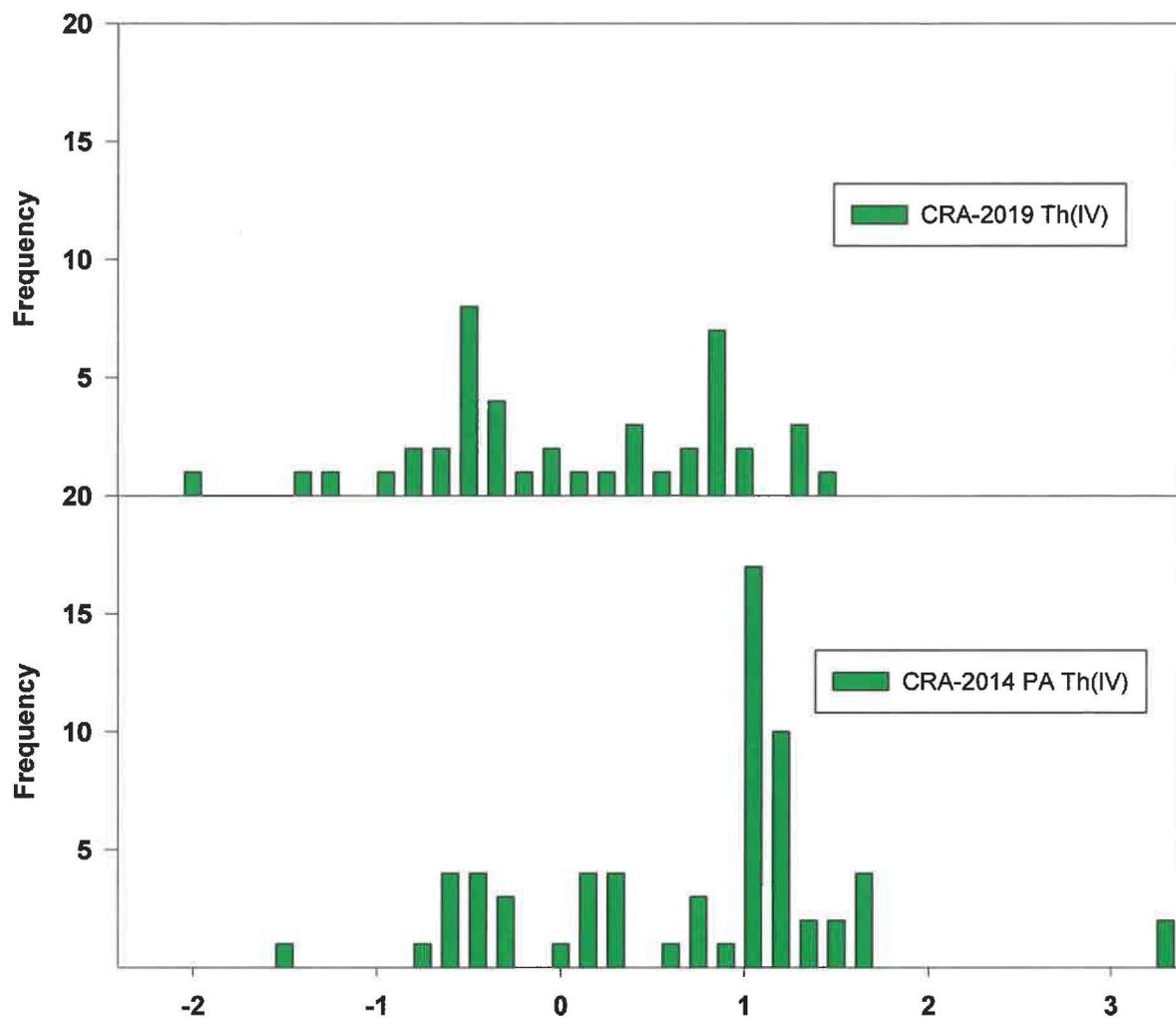
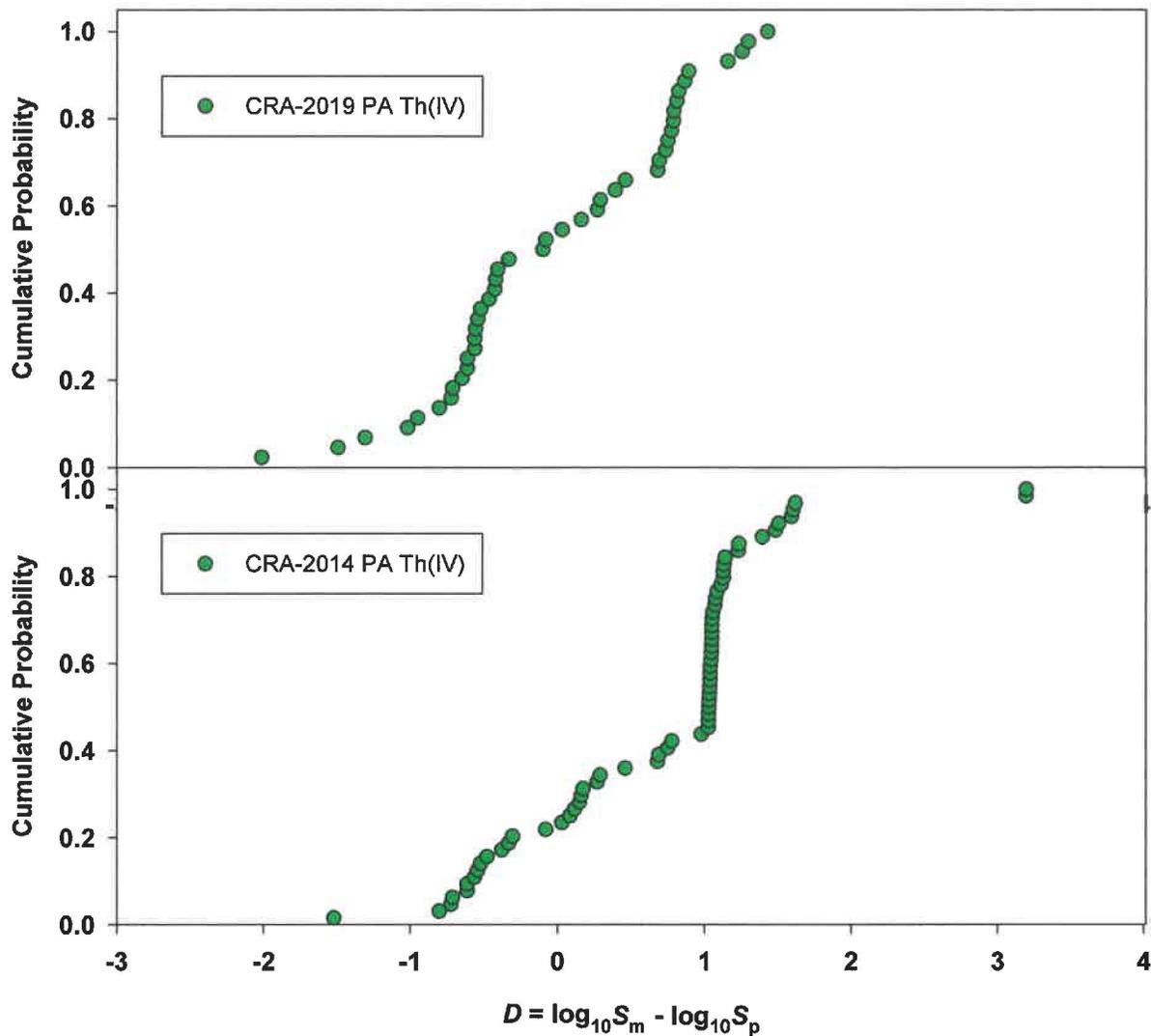


Figure 1. Histograms for CRA-2019 (top) and CRA-2014 (bottom) of the difference  $D = \log_{10} S_m - \log_{10} S_p$  for Th(IV) CRA-2019.



**Figure 2. CDF plots for CRA-2019 (top) and CRA-2014 (bottom) of the difference  $D = \log_{10} S_m - \log_{10} S_p$  for Th(IV) CRA-2019.**

**Table 5. Statistical Properties of the Distribution of  $D = \log_{10}(S_m) - \log_{10}(S_p)$  for Th(IV) in response to EPA selected data sets for CRA-2019 compared with the CRA-2014 PA data (Brush and Domski, 2013b).**

Analysis	Mean	Median	Standard Deviation	Minimum	Maximum
CRA-2014 PA	0.673	1.029	0.869	-1.52	3.19
CRA-2019 PA	-0.011	-0.090	0.821	-2.01	1.43

**Table 6. CDF of the Differences between Measured and Predicted Th(IV)  
Solubilities for CRA 2019 PA.  
This CDF Defines the Solubility Multiplier SOLVAR for the Oxidation-State IV Model  
Material SOLMOD4 in the WIPP PA Parameter Database.**

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D	$P(D < X)$ for CRA 2019 PA
-2.0098	0.0227
-1.4885	0.0455
-1.3031	0.0682
-1.0158	0.0909
-0.9475	0.1136
-0.8003	0.1364
-0.7203	0.1591
-0.7103	0.1818
-0.6458	0.2045
-0.6103	0.2273
-0.6103	0.2500
-0.5612	0.2727
-0.5603	0.2955
-0.5552	0.3182
-0.5403	0.3409
-0.5203	0.3636
-0.4645	0.3864
-0.4252	0.4091
-0.4192	0.4318
-0.4041	0.4545
-0.3303	0.4773
-0.0996	0.5000
-0.0803	0.5227
0.0297	0.5455
0.1597	0.5682
0.2697	0.5909
0.2897	0.6136
0.3919	0.6364
0.4597	0.6591
0.6797	0.6818

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Table 6 continued on next page

**Table 6. CDF of the Differences between the Measured and Predicted Th(IV) Solubilities for CRA 2019 PA (Cont.)**

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D	$P(D < X)$ for CRA 2019 PA
0.6897	0.7045
0.7340	0.7273
0.7497	0.7500
0.7740	0.7727
0.7873	0.7955
0.7890	0.8182
0.8085	0.8409
0.8228	0.8636
0.8640	0.8864
0.8897	0.9091
1.1567	0.9318
1.2526	0.9545
1.2953	0.9773
1.4266	1.0000

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### 3.2 Am(III) Solubility Model

This subsection describes the comparisons by this analysis of the experimentally measured Am(III) solubilities with those predicted using the WIPP Am(III) speciation and solubility model in EQ3/6. It includes information similar to that provided for Th(IV) in Subsection 3.1 (see above). It provides a revised CDF for the differences between our measured and predicted Am(III) solubilities in response to EPA selected data sets for CRA 2019 PA.

The measured Am(III) solubilities from published papers and unpublished reports evaluated previously (Xiong et al., 2005; and/or Xiong et al., 2011) include: Borkowski et al. (2009), Borkowski (2010), Bunyakina and Storozhenko (2004), Khalili et al. (1994), Lucchini et al. (2007), Makino et al. (1993), Meinrath and Kim (1991), Meinrath and Takeishi (1993), Peretrukhin et al. (1996), Rao et al. (1996a, 1996b, 1999), Runde et al. (1992), Runde and Kim (1995), Silva (1982), and Wood (2002) (see Xiong and Domski 2016).

The measured Am(III) solubilities from published papers and unpublished reports that Brush and Domski (2013a) evaluated for their analysis include: Borkowski et al. (2010), Borkowski (2012), Migdisov and Williams-Jones (2007), Neck et al. (2009), Rabung et al. (2008) and Vitorge and Tran The (1991) (see Table 7).

The evaluations of the above data in this analysis are similar to Xiong et al. (2005, 2011) and Brush and Domski (2013a, 2013b). The changes based on the data sets selected by EPA for construction of An(III) solubility uncertainty distribution are indicated as follows:

- Twenty-four samples from Khalili et al. (1994) in G-Seep brine which were included in SEN4, per EPA's instructions have since been eliminated based on DOE's reasoning of the poor quality of this data set.
- The 15 results in ERDA-6 from Rao et al. (1999), were included in this analysis, per EPA's instructions.
- The 8 samples in 5 M NaCl; 5 samples in 1 M MgCl<sub>2</sub>, 3 samples in 2.5 M MgCl<sub>2</sub>, 8 samples in 5 M MgCl<sub>2</sub>, 9 samples in 1 M CaCl<sub>2</sub>, and 23 samples in 3 M CaCl<sub>2</sub> from Neck et al. (2009), were included in this analysis, per EPA's instructions.
- Eleven 5 M NaCl samples of Runde and Kim (1995) were included in this analysis, per EPA's instructions.

**Table 7. Final Evaluations of Lab Studies for Comparisons of Measured and Predicted Nd(III) and Am(III) Solubilities.**

Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation and EQ3/6 Run Numbers for Results Included in This Analysis
Khalili et al. (1994)	Synthetic brine similar to GWB	pcH = 6.4, 8.4, 10.4, & 12.4	Nd(OH) <sub>3</sub> ·nH <sub>2</sub> O(am) at pcH = 6.4 & 8.4, Nd <sub>2</sub> (CO <sub>3</sub> ) <sub>3</sub> ·8H <sub>2</sub> O or “uncertain” at pcH = 10.4, NdO(OH) & Nd(OH) <sub>3</sub> or “uncertain” at pcH = 12.4	<p>Twenty-nine results for pcH = 6.4 &amp; 8.4 included by Xiong et al. (2005) because Am(OH)<sub>3</sub> is in the WIPP Am(III) model. Results for pcH = 10.4 &amp; 12.4 excluded by Xiong et al. (2005) because Nd<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub>·8H<sub>2</sub>O and NdO(OH) are not in the model, and because it is unclear that all of the solids were identified. Twenty-four of the 29 results included by Xiong et al. (2005) excluded by Xiong et al. (2011) and Brush and Domski (2013a) because the pcH in these runs exceeded 7.4, the pcH above which Borkowski concluded that complexation of Nd(III) by borate in GWB is significant (see text). Five results (5 duplicates) included by Brush and Domski (2013a). These results do not satisfy criterion G11 in Brush and Domski (2013b) and were not included in their revised analysis. In the analysis by Xiong and Domski (2015), although there were the results with borate that satisfy G11 and could be run by using the updated WIPP thermodynamic database, per our acceptance of the EPA Comment 3-C-4, these results were not used because the solubility controlling phase was Nd(OH)<sub>3</sub>·nH<sub>2</sub>O(am) in their experiments, which is not in the WIPP thermodynamic database.</p> <p>In this analysis, 24 results in G-Seep brine at pcH 8.4 are not included, per EPA’s instructions.</p>

Table 7 continued on next page

**Table 7. Final Evaluations of Lab Studies for Comparisons of Measured and Predicted Nd(III) and Am(III) Solubilities (Cont.)**

Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation and EQ3/6 Run Numbers for Results Included in This Analysis
Neck et al. (2009)	(A) 0.1, 0.5, 2.5, & 5.0 M NaCl; 0.1 M NaClO <sub>4</sub>	(A) pcH ≈ 7 to 13, 14, or even 15	(A) Nd(OH) <sub>3</sub> or Cm(OH) <sub>3</sub> (5 M NaCl & 1.0, 2.5, & 3.5 M CaCl <sub>2</sub> )	<p>Not considered by Xiong et al. (2011) because this paper came to their attention after completion of their analysis. All 161 results from NaCl and MgCl<sub>2</sub> solutions included by Brush and Domski (2013a). After application of criterion G11 by Brush and Domski (2013b), 8 results from NaCl solutions (no replicates) were included in this revised analysis: 95NeckNa.3i through 102NeckNa.3i, and 01NeckMg.3i through 42NeckMg.3i. After application of criterion G11 by Brush and Domski (2013b), 16 (no replicates) results from MgCl<sub>2</sub> solutions were included in their analysis: 17-21NeckMg.3i, 27-29NeckMg.3i, 35-42NeckMg.3i. All 46 results from CaCl<sub>2</sub> with pcH &lt; 10 included by Brush and Domski (2013a). Following the application of criterion G11 by Brush and Domski (2013b), 32 (2 replicates) results from CaCl<sub>2</sub> solutions were included in their analysis: 15NeckCa.3i through 46NeckCa.3i. Results with pcH &gt; 10 excluded because the complexes Ca(Cm(OH)<sub>3</sub>)<sup>2+</sup>, Ca<sub>2</sub>(Cm(OH)<sub>4</sub>)<sup>3+</sup>, and Ca<sub>3</sub>(Cm(OH)<sub>6</sub>)<sup>3+</sup> found to be important by Neck et al. (2009) are not in the WIPP Am(III) model. In the analysis by Xiong and Domski (2015), they included the results used by Brush and Domski (2013b).</p> <p>In this analysis, we include eight samples in 5 M NaCl; five samples in 1 M MgCl<sub>2</sub>, three samples in 2.5 M MgCl<sub>2</sub>, eight samples in 5 M MgCl<sub>2</sub>, nine samples in 1 M CaCl<sub>2</sub>, ten samples in 2.5 M CaCl<sub>2</sub>, and 13 samples in 3.5 M CaCl<sub>2</sub>, per EPA's instructions.</p>
	(B) 0.25, 1.0, 2.5, & 3.5 M MgCl <sub>2</sub>	(B) pcH ≈ 7 or 7.5 to 9		
	(C) 0.25, 1.0, 2.5, & 3.5 M CaCl <sub>2</sub>	(C) pcH ≈ 7-9 to 12		

Table 7 continued on next page

**Table 7. Final Evaluations of Lab Studies for Comparisons of Measured and Predicted Nd(III) and Am(III) Solubilities (Cont.)**

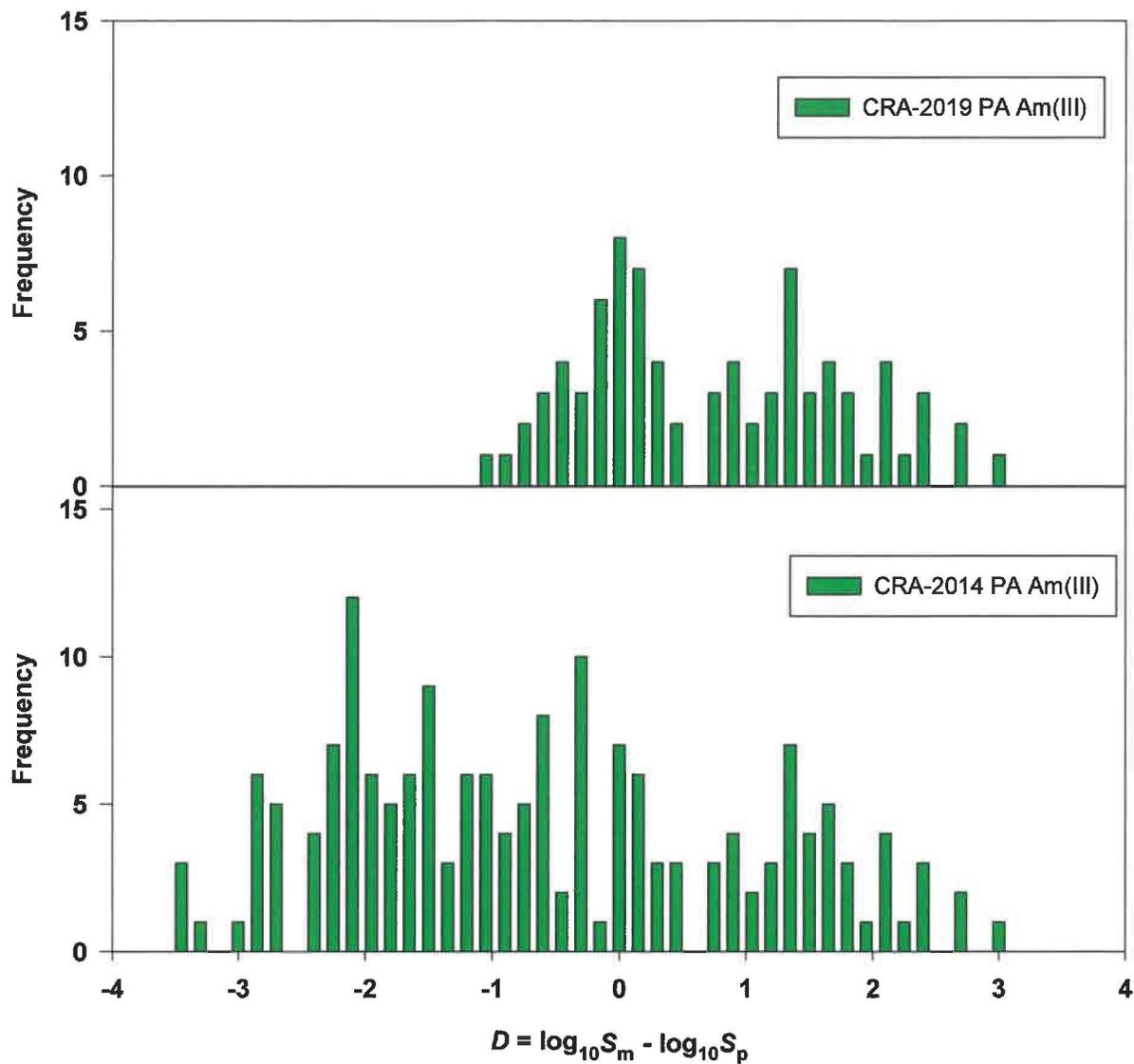
Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation and EQ3/6 Run Numbers for Results Included in This Analysis
Rao et al. (1999)	(A) 0.1-1.1 m NaHCO <sub>3</sub>	(A) pH = 7.9-8.3	NaNd(CO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O (all)	One-hundred-and-five results included in Xiong et al. (2005). Twenty-eight of the 105 results included by Xiong et al. (2005) excluded by Xiong et al. (2011) and Brush and Domski (2013a) because the pcH in these runs exceeded 7.4 or 8.1, the pcH above which Borkowski concluded that complexation of Nd(III) by borate in GWB or ERDA-6, respectively, is significant (see description of Borkowski et al., 2009, in text). Seventy-seven results (25 replicates) included by Brush and Domski (2013a), but only 71 of the EQ3/6 runs attained equilibrium. After application of criterion G11 by Brush and Domski (2013b), one of these results was included in their analysis: 46Rao99.3i. In Xiong and Domski (2015), 12 additional results from ERDA-6 containing borate were included (47 – 58Rao99.6o) using the updated database, that met all criteria, and per acceptance of the EPA Comment 3-C-5.
	(B) 0.1-2 m Na <sub>2</sub> CO <sub>3</sub>	(B) pH = 10.4-10.8		
	(C) 2 M NaCl & 0.1-0.5 m NaHCO <sub>3</sub>	(C) pH = 7.9-8.3		
	(D) 2 M NaCl & 0.1-2 m Na <sub>2</sub> CO <sub>3</sub>	(D) pH = 10.4-10.8		
	(E) 4 M NaCl & 0.1-2 m Na <sub>2</sub> CO <sub>3</sub>	(E) pcH = 10.4-10.8		
	(F) G Seep, a synthetic brine similar to GWB	(F) pcH = 5.6-7.8		
	(G) ERDA-6	(G) pcH = 6.2-10.6		
Runde & Kim (1995)	(A) 5.0 M NaCl	(A) pcH = 6.7-13.8	(A) Am(OH) <sub>3</sub> (cr) (B) NaAm(CO <sub>3</sub> ) <sub>3</sub> ·xH <sub>2</sub> O(cr)	Seventy one results (18 duplicates) included by Xiong et al. (2005, 2011) and by Brush and Domski (2013a). Following the application of criterion G11 by Brush and Domski (2013b), 6 results (no replicates) were included in their analysis: 10Run95.3i – 15Run95.3i. The results used in Brush and Domski (2013b) are included in the analysis by Xiong and Domski (2015).
	(B) 5.0 m NaCl & 0.01 atm CO <sub>2</sub>	(B) pcH = 6.6-9.3		

Figure 3 provides the histograms of the CRA-2019 PA and CRA-2014 PA frequency distributions for comparison. Figure 4 displays the CDF for the CRA-2019 PA and CRA-2014 for our comparison of experimentally measured and predicted Nd(III) and Am(III) solubilities in response to EPA's selected data sets for CRA 2019 PA. The current comparison includes a total of 82 measured and predicted solubilities. The current distribution peaks at Bin 0.00 with a frequency of 8 comparisons, and with a secondary peak at Bin 1.3 with 7 comparisons. Table 8 summarizes the statistical parameters of the current frequency distribution generated in this analysis with that of CRA-2014 PA (Brush and Domski, 2013b).

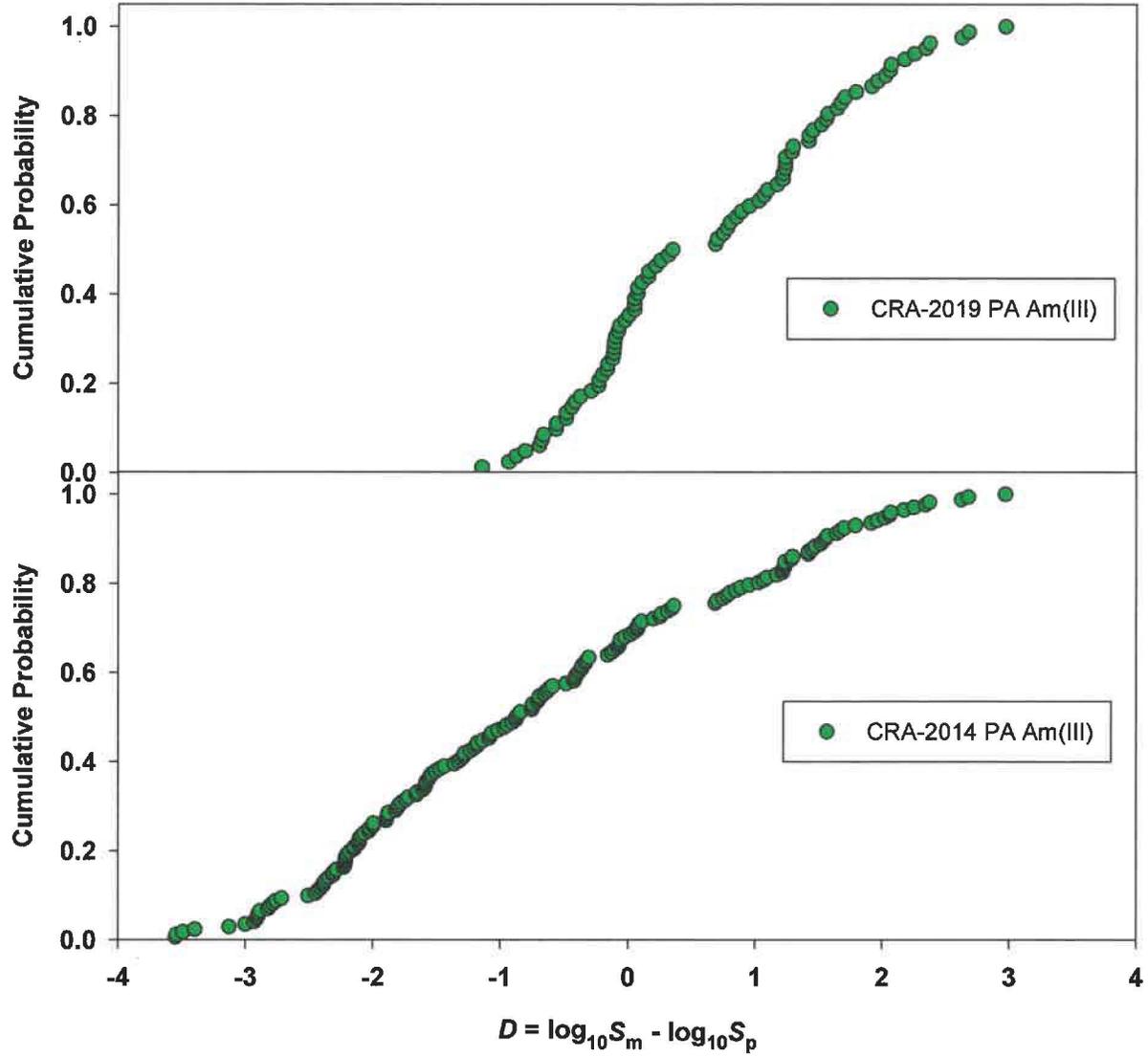
Figure 3 shows that the WIPP Am(III) model, using DATA0.FM4 over predicted ( $D < 0$ ) 28 of the 82 measured Nd(III) and Am(III) solubilities, and under predicted ( $D > 0$ ) 54 of the experimental solubilities. The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Am(III) solubilities are 0.651 and 0.516, respectively (Table 7), and the mean and median values of the sampled uncertainty factor are  $10^{0.651} = 4.480$  and  $10^{0.516} = 3.280$ . WIPP PA uses actinide solubilities that are the products of the baseline solubilities and the sampled uncertainty factors.

Table 9 provides values of the CDF for Nd(III) and Am(III) for CRA 2019 PA, which are also graphically presented in Figure 2. PA will use this CDF to define the solubility multiplier SOLVAR for the oxidation-state III model, material SOLMOD3 in the WIPP PA Parameter Database for the calculations for CRA 2019 PA. Brush et al. (2005) described the PA implementation of these results in detail.

Appendix B provides a breakdown of D values by author and study and the descriptive statistics for each data set.



**Figure 3. Histograms for CRA-2019 (top) and CRA-2014 (bottom) of the difference  $D = \log_{10} S_m - \log_{10} S_p$  for Nd(III) and Am(III).**



**Figure 4. CDF plots for CRA-2019 (top) and CRA-2014 (bottom) of the difference  $D = \log_{10} S_m - \log_{10} S_p$  for Nd(III) and Am(III).**

**Table 8. Statistical Properties of the Distribution of  $D = \log_{10}(S_m) - \log_{10}(S_p)$  for Nd(III) and Am(III) in response to EPA's selected data sets for CRA 2019 PA compared with the CRA-2014 PA data (Brush and Domski, 2013b).**

Analysis	Mean	Median	Standard Deviation	Minimum	Maximum
CRA-2014 PA	-0.678	-0.866	1.57	-3.55	2.97
CRA-2019 PA	0.651	0.516	1.01	-1.14	2.97

**Table 9. CDF of the Differences between the Measured and Predicted Nd(III) and Am(III) Solubilities for CRA 2019 PA.**  
**This CDF Defines the Solubility Multiplier SOLVAR for the Oxidation-State III Model Material SOLMOD3 in the WIPP PA Parameter Database.**

D	$P(D < X)$ for CRA 2019 PA
-1.142510	0.01220
-0.932358	0.02439
-0.875954	0.03659
-0.805676	0.04878
-0.694681	0.06098
-0.677855	0.07317
-0.665805	0.08537
-0.566530	0.09756
-0.562972	0.1098
-0.487951	0.1220
-0.486971	0.1341
-0.442671	0.1463
-0.417617	0.1585
-0.376826	0.1707
-0.291642	0.1829
-0.232110	0.1951
-0.228885	0.2073
-0.202533	0.2195
-0.166077	0.2317
-0.160532	0.2439
-0.122178	0.2561
-0.114771	0.2683
-0.112331	0.2805
-0.108348	0.2927
-0.100004	0.3049
-0.079188	0.3171
-0.069364	0.3293
-0.025201	0.3415
0.007030	0.3537
0.048993	0.3659
0.049039	0.3780

Table 9 continued on next page

**Table 9. CDF of the Differences between the Measured and Predicted Nd(III) and Am(III) Solubilities for CRA 2019 PA. (Cont.)**

D	$P(D < X)$ for CRA 2019 PA
0.049806	0.3902
0.074136	0.4024
0.074136	0.4146
0.106091	0.4268
0.155677	0.4390
0.160394	0.4512
0.215051	0.4634
0.254326	0.4756
0.313025	0.4878
0.346357	0.5000
0.685282	0.5122
0.701582	0.5244
0.749390	0.5366
0.778205	0.5488
0.802090	0.5610
0.850272	0.5732
0.889953	0.5854
0.952970	0.5976
1.028720	0.6098
1.068567	0.6220
1.095435	0.6341
1.170804	0.6463
1.215464	0.6585
1.217682	0.6707
1.232646	0.6829
1.236077	0.6951
1.236170	0.7073
1.285946	0.7195
1.295231	0.7317
1.418860	0.7439
1.419674	0.7561
1.451948	0.7683
1.518970	0.7805

Table 9 continued on next page

**Table 9. CDF of the Differences between the Measured and Predicted Nd(III) and Am(III) Solubilities for CRA 2019 PA. (Cont.)**

D	$P(D < X)$ for CRA 2019 PA
1.557256	0.7927
1.570299	0.8049
1.643798	0.8171
1.672233	0.8293
1.703691	0.8415
1.792566	0.8537
1.916789	0.8659
1.963935	0.8780
2.023033	0.8902
2.059652	0.9024
2.068615	0.9146
2.175439	0.9268
2.251789	0.9390
2.343682	0.9512
2.372998	0.9634
2.627935	0.9756
2.683488	0.9878
2.971474	1.0000

## 4 CONCLUSIONS

This revised analysis report provides the results of the new uncertainty analysis of the solubilities of Th(IV) and Am(III). SNL PA personnel will use the results of this uncertainty analysis for the calculations in response to EPA's selected data sets for CRA 2019 PA.

We used EQ3/6, Version 8.0a, and the Pitzer thermodynamic database DATA0.FM4 (Domski 2019), for this analysis. We carried out this analysis under Task 6 of AP-153, Rev. 1, the current analysis plan (AP) for WIPP near-field geochemical process modeling.

Our Th(IV) comparison included a total of 44 measured and predicted solubilities. The WIPP Th(IV) model over-predicted 23, and under-predicted 21 of the measured solubilities, respectively, in the current analysis. The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Th(IV) solubilities are -0.0114 and -0.090, respectively (Table 5). The mean and median values of the sampled uncertainty factor are  $10^{0.011} = 0.974$  and  $10^{-0.090} = 0.813$ . (The PA codes use actinide solubilities that are the products of the baseline solubilities and the sampled uncertainty factors).

Our Nd(III) and Am(III) comparison included a total of 82 measured and predicted solubilities. The WIPP Am(III) model under-predicted the majority of the measured solubilities, 54 of 82, and over predicted 28 of 82. The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Am(III) solubilities are 0.651 and 0.516, respectively (Table 8), and the mean and median values of the sampled uncertainty factor are  $10^{0.651} = 4.480$  and  $10^{0.516} = 3.280$ .

Tables 6 and 9 provide the CDFs of the uncertainties for the predicted Th(IV) and Am(III) solubilities in response to EPA's selected data sets for CRA 2019 PA.

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## Appendix A. Th(IV) Uncertainty Outputs

Table A.1

Author/Study	EQ3/6 Filename	D
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	15Fel91.3o	-0.520
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	23Fel91.3o	0.680
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	24Fel91.3o	0.690
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	25Fel91.3o	0.750
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	26Fel91.3o	-0.720
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	27Fel91.3o	-0.800
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	28Fel91.3o	-0.080
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	29Fel91.3o	-0.540
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	30Fel91.3o	-0.710
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	31Fel91.3o	-0.330
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	39Fel91.3o	0.270
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	40Fel91.3o	0.460
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	41Fel91.3o	0.160
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	42Fel91.3o	0.030
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	43Fel91.3o	-0.610
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	44Fel91.3o	-0.610
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	45Fel91.3o	0.290
Felmy et al. (1991), 3.0 M NaCl, 8.0 < pcH < 11.2	46Fel91.3o	-0.560
Altmaier et al. (2004), Fig 2 NaCl 5 M	01Alt04.3o	-1.489
Altmaier et al. (2004), Fig 2 NaCl 5 M	02Alt04.3o	-0.404
Altmaier et al. (2004), Fig 2 NaCl 5 M	03Alt04.3o	-0.947
Altmaier et al. (2004), Fig 2 NaCl 5 M	04Alt04.3o	-2.010
Altmaier et al. (2004), Fig 2 NaCl 5 M	05Alt04.3o	-1.016
Altmaier et al. (2004), Fig 2 MgCl2 2.5 M	06Alt04.3o	-0.555
Altmaier et al. (2004), Fig 2 MgCl2 2.5 M	07Alt04.3o	-0.419
Altmaier et al. (2004), Fig 2 MgCl2 2.5 M	08Alt04.3o	-0.646
Altmaier et al. (2004), Fig 2 MgCl2 2.5 M	09Alt04.3o	-0.465
Altmaier et al. (2004), Fig 2 MgCl2 2.5 M	10Alt04.3o	-1.303
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	01ALT06.3o	0.734
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	02ALT06.3o	0.787
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	03ALT06.3o	0.789
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	04ALT06.3o	0.864
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	05ALT06.3o	0.774
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	06ALT06.3o	0.823
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	07ALT06.3o	0.809
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	08ALT06.3o	0.890
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	09ALT06.3o	1.253
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	10ALT06.3o	1.295
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	11ALT06.3o	1.427
Altmaier et al. (2006), 4 M NaHCO3 - Na2CO3 - NaCl .02 M TIC	12ALT06.3o	1.157
Altmaier et al. (2008), 5.26 M CaCl2 Figure 5	01ALT08.3o	-0.100
Altmaier et al. (2008), 5.26 M CaCl2 Figure 5	02ALT08.3o	-0.561
Altmaier et al. (2008), 5.26 M CaCl2 Figure 5	03ALT08.3o	-0.425
Altmaier et al. (2008), 5.26 M CaCl2 Figure 5	04ALT08.3o	0.392

Table A.2

Study	Max	Min	Mean	Median	n
Felmy et al (1991)	0.75	-0.80	-0.12	-0.21	18
Altmaier et al. (2004)	-0.40	-2.01	-0.93	-0.80	10
Altmaier et al. (2006)	1.43	0.73	0.97	0.84	12
Altmaier et al. (2008)	0.39	-0.56	-0.17	-0.26	4

## Appendix B. Am(III) Uncertainty Outputs

Table B.1.

Author	EQ3/6 Filename	D
Neck et al. (2009), 1-M CaCl <sub>2</sub>	15NeckCa.3o	2.97
Neck et al. (2009), 1-M CaCl <sub>2</sub>	16NeckCa.3o	2.34
Neck et al. (2009), 1-M CaCl <sub>2</sub>	17NeckCa.3o	2.68
Neck et al. (2009), 1-M CaCl <sub>2</sub>	18NeckCa.3o	1.96
Neck et al. (2009), 1-M CaCl <sub>2</sub>	19NeckCa.3o	2.18
Neck et al. (2009), 1-M CaCl <sub>2</sub>	20NeckCa.3o	1.64
Neck et al. (2009), 1-M CaCl <sub>2</sub>	21NeckCa.3o	1.79
Neck et al. (2009), 1-M CaCl <sub>2</sub>	22NeckCa.3o	1.42
Neck et al. (2009), 1-M CaCl <sub>2</sub>	23NeckCa.3o	1.42
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	24NeckCa.3o	1.70
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	25NeckCa.3o	2.06
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	26NeckCa.3o	2.07
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	27NeckCa.3o	1.30
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	28NeckCa.3o	1.23
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	29NeckCa.3o	0.69
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	30NeckCa.3o	1.45
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	31NeckCa.3o	0.95
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	32NeckCa.3o	0.07
Neck et al. (2009), 2.5-M CaCl <sub>2</sub>	33NeckCa.3o	0.07
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	34NeckCa.3o	0.78
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	35NeckCa.3o	1.10
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	36NeckCa.3o	1.22
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	37NeckCa.3o	-0.08
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	38NeckCa.3o	0.31
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	39NeckCa.3o	0.05
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	40NeckCa.3o	0.35
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	41NeckCa.3o	-0.38
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	42NeckCa.3o	0.11
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	43NeckCa.3o	-0.07
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	44NeckCa.3o	-0.16
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	45NeckCa.3o	-0.49
Neck et al. (2009), 3.5-M CaCl <sub>2</sub>	46NeckCa.3o	-0.69
Neck et al. (2009), 1-M MgCl <sub>2</sub>	17NeckMg.3o	2.63
Neck et al. (2009), 1-M MgCl <sub>2</sub>	18NeckMg.3o	2.37
Neck et al. (2009), 1-M MgCl <sub>2</sub>	19NeckMg.3o	2.25
Neck et al. (2009), 1-M MgCl <sub>2</sub>	20NeckMg.3o	2.02
Neck et al. (2009), 1-M MgCl <sub>2</sub>	21NeckMg.3o	1.92
Neck et al. (2009), 2.5-M MgCl <sub>2</sub>	27NeckMg.3o	1.29
Neck et al. (2009), 2.5-M MgCl <sub>2</sub>	28NeckMg.3o	1.52
Neck et al. (2009), 2.5-M MgCl <sub>2</sub>	29NeckMg.3o	1.56
Neck et al. (2009), 5-M MgCl <sub>2</sub>	35NeckMg.3o	1.57
Neck et al. (2009), 5-M MgCl <sub>2</sub>	36NeckMg.3o	1.03
Neck et al. (2009), 5-M MgCl <sub>2</sub>	37NeckMg.3o	1.07
Neck et al. (2009), 5-M MgCl <sub>2</sub>	38NeckMg.3o	1.17
Neck et al. (2009), 5-M MgCl <sub>2</sub>	39NeckMg.3o	1.24

<b>Author</b>	<b>EQ3/6 Filename</b>	<b>D</b>
Neck et al. (2009), 5-M MgCl2	40NeckMg.3o	0.70
Neck et al. (2009), 5-M MgCl2	41NeckMg.3o	0.89
Neck et al. (2009), 5-M MgCl2	42NeckMg.3o	0.85
Neck et al. (2009), 5-M NaCl	095NeckNa.3o	1.67
Neck et al. (2009), 5-M NaCl	096NeckNa.3o	1.24
Neck et al. (2009), 5-M NaCl	097NeckNa.3o	0.75
Neck et al. (2009), 5-M NaCl	098NeckNa.3o	-0.11
Neck et al. (2009), 5-M NaCl	099NeckNa.3o	0.80
Neck et al. (2009), 5-M NaCl	100NeckNa.3o	0.25
Neck et al. (2009), 5-M NaCl	101NeckNa.3o	-0.03
Neck et al. (2009), 5-M NaCl	102NeckNa.3o	1.22
Rao et al. (1999)	01rao99.6o	0.05
Rao et al. (1999)	02rao99.6o	-0.57
Rao et al. (1999)	03rao99.6o	-0.23
Rao et al. (1999)	04rao99.6o	-0.20
Rao et al. (1999)	05rao99.6o	-0.17
Rao et al. (1999)	06rao99.6o	-0.11
Rao et al. (1999)	07rao99.6o	0.16
Rao et al. (1999)	08rao99.6o	0.01
Rao et al. (1999)	09rao99.6o	-0.11
Rao et al. (1999)	10rao99.6o	0.16
Rao et al. (1999)	11rao99.6o	-0.23
Rao et al. (1999)	12rao99.6o	-0.29
Rao et al. (1999)	13rao99.6o	-0.56
Rao et al. (1999)	14rao99.6o	-0.68
Rao et al. (1999)	15rao99.6o	-0.81
Runde & Kim (1994)	01Run95.3o	-0.12
Runde & Kim (1994)	02Run95.3o	-1.14
Runde & Kim (1994)	03Run95.3o	-0.10
Runde & Kim (1994)	04Run95.3o	0.22
Runde & Kim (1994)	05Run95.3o	-0.44
Runde & Kim (1994)	06Run95.3o	-0.67
Runde & Kim (1994)	07Run95.3o	-0.49
Runde & Kim (1994)	08Run95.3o	-0.93
Runde & Kim (1994)	09Run95.3o	-0.42
Runde & Kim (1994)	10Run95.3o	-0.88
Runde & Kim (1994)	11Run95.3o	0.05

Table B.2.

<b>Study</b>	<b>Max</b>	<b>Min</b>	<b>Mean</b>	<b>Median</b>	<b>n</b>
Neck et al. (2009)	2.971	-0.695	1.106	1.217	56
Rao et al. (1999)	0.160	-0.806	-0.239	-0.203	15
Runde & Kim (1994)	0.215	-1.143	-0.448	-0.443	11