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Sandia National Laboratories
Waste Isolation Pilot Plant

Uncertainty Analysis of Actinide Solubilities for the Replacement Panels Planned Change Request (RPPCR) Performance Assessment and the 2024 Compliance Recertification Application (CRA-2024)

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

Author:	signature on file Paul S. Domski, Org. 08882	6/12/2023 Date
Technical Reviewer:	Sungtae Kim Sungtae Kim, Org. 08881	6/12/2023 Date
QA Reviewer:	Shelly R. Nielsen Shelly R. Nielsen, Org. 08880	6-12-2023 Date
Management Reviewer:	Chammi Miller Chammi Miller, Org. 08882	06/12/23 Date

WIPP:4.4.1.1.1:TD:QA-L:RECERT:555726

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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 for the baseline solubility values (Domski, 2023a) in the WIPP Performance Assessment (PA) Parameter Database for the Replacement Panels Planned Change Request (RPPCR). 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, the "D" value, between the $\log_{10}$ of the measured data and the $\log_{10}$ of the data simulated using the WIPP model. The D values are used to construct empirical cumulative distribution functions (CDF) which are then sampled by the PA code. Multiple studies for both the An(III) [1]and the An(IV) model were included in this uncertainty analysis, and were used to build the oxidation-state specific uncertainty distributions.

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 1×10^{-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). See Section 2.2.8 of AP-153, Revision 1 Brush et al. (2012) for a discussion of U(VI). GWB is a synthetic brine representative of intergranular Salado Formation (Fm.) brines at or near the stratigraphic horizon of the repository (Krumhansl et al., 1991; Snider, 2003). ERDA-6 (Popielak et al., 1983) is a synthetic brine representative of fluids in brine reservoirs in the Castile Fm., which underlies the Salado Fm.

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

This analysis report documents the actinide uncertainty distributions for the RPPCR/CRA-2024 . This uncertainty analysis differs from previous uncertainty analyses by use of the new WIPP database for EQ3/6, DATA0.FM6, and the application of a new set of literature screening criteria (Domski and Nemer, 2023) for choosing experimental studies.

The difference between the current uncertainty analysis and that documented in Knerr (2021) which addresses the Environmental Protection Agency (EPA) comment CC1-GEOCHEM-1 for the 2019 Deferred Performance Assessment Compliance Recertification Application (CRA-2019 DPA) is: 1) A new literature search was performed;2) new screening criteria (Domski and Nemer, 2023) were applied to the experimental studies used to compare the WIPP models; and 3) the use of a different WIPP database, DATA0.FM6 (Domski, 2023b).

The EQ3/6 software package, Version 8.0a, (Wolery and Jarek, 2003; Wolery, 2008; Wolery et al., 2010; Xiong, 2011) is used for this analysis. The updated WIPP thermodynamic database

[1] The term An(-) is the general term used for actinide with the value in parenthesis denoting its oxidation state.

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(DB) DATA0.FM6 (Domski, 2023b) was used for this analysis and is a deviation from AP-153, Revision 1(Brush et al., 2012) which specifies DATA0.FM1.

We conducted 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 that 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.

In this analysis report the U.S. Department of Energy (DOE) have addressed issues 9 and 10 of the U.S. Environmental Protection Agency (EPA) (Peake, 2022). These issues are further discussed in the following sections.

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Table 1. Abbreviations, Acronyms, Initialisms, etc.

Abbreviation, Acronym, or Initialism	Definition
Am, Am(III)	americium, americium in the +III oxidation state
am	amorphous
An(-)	general term used for actinide with the value in parenthesis denoting its oxidation state
AP	analysis plan
aq	aqueous (dissolved in an aqueous solution)
atm	atmosphere(s)
C, c	carbon, crystalline
Ca, Ca^{2+}	calcium, calcium ion
CAP	(SNL/WIPP QA) corrective action plan
CCA	Compliance Certification Application
CDF	cumulative distribution function
citrate	$(CH_2COO)_2C(OH)(COO)^{3-}$ or $(CH_2CO_2)_2C(OH)(CO_2)^{3-}$
Cl, Cl^-	chloride, chloride ion
ClO_4, ClO_4^-	perchlorate, perchlorate ion
Cm, Cm(III)	curium, curium in the +III oxidation state
CMS	(Sandia/WIPP software) Configuration Management System
CO_3, CO_3^{2-}	carbonate, carbonate ion
col	colloidal (suspended in an aqueous or other solution)
CPR	cellulosic, plastic and rubber (materials)
cr	crystalline

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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
CRA-2019 DPA	The fourth WIPP Compliance Recertification Application, Deferred Performance Assessment submitted to the EPA in March 2019
RPPCR	Replacement Panels Planned Change Request
D	the difference(s) between logs (base 10) of the measured and predicted actinide solubilities (Sm and Sp)
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
fCO_2	fugacity (similar to the partial pressure) of CO_2
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
I	ionic strength, defined by $I = ½ \times \Sigma_i(M_i \times z_i^2)$, in which M_i and z_i are the molarity and charge of species i

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

Abbreviation, Acronym, or Initialism	Definition
I/O	(EQ3/6) input and output files
K, K^+	potassium, potassium ion
LIB	Library
log	the common logarithm or logarithm (base 10)
M	molar, $mol \cdot dm^{-3}$
m	molal, $mol \cdot kg^{-1}$
mcr	microcrystalline
Mg, Mg^{2+}	magnesium, magnesium ion
mM	millimolar
N	bin number (see Section 2 for explanation)
Na, Na^+	sodium, sodium ion
Nd, Nd(III)	neodymium, neodymium in the +III oxidation state
NO_3, NO_3^-	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^-	hydroxide, hydroxide ion
P	probability
PA	performance assessment, may refer to both the PA code, and/or the WIPP PA group
PABC	(WIPP) Performance Assessment Baseline Calculations, carried out in 2005 and 2009
pcH	the negative log (base 10) of the molar concentration of H^+

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

Abbreviation, Acronym, or Initialism	Definition
p_{CO_2}	partial pressure (similar to the fugacity) of CO_2
pmH	the negative log (base 10) of the molal concentration of H^+
ppm	parts per million
pH	the negative log (base 10) of the activity of 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
SO_4, 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
µm	micrometer
whewellite	$CaC_2O_4 \cdot H_2O$
WIPP	Waste Isolation Pilot Plant
XRD	X-ray diffraction
Zr, Zr(IV)	zirconium, zirconium in the +IV oxidation state
μ^0, μ^0/RT	standard chemical potential, dimensionless standard chemical potential

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2 METHODS

The method employed in the actinide uncertainty analysis has gone largely unchanged since the Compliance Certification Application (CCA) PA. The core of the method is the creation of empirical 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 CDFs are sampled by the PA code and used as multiplier for the baseline solubility values for performance assessment.

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 (see below).
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.FM6 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 prepare empirical cumulative distribution functions.
7. Document all of the above steps in an analysis report (this report).
8. Provide a memo(s) for 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 RPPCR/CRA-2024 .

A new literature search was initiated for the RPPCR and CRA-2024. This literature search was broader based than previous searches in accordance with G1 and G6 (see below) (Domski and Nemer, 2023). The literature search for the RPPCR and CRA-2024 included the period up to October 9, 2022, which is 3-months before the parameter cutoff date for RPPCR and CRA-2024 of January 9, 2023 (see G1 below). The starting date for the literature search was not specified as criterion G1 (see below) does not specify a start date. This broad-based screening was facilitated by the use of Python scripts to search two databases: 1) Web of Science, to obtain journal articles and conference papers (https://app.webofknowledge.com/author/search), and 2) OSTI, to obtain US government reports (i.e., the G2 criterion) (https://www.osti.gov). The Python scripts and their application are explained in Nemer et al. (2023). The updated literature screening criteria G1 (Domski and Nemer, 2023) combined with this analysis report directly address and resolve Issue 9 of the EPA (Peake, 2022) by setting and implementing a cutoff date for the eligible literature inclusion date.

The implementation of new screening criteria also required re-screening literature that was previously screened to determine if data previously screened out would be screened in under the new criteria, and if data previously screened in might be screened out under the new criteria.

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The literature screening that was completed for RPPCR and CRA-2024 identified eight potential experimental studies for the An(IV) actinides, however, a detailed examination of these studies revealed that none of them met all of the screening criteria (see below), and as such none were included in the current uncertainty analysis. Re-screening of previously screened literature using the new criteria (Domski and Nemer, 2023) identified an additional two data points from the Altmaier et al. (2004) reference. These two data points were included with the five data points that resulted from the application of the new G10 criterion as reported by Domski and Nemer (2023) for the An(IV) uncertainty distribution.

The An(III) literature screening identified one potential solubility study which required in-depth screening to determine its eligibility for the RPPCR/CRA-2024 uncertainty analysis. Unfortunately it did not meet the screening criteria, and is discussed in detail in the An(III) section (see below). Literature previously screened for past uncertainty analyses was re-screened for the RPPCR/CRA-2024 , and did not identify any additional data that could be used in the RPPCR/CRA-2024 An(III) uncertainty analysis. Therefore, the RPPCR/CRA-2024 An(III) uncertainty analysis includes essentially the same data as that in Domski and Nemer (2023), but some minor changes due to the more detailed re-screening.

The WIPP screening criteria are used to determine which literature/experimental studies are included in the actinide uncertainty analysis, and are documented in Domski and Nemer (2023). The criteria were presented to, and accepted by the EPA at the September 25, 2022 technical exchange meeting. From Domski and Nemer (2023):

Applicability:
These screening criteria are applicable to the selection of experimental data from the scientific literature for the purpose of validating the WIPP actinide solubility model under conditions modeled in PA. In this context, it is necessary to differentiate the two types of data typically considered in modeling, training data and test data. Training Data are those data used to fit free parameters to a given model, e.g., laboratory measurements of solubility which are used to fit the log K of a specific mineral phase. These screening criteria are ***not*** *intended for use in selecting Training Data, or to fit free parameters to the WIPP actinide solubility model for inclusion in the thermodynamic database. These screening criteria are applicable only to the selection of Test Data. Test Data are those data used to validate the WIPP actinide solubility model. For example, solubility measurements taken from the literature are compared to those predicted by the model parameterized with the log K fitted with the Training Data. The Test Data (S_m – **S**olubility **m**easured) and the model predictions (S_s – **S**olubility **s**imulated) are used to calculate the difference, or D values as $D = \log_{10}(S_m) - \log_{10}(S_s)$. The D values are then used to create an empirical distribution of model-prediction errors, which are sampled in PA and added to the $\log_{10}$ of the baseline solubility values.*

The new screening criteria, G-1 through G-10, are listed below:
G1 – Time Frame: *Preference given to data from studies after January 1990 with the emphasis on the period between the closure date of the most recent recertification cycle and the current recertification cycle. The closure date for the literature search is set to 3-months before the required parameter feed to performance assessment for the current recertification cycle. Studies prior to 1990 may be acceptable, provided the experimental and analytical methods are provided and defensible, and satisfies the remainder of the criteria listed herein.*

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G2 – Sources: *Include data from studies in peer-reviewed journals unpublished undergraduate/graduate theses or dissertations, national and international laboratories, and organizations (e.g., officially released reports from government and educational national and international laboratories.), as well as studies originating from industry.*
G3 – Type of Data: *Include only those studies that provide solubility data. Exclude studies that do not provide solubility data (e.g., studies of corrosion, leaching, sorption, or transport).*
G4 – Solvent and Composition: *Include only data from studies in which water was the solvent and the solution composition characterized; exclude studies in which no aqueous phase was present (e.g., solids, molten metal or salts, or pure organic liquids).*
G5 – Temperature and Pressure: *Include only data obtained from studies that were performed at atmospheric pressures and temperatures of 20 - 30°C.*
G6 – Use of Oxidation-State Analogs*: Include data from studies that contain actinides currently in the WIPP model, or their appropriate oxidation-state analogs.*
G7 – Phase Characterization and Thermodynamic Equilibrium: *Include data from studies with a well-characterized solubility-controlling solid (both the starting and final solid phases) for which solubility parameters are in the WIPP model. Characterization of the final phase(s) may be complicated due to the presence of low-crystallinity hydrated phases, which may not be easily identified. Methods used for phase identification must be well documented and appropriate for the experimental conditions. The solution separation technique (i.e., dissolved from colloidal) must be well documented and appropriate for the experimental conditions. The experimental procedure should discuss solid-phase synthesis procedures, including descriptions of precursors (solids and solutions); and whether equilibrium (including meta-stability) was obtained. Determination of equilibrium typically includes demonstration of stable concentrations and/or chemical conditions as a function of time.*
G8 – Applicability*: Include data from studies in which the participating chemical components (i.e., aqueous species and solid phases that may affect actinide solubility) are included in the WIPP model. Exceptions include the use of non-participating cations and/or anions, or additional known and characterized dissolved elements that have been demonstrated to not strongly complex or effect the solubility of the actinide(s) of interest.*
G9 – Repeatability and Reproducibility: *Include only data from studies in which: 1) The investigators provided a sufficient description of their experiments to allow the work to be repeated by a qualified investigator; and 2) sufficient experimental data are provided to allow computer simulation of the experiments.*
G10 – WIPP-Relevant Conditions: *Include data from experimental studies carried out under conditions [e.g., pcH, ionic strength, and total inorganic carbon (TIC)] similar to those expected for WIPP, as designated by the Chemical Conditions Conceptual Model.*
Experimental studies/data included in the WIPP actinide uncertainty analysis must meet the following conditions:

Parameter ranges for G10 literature screening criteria.

G10 Conditions	
Ionic Strength (m)	$5.0 \leq I \leq 9.5$ m[2]

[2] The WIPP brines are predominantly sodium – chloride type brines but include an array of other dissolved ionic species (e.g., K^+, Mg^{2+}, Ca^{2+}, SO_4^{2-}, HCO_3^-, $B(OH)_4^-$, and Br^-), and as such the G10 ionic strength range mainly reflects the Na - Cl predominance. However, in the scientific literature actinide/lanthanide solubility studies are

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G10 Conditions	
	$0.0 \leq Mg \leq 1.6$ M
	$0.0 \leq Ca \leq 0.5$ M
pcH	$8.5 \leq pcH \leq 10.7$
Total Inorganic Carbon	$TIC = 0.0 - 2.0 \times 10^{-2}$ M

EQ3/6 input files used for the CRA-2019 DPA and to address EPA comment CC1-GEOCHEM-1 (Knerr, 2012) were used for the RPPCR/CRA-2024 uncertainty analysis. All of the laboratory studies, with the exception of one, Rao et al. (1999), used a form of Am-hydroxide as the solubility controlling phase, and were simulated with EQ3NR. For Rao et al. (1999), the solubility-controlling phase was $NaAm(CO_3)_2 \cdot 6H_2O$(cr), and it was necessary to use EQ6 to include CO_2(g) as a fixed-fugacity gas. 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 archived to the SNL/WIPP Records Center.

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 (archived in the records package). The results for the An(III) and An(IV) are compiled in the spreadsheets *An(III)_RPPCR_Uncertainty.xlsx* and *AN(IV)_RPPCR_Uncertainty.xlsx*, respectively, both of which are archived in the records package. Each of the laboratory studies is saved on separate tabs that are clearly labeled. These two spreadsheets also contain the probability distributions and CDF and histogram plots.

The frequency distributions were generated from 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. Instead, a constructed cumulative-distribution function (CDF) was defined from the raw data rather than the binned histogram data.

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

typically performed in single electrolyte solutions, mainly NaCl, $MgCl_2$, and $CaCl_2$, therefore, it is necessary to set WIPP appropriate upper bounds on dissolved Mg and Ca in this criterion. Note that the Mg and Ca limits are applicable to both single and multi-electrolyte solutions, however, the lower limit of ionic strength (5.0 m) is not applicable to Mg and Ca single-electrolyte systems.

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model); positive values of D indicate that the model predicted a solubility less than the measured value (underprediction). The Excel files "*An(III)_RPPCR_Uncertainty.xlsx*" and "*AN(IV)_RPPCR_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)_RPPCR_Uncertainty.xlsx*" and "*AN(IV)_RPPCR_Uncertainty.xlsx*" and all of the other spreadsheets to the archived file RPPCR-CRA2024_An-Uncert.ZIP, which is stored at "/data/cvs/CVSLIB/WIPP_EXTERNAL/ap153_files/RPPCR".

In this analysis, we use DATA0.FM6 (Domski, 2023b) to perform calculations to construct the uncertainty distributions based on the data sets meeting the literature screening criteria of Domski and Nemer (2023). We follow the rounding rules of Gamsjäger et al. (2012) for the numbers in the CDFs generated in this analysis.

Table 2 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 2. 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.FM6	In zip file Uncertainty_RPPCR.ZIP, /data/cvs/CVSLIB/WIPP_EXTERNAL/ap153_files/RPPCR
Excel macro GetEQData_v101f.xls	In zip file Uncertainty_RPPCR.ZIP, /data/cvs/CVSLIB/WIPP_EXTERNAL/ap153_files/RPPCR
EQ3/6 I/O files and Excel spreadsheets with input and extracted data	In zip file Uncertainty_RPPCR.ZIP, /data/cvs/CVSLIB/WIPP_EXTERNAL/ap153_files/RPPCR

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3 RESULTS

This section provides the results of the RPPCR actinide solubility uncertainty analysis for An(IV) (Subsection 3.1) and An(III) (Subsection 3.2). Brief descriptions of the literature that required close scrutiny to determine if they qualified for inclusion in the analysis are provided in Table 3 for the +IV actinides and Table 6 for the +III actinides. The implementation of the new G2 and G6 criteria of Domski and Nemer (2023) allowed a larger selection of literature to be included in the screening, both in terms of publication date, i.e., consideration of papers from before 1990 (G2), and in terms of oxidation state analogs (G6). For previous uncertainty analyses the An(III) data was selected from solubility studies for Am(III), Nd(III), and Cm(III), and the An(IV) data was selected from Th(IV) solubility studies. For the current study, in addition to the three elements mentioned above Pu(III) studies were also considered for inclusion in the An(III) uncertainty distribution. Likewise for An(IV), Pu(IV) and U(IV) studies were considered. The results of the broader screening did not yield any additional studies for the +IV oxidation state, though two additional data points from a previously screened study were admitted. For the +III oxidation state one additional study was found that required close scrutiny under the current criteria to determine its eligibility for inclusion in the uncertainty analysis. The results of the expanded study are summarized in Nemer et al. (2023).

3.1 An(IV) Solubility Model

Literature Screening

A literature search was performed for the RPPCR to identify experimental studies for inclusion in the An(IV) uncertainty analysis. The literature search identified eight potential studies, however, none of these studies met all the screening criteria, and no data were identified for inclusion in the An(IV) uncertainty distribution. See below for a brief overview of each study.

Domski and Nemer (2023) applied the updated screening criterion G10 to references that were screened in for the An(IV) CRA-2019 DPA uncertainty analysis. This resulted in the exclusion of 39 of 44 experimental data points from the An(IV) uncertainty distribution. For the RPPCR a comprehensive re-screening of the experimental studies which had previously been screened in or out in previous uncertainty analyses (e.g., Brush and Domski, 2013; Domski, 2019a; Knerr 2021) was undertaken by application of the screening criteria of Domski and Nemer (2023). This resulted in the identification of two additional data points from the study of Altmaier et al. (2004) (see Table 3). Therefore, the number of data points included in the RPPCR/CRA-2024 An(IV) actinide uncertainty analysis is seven.

An(IV) Studies Considered for the RPPCR/CRA-2024 Actinide Uncertainty Analysis

Eight potential An(IV) experimental solubility studies were identified for possible inclusion in the An(IV) actinide uncertainty analysis for the RPPCR/CRA-2024 . These studies are discussed below.

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Adam et al. (2021) examined the impact of cement additives adipic acid, methyl acrylate, citric acid, melamine, ethylene glycol, phthalic acid and gluconic acid on the solubility of Nd(III), Th(IV), and U(VI). A total of 36 results met the G10 condition for pmH ≈ 9, 18 results for 5.61 m NaCl, and 18 results for 1.03 m $MgCl_2$ for all additives (G8) with the exception of gluconate, as gluconate was the only additive observed to have any impact on solubility besides citrate which is included in the An(IV) model. However, these results did not meet the G7 criterion, the solubility-controlling phase differs from that in the WIPP Th(IV) model, there was no post-test solids characterization, and the post-test phase-separation method (ultrafiltration) was different than that employed in the studies used to parameterize the Th(IV) WIPP model.

Altmaier et al. (2008b) studied the solubility of $PuO_2.xH_2O$ in solutions of $MgCl_2$ and $CaCl_2$ at pcH values of ~9 – 12. The data collected for the 0.25 M MgCl2 solutions were the only data meeting the G10 ionic strength and pcH conditions. However, there was no post-test solids characterization and the samples were processed using ultrafiltration, both conditions in conflict with the G7 criterion.

Dumpala et al. (2021) examined the fractions of colloidal versus dissolved Th(IV) in sodium perchlorate solutions of varying ionic strengths (0 – 3 M) and pH values ranging from 5 – 13. All of the experimental results were excluded because they did not meet the conditions of G7, the solubility-controlling phase was not clearly defined/characterized, G9 because apparently the pH was not measured during sampling, and the G10 ionic strength condition was not met.

Kiefer et al. (2022) investigated the effect of temperature, aging time, and pH on the structure and solubility of freshly precipitated ThO_2(am, hyd). All of the experiments in this study were performed in 0.1 M NaCl solutions at pH values of ~3.5 – 7. All results of this study were excluded based on G10 ionic strength and pH conditions.

Kobayashi et al. (2009) evaluated the solubility of Th(IV) in sodium perchlorate solutions up to 2.0 M with oxalic acid and malonic acid (0.02 and 0.1 M) at low pH of ~4 – 5 from oversaturation. A Th(IV) - oxalate phase was suspected but not identified, and no solid phase was observed in the malonic acid experiment. All data from this study was excluded based on the pH and ionic strength conditions of G10, and the lack of a solubility-controlling phase (G7).

Kobayashi et al. (2021) studied the solubility of PuO_2(am,hyd) and formation of Pu(IV) carbonate complexes in 0.1 – 5.0 M sodium nitrate solutions. The results of the study were screened out based on criterion G8 because nitrate is not included in the WIPP An(IV) model, and was suspected of influencing the solubility results.

Rai et al. (1990) examined the solubility of $UO_2{\cdot}xH_2O$ in dilute solutions (maximum ionic strength ~0.5 M) across a range of pH ~2 – 12. All results were excluded because they failed to meet the G10 ionic strength condition.

Tasi et al. (2018) studied the effect of α–D–isosaccharinic (ISA) acid on the solubility of PuO_2 (ncr, hyd). All of the data were excluded because α–D–isosaccharinic is not included in the WIPP An(IV) model (G8).

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Table 3 provides the studies and the rationale used for including or excluding the data from each in the RPPCR/CRA-2024 actinide uncertainty analysis.

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Table 3. 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 of Literature / Experimental Results
Adam et al. (2021)	(A) 2.64 m NaCl (B) 5.61 m NaCl (C) 1.03 m $MgCl_2$ (D) 3.87 m $MgCl_2$ (E) 1.02 m $CaCl_2$ (F) 3.91 m $CaCl_2$	(A) ~9, ~13 pmH (B) ~9, ~13 pmH (C) ~9 pmH (D) ~9 pmH (E) ~9, ~12 pmH (F) ~9, ~12 pmH	ThO_2(ncr, hyd) nrc = nanocrystalline hyd = hydrated	This paper examines the impact of cement additives adipic acid, methyl acrylate, citric acid, melamine, ethylene glycol, phthalic acid and gluconic acid on the solubility of Nd(III), Th(IV), and U(VI). A total of 36 results met the G10 ionic strength and pmH ≈ 9 conditions with 18 results for 5.61 m NaCl, and 18 results for 1.03 m $MgCl_2$. Data for all additives, with the exception of gluconate, met the G8 criterion. However, none of the results met the G7 criterion, the solubility-controlling phase differs from that in the WIPP Th(IV) model, there was no post-test solids characterization, and the post-test phase-separation method (ultrafiltration) was different than employed in the studies used to parameterize the Th(IV) WIPP model.
Altmaier et al. (2004)	(A) 0.5 M NaCl (B) 5 M NaCl (C) 0.25 M $MgCl_2$ (I = 0.75 M) (D) 2.5 M $MgCl_2$ (I = 7.5 M) (E) 4.5 M $MgCl_2$ (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_2(cr) & $ThO_n(OH)_{4-2n}$ xH_2O(am), which they referred to as "$Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am)"	Five 5.0 M NaCl uncentrifuged results with pcH ~10 accepted. Two 0.25 M $MgCl_2$ results with pcH ~9 accepted. Seven results satisfied criterion G10 and were also included in this analysis. Ultracentrifuged results were excluded because the post-test 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.
Altmaier et al. (2005)	(A) Mostly $NaHCO_3$-Na_2CO_3-NaCl-NaOH with I = 0.5 M (B) Some with	(A) pcH = 4.5-7.5 & 8.5-13.5 (B) pcH = 13	$Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am)	All results were below the G10 ionic strength limit. In addition, the $Th(OH)_y(CO_3)_z^{4-y-2z}$ complex(es) found to be important in some samples by Altmaier et al. (2005) are not in the WIPP Th(IV) model.

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	0.25-2 M Na_2CO_3 & 0.1 M NaOH			
Altmaier et al. (2006)	$NaHCO_3$-Na_2CO_3-NaCl with I = 0.1-4 M.	pcH = 8–11	$Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am)	All results below the G10 ionic strength limit.
Altmaier et al. (2008a)	(A) 0.2–5.26 m $CaCl_2$ (I = 0.6–15.78 m) (experiments with Th(IV)) (B) 2.40 m $Ca(ClO_4)_2$ (I = 7.20 m) (Th(IV))	pmH = 9.3-12.1 (all Th(IV))	Solutions with $CaCl_2$ < 1 M (entire pH range) or $CaCl_2$ > 1 M (pH < 11.5): $Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am). Solutions with $CaCl_2$ > 1 M (pH > 11.5): unidentified, possibly a Ca-Th solid(cr).	None of the experimental results meet the G7 criterion because the samples were the product of either ultrafiltration or ultracentrifugation. In addition the results fail to meet the ionic strength and pcH requirements of the G10 criterion.
Altmaier et al. (2008b)	(A) 0.25 $MgCl_2$ (B) 3.5 M $MgCl_2$ (C) 3.5 - 4 M $CaCl_2$	(A) pcH ~9 (B) pcH ~8.8 (C) pcH ~10 - 12	$PuO_2 \cdot xH_2O$	The data for the 0.25 M $MgCl_2$ experiments were closely scrutinized for inclusion in the analysis. However, because the samples were subject to ultrafiltration, which is inconsistent with the WIPP An(IV) model, and because there was no post-test analysis of the solubility-controlling phase (G7) these data were excluded. All other data screened out based on the ionic strength condition of G10.
Baston et al. (1996)	Nirex (0.02 M)	pH = 10-12	ThO_2(am)	All results were below the G10 ionic strength limit.
Borkowski et al. (2012)	(A) HCO_3^-/CO_3^{2-}-free & HCO_3^-/CO_3^{2-}-bearing GWB, diluted to 95% of its specified concentrations	(A) pcH = 6.8–10.13 (GWB: carbonate-free, or 10^{-2} or 10^{-3} M carbonate; & over- & undersaturation)	Solids precipitated in oversaturation runs not reported. "Th oxyhydroxide" (composition not reported) used in	Unfortunately none of the results from this study qualified for use in the uncertainty analysis because the post-test phase characterization (G7) was not documented.

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	(B) HCO_3^-/CO_3^{2-}-free & HCO_3^-/CO_3^{2-}-bearing ERDA-6, diluted to 95% of its specified concentrations	(B) pcH = 7.86–11.04 (ERDA-6: carbonate-free, or 10^{-2} or 10^{-3} M carbonate; & over- & undersaturation)	undersaturation runs	
Borkowski (2012)	Same as above	Same as above	Same as above	Same as above
Brendebach et al. (2007)	0.2-4.5 M $CaCl_2$ (I = 0.6–13.5 M) (experiments with Th(IV))	pcH ≈ 9.3-12.1 (all Th(IV)).	Solutions with $CaCl_2$ < 1 M (entire pH range) or $CaCl_2$ > 1 M (pH < 11.5): $Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am). Solutions with $CaCl_2$ > 1 M (pH > 11.5): unidentified, possibly a Ca-Th solid(cr).	The results of Brendebach et al. (2007) were superseded by those of Altmaier et al. (2008a) (see above). Therefore, the results of Brendebach et al. (2007) are excluded from this analysis for the same reasons that Altmaier et al. (2008a) were excluded.
Colàs et al. (2011)	0.5 M $NaClO_4$ with 10^{-6} to 10^{-1} M gluconate	pcH ≈ 9-13	Th hydroxide ($ThO_2 \cdot xH_2O$)	All results were excluded based on G8, gluconate is not in the WIPP Th(IV) model.
Dumpala et al. (2021)	0.0, 0.1, 1.0, and 3.0 M $NaClO_4$	Initial pH 5.0, 7.0, 9.0, 13.0	"neither completely $Th(OH)_4$ nor ThO_2, can bebcalled as amorphous hydrous oxyhydroxide $ThO_n(OH)_{4-2n} \cdot xH_2O$"	All results excluded because the G10 pH and ionic strength condition was not met, and the pH was not measured at sampling times.
Fellhauer et al. (2010)	1.02–5.26 m $CaCl_2$ (I = 3.06–15.78 m) (experiments with Np(IV) and Pu(IV))	pcH ≈ 11-12 (Np(IV)) pcH ≈ 10-12 (Pu(IV))	$NpO_2 \cdot xH_2O$(am) or $PuO_2 \cdot xH_2O$(am)	All experimental results excluded because they failed to meet the G10 ionic strength condition.

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
Felipe-Sotelo et al. (2015)	$Ca(OH)_2$ saturated solutions	pH ≈ 12	ThO_2	Felipe-Sotelo et al. (2015) investigated the effect of organic complexants, EDTA and diethylenetriamine pentaacetate (DTPA), at 95% $Ca(OH)_2$ saturation and high pH conditions (i.e., pH ≈ 12) on ThO2 solubility. The experimental procedures used are not clearly described in the text (G9). The authors do not provide the solution chemistry for the solubility experiments, and neither is the ionic strength of these experiments provided in the paper (G4). All results were excluded because of the high pH conditions (G10), the unknown solution chemistry and ionic strength (G4), and the lack of information regarding the experimental procedures (G9).
Felipe-Sotelo et al. (2017)	Experiments carried out in two solutions: 1) NaOH w/ 95% saturated Ca(OH)2; and 2) NRVB equilibrated water. Ionic strength is not specified. NRVB is Nirex Reference Vault Backfill	pH 11.8 – 12.8	ThO_2(am)	This study examined thorium and uranium solubility in four different high-pH (~12) solutions. Two of the solutions were at low ionic strength values, a sodium hydroxide solution with Na+ = 460 mg/L, and $Ca(OH)_2$ solution with Ca^{2+} = 380 mg/L. The other solutions contained lithium and cesium in high concentrations, and neither of these elements are included in the WIPP model. All results were excluded because of the high pH conditions (G10), the unknown ionic strength, and the presence of elements not in the WIPP model (G8).
Felmy et al. (1991)	(A) 0.1 M $NaClO_4$ (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)	All experimental results were excluded because they failed to meet the G10 ionic strength condition.
Felmy et al. (2006)	(A) 0.5 M $NaNO_3$ &	(A) pcH = 5.2–12.2	Th(IV) hydrous oxide	All results were excluded based on G8, because NO_3^- is

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	0.01 M citrate (B) 3.0 M $NaNO_3$ & 0.01 M citrate (C) 6.0 M $NaNO_3$ & 0.01 M citrate (D) 0.5 M $NaNO_3$ & 10^{-4} to 10^{-2} M citrate (E) 0.5 M $NaNO_3$ & 10^{-4} to 10^{-2} M citrate	(B) pcH = 5.0–11.8 (C) pcH = 4.7–12.1 (D) pcH = 6-8 (E) pcH = 9.5-10.1	(ThO_2(am)) (all)	not in the WIPP Th(IV) model.
Kiefer et al. (2022)	0.1 M NaCl	pH ~ 3.5 - 7	ThO_2(am, hyd, fresh)	All results were excluded based on G10 ionic strength and pH conditions.
Kim et al. (2010)	Distilled H_2O; dilute solutions with various combinations of $HClO_4$, Na_2CO_3, $NaClO_4$ and $CaCl_2$; and naturally occurring, dilute granitic groundwater; I = 0–225 mM (all solutions)	pH = 8.6–11.2 (all solutions)	ThO_2(cr) (all solutions)	All results excluded because they failed to meet the G10 ionic strength condition.
Kitamura et al. (2013)	Cement pore water extracted by squeezing	pH ~12 – 13.5	ThO_2(am,aged)	All results were excluded because they failed to meet the G10 pH condition.
Kobayashi et al. (2009)	0.2, 0.6, 1.0, and 2.0 M $NaClO_4$ solutions with oxalic and malonic acid at 0.02 and 0.1 M.	pH ~ 4 – 5	A Th(IV) oxalate phase was suspected but not identified. No solid observed in the malonic acid exp.	All results were excluded based on G7 – no solid phase characterization, G8, malonic acid is not in the Th(IV) WIPP model, and G10 the pH and ionic strength conditions not met.
Kobayashi et al.	TIC = 0.16 M	pcH ~9 - 11	PuO_2(am,hyd)	All results were excluded. Nitrate is included in the

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
(2021)	(A) 0.5 M $NaNO_3$ (B) 2.0 M $NaNO_3$			WIPP An(IV) model (G8).
Myllykyla et al. (2017)	0.1 M NaCl solutions	pH 6.5 - ~9	ThO_2(cr)	All results were excluded because they failed to meet the G10 ionic strength condition, and the G7 solids characterization conditions.
Neck et al. (2002)	0.5 M NaCl	pcH = 1.11–13.7	$Th(OH)_4$(am) or $ThO_2 \cdot xH_2O$(am)	All results were excluded because they failed to meet the G10 ionic strength condition.
Neck et al. (2003)	(A) 0.1 M NaCl (B) 0.5 M NaCl	(A) pcH = 1.00-1.75 (B) pcH = 1.03-1.28 & 11.03-13.43	ThO_2(cr) or $ThO_2 \cdot xH_2O$(mcr)	All results were excluded because they failed to meet the G10 ionic strength condition.
Nishikawa et al. (2018)	0.5 M $NaClO_4$ and $HClO_4$ solutions	pcH 2-8	ThO_2(am)	All results were excluded because they failed to meet the G10 pcH and ionic strength conditions.
Östhols et al. (1994)	0.5 $NaClO_4$ & 0, 0.1, & 0.97 atm CO_2	pH = 3.31–7.36 (continuous titrations) & 8.21-10.45 (batch experiments)	ThO_2(mcr)	All results were excluded because they failed to meet the G10 ionic strength condition.
Prodromou et al. (2014)	0.1 $NaClO_4$ with humic acid	Not provided	$Th(OH)_4$	All results were excluded because they failed to meet the G8 criterion, humic acid is not included in the WIPP model.
Rai et al. (1990)	~0.05 M water	pH ~ 2 - 12	$UO_2 \cdot xH_2O$	All results were excluded because they failed to meet the G10 ionic strength condition.
Rai et al. (1995)	(A) 0.005-0.25 M $NaHCO_3$ (B) 0.1-2.0 M Na_2CO_3	Not provided	ThO_2(am)	Conference proceedings that do not include all experimental conditions such as pH. All results were excluded because they failed to meet the requirements of G9, and G10 (ionic strength).

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	(C) 0.1-2.0 M Na_2CO_3 & 0.1 M NaOH (D) 0.1-0.5 M NaOH & 1.0 M Na_2CO_3			
Rai et al. (1997)	(A) 0.2 m NaCl (used only for U solubilities) (B) 1.0 m NaCl (used only for U solubilities) (C) 3.2 m NaCl (data from Felmy et al., 1991) (D) 4.0 m NaCl, (E) 6.0 m NaCl, (F) 1.0 m $MgCl_2$ (I = 3 m), (G) 1.82 m $MgCl_2$ (I = 5.46 m), & (H) 3.0 m $MgCl_2$ (I = 9 m)	(A) U not included in this analysis (B) U not included in this analysis (C) See entry for 3.0 M NaCl for Felmy et al. (1991) in this table (D) pcH = 3.02-5.16 (E) pcH = 3.32-5.28 (F) pcH = 3.84-4.94 (G) pcH = 3.17-5.46 (H) pcH = 3.52-5.65	ThO_2(am)	All results were excluded because they failed to meet the G10 ionic strength and/or pcH conditions.
Rai et al. (2000)	(A) 0.1 M NaCl (B) 0.1 M NaCl (C) 0.1 M NaCl (D) 0.1 M NaCl (E) 0.1 M NaCl	(A) pH = 2.0-4.7 & 1.5-2.2 (B) pH= 4.2-5.1 & 2.0-2.1 (C) pH = 1.9-4.2 & 1.9-3.0 (D) pH = 1.3-3.6 (E) pH = 1.4-3.5	(A) ThO_2(am) & ThO_2(am to cr) (B) ThO_2(am) & ThO_2(am to cr) (C) ThO_2(am) & ThO_2(am to cr) (D) ThO_2(cr) (E) ThO_2(cr)	All results were excluded because they failed to meet the G10 ionic strength and/or pcH conditions.

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
Rai et al. (2005)	HCO_3^-/CO_3^{2-}-free & HCO_3^-/CO_3^{2-}-bearing (up to 6.2 m) solutions with numerous other solutes dissolved from borosilicate glass	pH ≈ 0.5–12	Not determined. May have included a Th- & Si-bearing solid(s)	All results were excluded based on the G7 and G8 requirements: (1) Solubility-controlling solid(s) were not identified; and (2) several components of the glass, especially SiO_2 and Al_2O_3, are not in the WIPP model.
Rai et al. (2008)	(A) 0.0025-0.1 M Na_2SiO_3, with "carbonate impurities" (B) 0.0025-0.14 M Na_2SiO_3, HCO_3^-/CO_3^{2-}-free (C) 0.0356 M Na_2SiO_3, (fixed) (D) 0.0157 M Na_2SiO_3, (fixed)	(A) pH 11, 12, or 13 (B) pH = 10, 12, or 13 (C) pH = 10–13.3 (variable) (D) pH = 10–13.3 (variable)	ThO_2(am) containing NO_3^- impurities form preparation and SiO_2, probably sorbed to the surface (all experiments)	All results were excluded based on the G8 requirements, SiO_2 is not included in the WIPP model.
Rai et al. (2009)	(A) 0.0002–0.2 m ISA (B) 0.008 or 0.08 m ISA	(A) pH ≈ 6 or 12 (fixed) (B) pH = 4.5–12	ThO_2(am) (all experiments)	All results were excluded based on the G8 requirements because the $Th(OH)ISA^{2+}$, $Th(OH)_3(ISA)_2^-$, and $Th(OH)_4(ISA)_2^{2-}$ complexes found to be important by Rai et al. (2009) are not in the WIPP Th(IV) model.
Tasi et al. (2018)	0.1m NaCl–NaOH–NaISA–$CaCl_2$ solutions	8 ≤ pHm ≤ 12.5	PuO_2 (ncr, hyd) nrc = nanocrystalline hyd = hydrated	This study examined the effect of α–D–isosaccharinic (ISA) acid on the solubility of PuO_2. All of the data were excluded because α–D–isosaccharinic is not included in the WIPP An(IV) model (G8).

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Reference	Solution(s) and Ionic Strength(s)	pH, pcH, or pmH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
Vandenborre et al. (2008)	0.01 M NaCl	pH = 3.0 or 4.0	ThO_2(cr) spheres with an average diameter of 500 μm	All results were excluded because they failed to meet the G10 ionic strength condition.
Vandenborre et al. (2010)	0.01 M NaCl	pH = 0.0 to 5.2	ThO_2(cr) spheres with an average diameter of 500 μm	All results were excluded because they failed to meet the G10 ionic strength condition.
Wood (2005)	(A) 0.1, 1.0, & 3.0 M $NaNO_3$; 100–500 μM citrate, EDTA; & oxalate (B) 0.1 M $NaNO_3$; 100–500 μM citrate, EDTA; & oxalate	(A) pH ≈ 2–10.5 (B) pH ≈ 3–12	(A) CeO_2 (crystallinity and hydration state unspecified) (B) "A relatively poorly crystalline form of ThO_2"	All results were excluded based on the G8 requirements because NO_3^- is not in the WIPP Th(IV) model.
Xia et al. (2003)	(A) 0.5 M $NaNO_3$ & 0.01 M EDTA (B)) 3.0 M $NaNO_3$ & 0.01 M EDTA (C)) 6.0 M $NaNO_3$ & 0.01 M EDTA (D)) 0.5 M $NaNO_3$ & 10^{-5}–10^{-2} M EDTA (E) 0.5 M $NaNO_3$ & 10^{-5}–10^{-2} M EDTA (F) 0.5 M $NaNO_3$ & 10^{-5}–10^{-2} M EDTA (G) 0.5 M $NaNO_3$ & 10^{-5}–10^{-2} M EDTA	(A) pcH = 4.3–11.8 (B) pcH = 6.7–12.8 (C) pcH = 6.8–13.2 (D) pcH = 8 (E) pcH = 10 (F) pcH = 8 (G) pcH = 10	Th(IV) hydrous oxide (ThO_2(am)) (all)	All results were excluded based on the G8 requirements because NO_3^- is not in the WIPP Th(IV) model.

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The An(IV) Uncertainty Analysis

This section describes the comparisons of experimentally measured An(IV) solubility and the solubilities predicted using the WIPP An(IV) speciation and solubility model included in the EQ3/6 thermodynamic database, DATA0.FM6.

EQ3NR input files were created to simulate the seven experimental results from Altmaier et al. (2004) that met the screening criteria. Five of these results were for 5.0 M NaCl solutions and two for 0.25 M $MgCl_2$ solutions. These input files were run using DATA0.FM6 as the thermodynamic database, and the results were extracted

Figure 1 provides the histogram and Figure 2 provides the empirical CDF for the comparisons of experimentally measured and predicted Th(IV) solubilities for the RPPCR/CRA-2024 . The current comparison includes a total of seven measured and predicted solubilities. Table 4 summarizes the statistical parameters of the current frequency distribution for RPPCR/CRA-2024 compared with the CRA-2019 DPA.

Inspection of Figure 1 shows that the WIPP Th(IV) model, the EQ3/6 code, and the DB DATA0.FM6 overpredicted 5 ($D < 0$) of the 7 measured solubilities included in the current analysis and underpredicted 2 ($D > 0$). The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Th(IV) solubilities are -0.758 and -0.947, respectively (Table 4), and the mean and median values of the sampled uncertainty factor are $10^{-0.758} = 0.174$ and $10^{-0.947} = 0.113$. (The WIPP PA codes uses actinide solubilities that are the products of the baseline solubilities and the sampled uncertainty factors).

The sorted D-values and their cumulative probability values, i.e., cumulative distribution function (CDF) for Th(IV) for the RPPCR/CRA-2024 are tabulated in Table 5 and are shown graphically in Figure 2. The PA code 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 the selected data sets for the RPPCR/CRA-2024 .

Table 4 provides the descriptive statistics for the RPPCR/CRA-2024 An(IV) actinides and compares them to the CRA-2019 DPA (Domski, 2019a) values. Appendix A provides a breakdown of D values by author and study the statistics for each data set, and plots of the measured experimental data compared to the EQ3NR modeled data.

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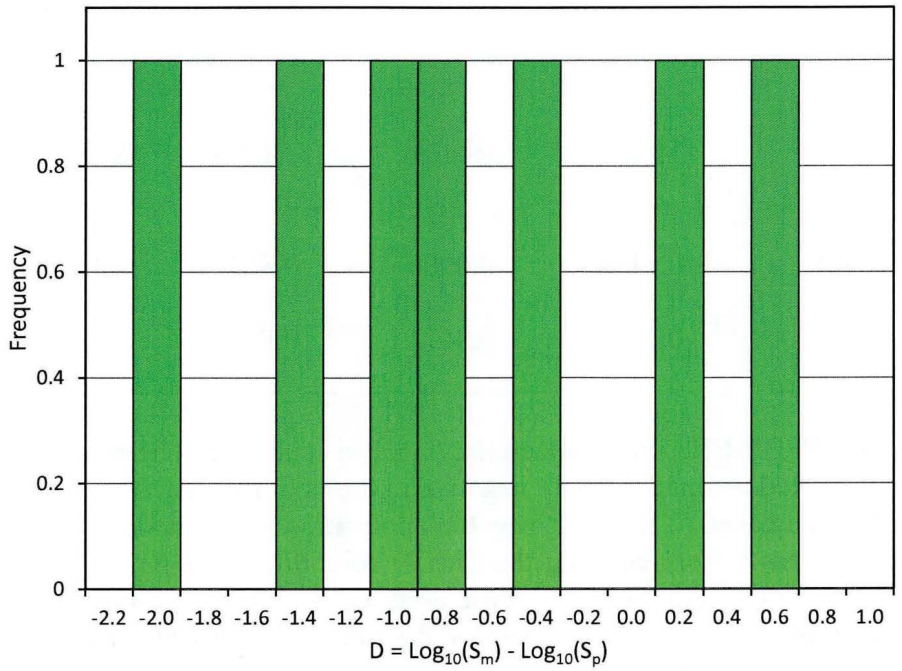


Figure 1. Histogram for the RPPCR/CRA-2024 of the difference D = log10 Sm – log10 Sp for Th(IV).

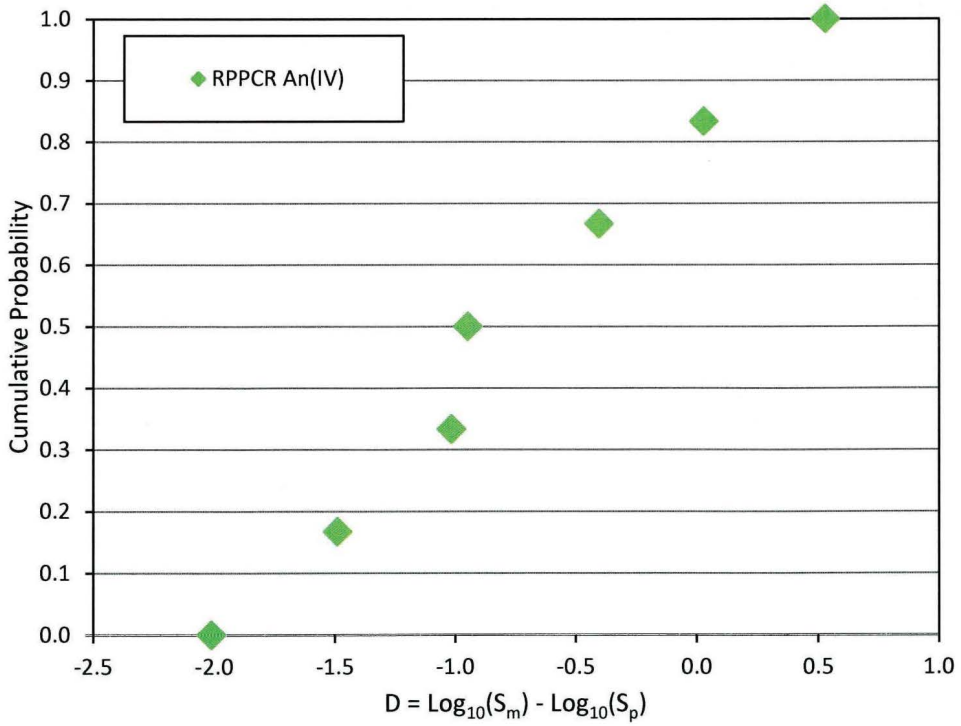


Figure 2. CDF plot for RPPCR/CRA-2024 of the difference D = log10 Sm – log10 Sp for Th(IV).

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Table 4. Statistical Properties of the Distribution of D = log10(Sm) – log10(Sp) for Th(IV) for the RPPCR/CRA-2024 compared with the CRA-2019 DPA data (Domski, 2019a).

Analysis	Mean	Median	Standard Deviation	Minimum	Maximum
CRA-2019 DPA	–0.0114	–0.0900	0.821	–2.01	1.43
RPPCR/CRA-2024	-0.758	-0.947	0.876	-2.01	0.53

The results of the RPPCR/CRA-2024 uncertainty analysis show that the mean and maximum of the distribution is shifted to smaller (more negative in terms of the mean) values compared to the CRA-2019 DPA. The cause of the shift was the application of the update G10 criterion which eliminated data that was underpredicting the Th(IV) solubility. The Th(IV) model included in DATA0.FM6 is the same as that in the database used, DATA0.FM4 (Domski, 2019b), therefore, there was no impact of using the new database on the calculated Th(IV) concentrations.

Table 5. CDF of the Differences between Measured and Predicted Th(IV) Solubilities for RPPCR/CRA-2024 . This CDF Defines the Solubility Multiplier SOLVAR for the Oxidation-State IV Model Material SOLMOD4 in the WIPP PA Parameter Database.

D	*P(D<X)* for the RPPCR/CRA-2024
-2.010	0.0000
-1.489	0.1667
-1.016	0.3333
-0.947	0.5000
-0.404	0.6667
0.029	0.8333
0.529	1.0000

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3.2 An(III) Solubility Model

Literature Screening

A new literature search was performed for the RPPCR/CRA-2024 to identify experimental studies for inclusion in the An(III) uncertainty analysis. The An(III) literature search included the following elements: Am(III), Nd(III), Cm(III), and Pu(III). The literature search identified one potential study, that of Adam et al. (2021), which after careful consideration did not provide any additional data to the +III uncertainty distribution. A discussion of Adam et al. (2021) is provided below.

Domski and Nemer (2023) applied the updated screening criterion G10 to references that were screened in for the An(III) uncertainty analysis in response to EPA's completeness comment CC1-GEOCHEM-1 as documented in Knerr (2021). This resulted in the exclusion of 97 of 129 experimental data points from the An(III) uncertainty distribution. For the RPPCR/CRA-2024 these literature were rescreened as were the experimental studies that had previously been screened in or out in previous uncertainty analyses (e.g., Brush and Domski, 2013; Domski, 2019a; Knerr 2021) by application of the screening criteria of Domski and Nemer (2023). However, the rescreening did not identify any new data to be included in the RPPCR/CRA-2024 An(III) uncertainty distribution.

An(III) Studies Considered for the RPPCR/CRA-2024 Actinide Uncertainty Analysis

One potential An(III) experimental solubility study, that of Adam et al. (2021), was identified for possible inclusion in the RPPCR/CRA-2024 +III actinide uncertainty analysis.

Adam et al. (2021) investigated the impact of cement additives adipic acid, methyl acrylate, citric acid, melamine, ethylene glycol, phthalic acid and gluconic acid[3] on the solubility of Nd(III), Th(IV), and U(VI) in carbonate-free experiments. The ligands were investigated singly and not as a composite mixture. The solubility-controlling phase used in the Nd experiments was identified as $Nd(OH)_3(s)$ where the designation of "(s)", rather than (am) or (cr), indicates that a crystalline form $Nd(OH)_3$ was used as a starting material in the experiment but that the solubility behavior may have been dominated by processes and/or characteristics at the mineral – aqueous interface and may not reflect the expected solubility of a crystalline solid, from Neck et al. (2009):

> *"The use of the notation "crystalline" and "amorphous" to describe a solid phase and thereby its solubility is an oversimplification. The X-ray diffraction (XRD) data give only information of the bulk structure, while the solubility is determined by the surface characteristics or by fractions of smaller ("amorphous") particles included in a bulk crystalline solid."*

When applied to solubility controlling mineral phases the "(s)" designation is used consistently in studies that originate from the Karlsruhe Institute of Technology (KIT), including Adam et al. (2021), and others such as Neck et al. (2009), Herm et al. (2015), and Hinz et al. (2015).

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[3] - Gluconic acid was only used with the Th(IV) and U(VI) experiments, but not with the Nd(III) experiments.

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The Nd(III) experiments of Adam et al. (2021) were run in matrix solutions of NaCl (2.64 and 5.61 m), $MgCl_2$ (1.03 and 3.87 m), and $CaCl_2$ (1.02 and 3.91 m). The 5.61 m NaCl and the 1.03 $MgCl_2$ matrix solutions met the G10 ionic strength criterion, but excluded the 1.02 and 3.91 m $CaCl_2$, the 2.64 m NaCl, and the 3.87 m $MgCl_2$ solutions because they exceeded the ionic strength conditions of G10. The ligand concentrations were "…fixed at 0.025 M for all ligands except adipic acid, melamine and phthalic acid. Due to the reduced water solubility of these ligands, lower concentrations (0.001–0.003 M) were used." The results of the experiments showed that with the exception of citrate none of the other ligands affected $Nd(OH)_3(s)$ solubility. Therefore, 18 of 29 solubility measurements for the 5.61 m NaCl satisfied both the ionic strength and pcH conditions of G10, and 12 of 12 solubility measurements in 1.03 m $MgCl_2$ met the G10 conditions and could be included. These data also meet the G8 conditions because the ligands not included in the WIPP model (melamine, phthalate, adipate, methyl acrylate, and ethylene glycol) were shown to be non-participating, while citrate, which was observed to increase solubility, is included in the WIPP An(III) model. The only disqualifying issue with this study was that the authors did not report having performed any post-test phase characterization, therefore, based on criterion G7 none of the data from this study could be included in the RPPCR/CRA-2024 uncertainty analysis.

All of the literature cited in Table 6 (see below) were re-screened for the RPPCR/CRA-2024 , and none of the references provided any additional data to be included in the RPPCR/CRA-2024 actinide uncertainty analysis. Therefore, the same references, and data, as were screened in by the analysis of Domski and Nemer (2023) were used to construct the An(III) uncertainty distribution for the RPPCR/CRA-2024 . Table 6 provides the references, conditions, and screening rationale for literature that was closely scrutinized for the RPPCR/CRA-2024 .

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Table 6. Final Evaluations of Laboratory 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 of Literature / Experimental Results
Adam et al. (2021)	(A) 2.64 m NaCl (B) 5.61 m NaCl (C) 1.03 m $MgCl_2$ (D) 3.87 m $MgCl_2$ (E) 1.02 m $CaCl_2$ (F) 3.91 m $CaCl_2$	(A) ~9, ~13 pmH (B) ~9, ~13 pmH (C) ~9 pmH (D) ~9 pmH (E) ~9, ~12 pmH (F) ~9, ~12 pmH	$Nd(OH)_3$(s) was the starting material, there was no post-test characterization.	This paper examines the impact of cement additives adipic acid, methyl acrylate, citric acid, melamine, ethylene glycol, phthalic acid and gluconic acid on the solubility of Nd(III), Th(IV), and U(VI). A total of 30 results met the G10 condition for ionic strength and pmH ≈ 9 - 10, 18 results for 5.61 m NaCl, and 12 results for 1.03 m $MgCl_2$ for all additives (G8). However, these results did not meet the G7 criterion as there was no post-test solids characterization, no results included in the current analysis.
Borkowski et al. (2009)	(A) 5 M NaCl (B) GWB, diluted to 95% of its specified concentrations (C) ERDA-6, diluted to 95% of its specified concentrations	(A) pcH = 8.2–13.0 (B) pcH = 6.6–8.7 (C) pcH = 7.9–10.3	Presumably $Nd(OH)_3$, based on their thermodynamic predictions.	EPA stated during the CRA-2014 PA review that DOE must recalculate the Am(III) uncertainty distribution and exclude the Borkwski et al. (2009) data (Edwards 2015, Comment 3-C-5). These results are excluded because of the lack of characterization data of the solubility-controlling phase (G7). This comment applies to Borkowski (2010), Borkowski et al. (2010), Borkowski (2012),and Lucchini et al. (2007).
Borkowski (2010)	(A) 5 M NaCl (B) GWB, diluted to 95% of its specified concentrations of solutes to avoid coprecipitation of Nd by evaporite minerals (C) ERDA-6, diluted to 95% of its specified concentrations of	(A) pcH = 8.14–13.17 (B) pcH = 6.54–8.64 (C) pcH = 7.55–10.62	Presumably $Nd(OH)_3$, based on their thermodynamic predictions.	Same as above.

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	solutes to avoid coprecipitation of Nd by evaporite minerals			
Borkowski et al. (2010)	1, 2, 3, 4, or 5 M NaCl with 0.00. 0.01, 0.02, 0.03, or 0.04 M $Na_2B_4O_7$; all runs from undersaturation	pcH = 8.59 ± 0.04 (constant, all runs)	Presumably $Nd(OH)_3$, which was used to start these runs	Same as above.
Borkowski (2012)	Same as above	Same as above	Same as above	Same as above
Bunyakina & Storozhenko (2004)	Solutions saturated with $MgSO_4$ and/or $Nd_2(SO_4)_3$. Ionic strengths not provided.	Not provided	$Nd_2(SO_4)_3{\cdot}8H_2O$	All results excluded because $Nd_2(SO_4)_3{\cdot}8H_2O$ is not in the WIPP Am(III) model (G7).
Herm et al. (2015)	(A) NaCl (m) 0.1, 0.51, 1.02, 2.64, 5.51 (B) $MgCl_2$ (m) 0.25, 2.80, 4.05, 5.24 (C) $CaCl_2$ (m) 2.85, 4.02 At NO_3 (m) from 0.0 to 8.04 (see paper for details)	(A) pmH ~8 – 13 (B) pmH ~8 – 12 (C) pmH ~8 – 12	$Nd(OH)_3$(s)	This study was identified and included for the uncertainty analysis documented in Knerr (2021). Ten results (4 NaCl and 6 $MgCl_2$) were included in Knerr (2021). The G10 criterion was applied to the results by Domski and Nemer (2023) which resulted in screening out 2 of the 4 NaCl results and 5 of the 6 $MgCl_2$ results, with 3 results included in the RPPCR/CRA-2024 uncertainty analysis.
Hinz et al. (2015)	(A) NaCl (M) 0.1, 1.0, 5.0 (B) NaCl (M) 0.1, 5.0 (C) $MgCl_2$ (M) 0.25, 1.0, 3.5 (D) $MgCl_2$ (M) 0.25,	(A) pcH 7 – 13 (B) pcH 6 – 9 (C) pcH 6 – 9 (D) pcH 6 – 9	(A) $Nd(OH)_3$(am) (B) $Nd[B_9O_{13}(OH)_4]$ (C) $Nd(OH)_3$(am) (D) $Nd[B_9O_{13}(OH)_4]$	This study was identified, and select results were included for demonstration in the uncertainty analysis documented in Knerr (2021). This study did not meet the G8 criterion used in the Knerr (2021) analysis because the An(III) model does not borate. However, under the revised screening criteron (G8) results may be included if the

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	3.5 (E) $CaCl_2$ (M)0.25, 1.0, 3.5	(E) pcH 8 – 12	(E) $Nd(OH)_3$(am)	*"non-participating cations and/or anions, or additional known and characterized dissolved elements that have been demonstrated to not strongly complex or effect the solubility of the actinide(s) of interest."*. Hinz et al. (2015) state: "*No significant effect of borate on Nd(III) solubility is observed in near neutral to slightly alkaline pH values (7 ≤ pHc ≤ 9) and [B]tot ≤ 0.04 M (dilute salt systems: 0.1 and 1.0 M NaCl; 0.25 M $MgCl_2$) or [B]tot ≤ 0.004 M (concentrated salt systems: 5.0 M NaCl; 1.0 and 3.5 M $MgCl_2$). Under these conditions, the concentration of Nd(III) is in good agreement with borate-free solubility data obtained under analogous pHc and ionic strength.*" Based on this information experimental data provided by the author was screened resulting in two data points for 5.0 M NaCl with 0.004 M B_{total}, one data point for 0.25 M $MgCl_2$ with 0.004 M B_{total}, one data point for 0.04 M B_{total}, and one data point for 1.0 M $MgCl_2$ with 0.004 M B_{total}. Because the dataset contained replicates collected at approximately the same pcH the average values of pcH and log[Nd] were used for each replicate in this analysis for a total of five included results. See *An(III)_RPPCR_Uncertainty.xlsx* in the records package for details. The remaining results were screened out because they didn't meet the ionic strength and/or pH conditions of G10, and because of the effect of borate on the solubility of An(III).
Khalili et al. (1994)	Synthetic brine	pcH = 6.4, 8.4, 10.4,	$Nd(OH)_3 \cdot nH_2O$(am)	The DOE and EPA agreed at the September 2018

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	similar to GWB	& 12.4	at pcH = 6.4 & 8.4, $Nd_2(CO_3)_3 \cdot 8H_2O$ or "uncertain" at pcH = 10.4, NdO(OH) & $Nd(OH)_3$ or "uncertain" at pcH = 12.4	technical exchange meeting that the results of this study should not be included in the actinide uncertainty analysis (Page 105, U.S. EPA, 2022). All results were excluded because the conditions of G7 were not met - post-test solids characterization, G9 insufficient experimental procedure provided and uncertain laboratory conditions (e.g., exposure to $CO_2(g)$), and G4 poorly defined aqueous composition.
Lucchini et al. (2007)	(A) GWB (B) ERDA-6	(A) pcH = 6.6–8.7 (B) pcH = 7.9–10.3	$Nd(OH)_3$ (both solutions), based on their thermodynamic predictions.	Excluded because Borkowski et al. (2009) and Borkowski (2010) superseded this paper.
Makino et al. (1993)	0.01 M $NaClO_4$ & 0.1 ppm CO_2	pcH = 7–13	$Nd(OH)_3$(cr) with lesser amounts of $Nd(OH)CO_3$(cr) (unclear whether both of these solids were in equilibrium one was replacing another).	All results were excluded because they do not meet the ionic strength conditions of G10. In addition, neither the TIC concentration of the aqueous phase nor the pCO_2 of the gaseous phase was provided, thus precluding predictions of Nd(III) solubilities for their experiments (G9).
Meinrath & Kim (1991)	(A) 0.1 M $NaClO_4$ & 0.00031 atm CO_2	(A) pH = 4.1-4.3	(A) $Nd_2(CO_3)_3$ & $Nd(OH)CO_3$ initially, $Nd(OH)CO_3$ after 2 weeks	All results were excluded because they do not meet the ionic strength and pH conditions of G10. In addition, the results from (B) and (C) were excluded because $Nd_2(CO_3)_3$ is not in the model (G8).
	(B) 0.1 M $NaClO_4$ & 0.01 atm CO_2	(B) pH = 5.2-5.4	(B) $Nd_2(CO_3)_3$	
	(C) 0.1 M $NaClO_4$ & 1 atm CO_2	(C) pH = 5.7-6.2	(C) $Nd_2(CO_3)_3$	
Meinrath & Takeishi (1993)	(A) 0.1 M $NaClO_4$ & 0.0003 atm CO_2	(A) pH = 5.6-6.6	(A) $Nd_2(CO_3)_3$ & $Nd(OH)CO_3$ initially, $Nd(OH)CO_3$(mcr) after 5-7 days	All results were excluded because they do not meet the ionic strength and pH conditions of G10. In addition, the results from (C), (D), and (E) were excluded because $Nd_2(CO_3)_3$ is not in the model (G8).

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	(B) 0.1 M $NaClO_4$ & 0.003 atm CO_2	(B) pH = 5.4-6.3	(B) $Nd_2(CO_3)_3$ & $Nd(OH)CO_3$ initially, $Nd(OH)CO_3$(mcr) after 30-40 days	
	(C) 0.1 M $NaClO_4$ & 0.01 atm CO_2	(C) pH = 5.2-6.2	(C) $Nd_2(CO_3)_3 \cdot$(4.5-6 H_2O)	
	(D) 0.1 M $NaClO_4$ & 0.08 atm CO_2	(D) pH = 4.7-5.9	(D) $Nd_2(CO_3)_3 \cdot$(4.5-6 H_2O)	
	(E) 0.1 M $NaClO_4$ & 1 atm CO_2	(E) pH = 4.2-5.2	(E) $Nd_2(CO_3)_3 \cdot$(4.5-6 H_2O)	
Migdisov and Williams-Jones (2007)	$HClO_4$ =$NaHClO_4$ (total ClO_4^- = 0.057-1.97 M	pH = 1.04-1.65 (at 25 °C)	NdF_3	All results were excluded because Migdisov and Williams-Jones (2007) carried out their experiments at 150, 200, and 250 °C in violation of G5, and because F is not in the WPP Am(IIII) model (G8).
Neck et al. (2009)	(A) 0.1, 0.5, 2.5, & 5.0 M NaCl; 0.1 M $NaClO_4$	(A) pcH ≈ 7 to 13, 14, or even 15	(A) $Nd(OH)_3$ or $Cm(OH)_3$ (5 M NaCl &1.0, 2.5, & 3.5 M $CaCl_2$)	Results included in Knerr (2021) were screened with G10 and documented in Domski and Nemer (2023). The results accepted in Knerr (2021) included 8 data points from (A) 5.0 M NaCl; 16 from (B) 5 from 1.0 M $MgCl_2$, 3 from 2.5 M $MgCl_2$, and 8 from 3.5 M $MgCl_2$; and 32 from (C) 9 from 1.0 M $CaCl_2$, 10 from 2.5 M $CaCl_2$, and 13 from 3.5 M $CaCl_2$, for a total of 56 included data points. Application of G10 reduced the number of included data points to 5 for 1.0 M $MgCl_2$.
	(B) 0.25, 1.0, 2.5, & 3.5 M $MgCl_2$	(B) pcH ≈ 7 or 7.5 to 9		
	(C) 0.25, 1.0, 2.5, & 3.5 M $CaCl_2$	(C) pcH ≈ 7-9 to 12		
Peretrukhin et al. (1996)	1 & 5 M NaOH (experiments with Am(III))	Not provided	$Am(OH)_3$ aged by boiling for 3 hours (undersaturation runs) or $Am(OH)_3$, crystallinity unspecified (oversaturation runs)	All results were excluded because none of the data met the conditions of G10, pH values were not provided. Experimental procedures not clearly defined (G9).

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
Rabung et al. 2008	(A) 0.1–3.5 M $CaCl_2$	pcH ≈ 11.7	$Cm(OH)_3$(am) (all)	All results were excluded because none of the data met the conditions of G10, (A) met the ionic strength condition, but not pH. Solubility data for (A) not provided (G9).
	(B) 1.0 M $CaCl_2$	pcH = 10.8–11.9		
	(C) 2.5 M $CaCl_2$	pcH = 10.8–11.9		
	(D) 3.5 M $CaCl_2$	pcH = 10.8–11.9		
Rao et al. (1996a)	0.1 M NaCl	pH = 4.8-9.0 (filtered samples in their Fig. 1) & 5.1-9.0 (41-day samples in their Fig. 2.	$Nd(OH)_3$(cr)	All results were excluded because none of the data met the conditions of G10, all data were below the ionic strength and pH thresholds.
Rao et al. (1996b)	(A) 0.1-1.0 M $NaHCO_3$ (B) 0.1-2.0 M Na_2CO_3	Not provided	$NaNd(CO_3)_2 \cdot 6H_2O$(cr) (all)	All results were excluded because none of the data met the conditions of G10, most exceeded the maximum TIC.
Rao et al. (1999)	(A) 0.1-1.1 m $NaHCO_3$	(A) pH = 7.9-8.3	$NaNd(CO_3)_2 \cdot 6H_2O$ (all)	All results were excluded for (A) through (E) because none meet the ionic strength condition of G10. Data from (F) is excluded because the data does not meet the pcH condition of G10. Eleven of the results from (G) (see Figure 6a in reference) met all of the conditions of G10 and are included in the current analysis.
	(B) 0.1-2 m Na_2CO_3	(B) pH = 10.4-10.8		
	(C) 2 M NaCl & 0.1-0.5 m $NaHCO_3$	(C) pH = 7.9-8.3		
	(D) 2 M NaCl & 0.1-2 m Na_2CO_3	(D) pH = 10.4-10.8		
	(E) 4 M NaCl & 0.1-2 m Na2CO3	(E) pcH = 10.4-10.8		
	(F) G Seep, a synthetic brine similar	(F) pcH = 5.6-7.8		

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
	to GWB			
	(G) ERDA-6	(G) pcH = 6.2-10.6		
Runde et al. (1992)	0.1 M $NaClO_4$ & 0.0003, 0.01, & 1 atm CO_2	pH = 5.5 – 6.3	$Nd(OH)CO_3$ at 0.0003 atm CO_2, $Nd_2(C0_3)_3$ at 0.01 & 1 atm CO_2	All results were excluded because only K_{sp}'s reported, not the actual solubility data.
Runde & Kim (1995)	(A) 5.0 M NaCl (B) 5.0 m NaCl & 0.01 atm CO_2	(A) pcH = 6.7–13.8 (B) pcH = 6.6-9.3	(A) $Am(OH)_3$(cr) (B) $NaAm(CO_3)_3 \cdot xH_2O$(cr)	(A) Five results included that met the conditions of the G10 criterion. (B) All results were excluded, the TIC exceeded 0.02 M for all results that met the G10 pcH conditions.
Silva (1982)	(A) 0.1 M $NaClO_4$ (B) 0.1 M $NaClO_4$	(A) pH = 5.7-9.5 (B) pH = 7.0-9.4	(A) $Nd(OH)_3$(cr) (B) $Am(OH)_3$(cr)	All results were excluded because the ionic strength condition of G10 was not met.
Vitorge and Tran The (1991)	(A) "Lime water" (composition unspecified) with 0-10 M KOH (B) "Lime water" (composition unspecified) (C) "Cement water" (composition unspecified) with 0-10 M KOH	Unspecified for (A), (B), and (C)	$Am(OH)_3$?	All results were excluded because the compositions of the solutions were not specified (G9).
Wood et al. (2002)	(A) 0.03 m $NaTr^A$ (30 °C) (B) 0.03, 0.10, & 1.0 m $NaTr^A$ (50 °C)	(A) pmH = 7.01-9.13 (B) pmH = 6.494–9.548 (I = 0.03 m) pmH = 6.653-6.711 (I = 0.10 m) pmH = 6.681-6.809	$Nd(OH)_3$(cr)	The results at 30 °C were excluded because they did not satisfy the ionic strength condition of G10. All other results excluded because they exceeded the maximum temperature (30 °C) specified by G5.

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
		(I = 1.0 m)		
	(C) 0.03, 0.10, & 1.0 m $NaTr^A$ (100 °C)	(C) pmH = 5.362–8.421 (I = 0.03 m) pmH = 5.739-5.744 (I = 0.10 m) pmH = 5.857-5.859 (I = 1.0 m)		
	(D) 0.03, 0.010, & 1.0 m $NaTr^A$ (150 °C)	(D) pmH = 4.791-5.890 (I = 0.03 m) pmH = 5.017-5.021 (I = 0.10 m) pmH = 5.139-5.141 (I = 1.0 m)		
	(E) 0.03, 0.10, & 1.0 m $NaTr^A$ (200 °C)	(E) pmH = 4.419-5.088 (I = 0.03 m) pmH = 4.472-4.473 (I = 0.10 m) pmH = 4.558-4.560 (I = 1.0 m)		
	(F) 0.03, 0.10, & 1.0 m $NaTr^A$ (250 °C)	(F) pmH = 3.873-4.873 (I = 0.03 m) pmH = 4.050-4.068 (I = 0.10 m) pmH = 4.180-4.182 (I = 1.0 m)		
	(G) 0.03 & 0.10 m (290 °C)	(G) pmH = 3.493–6.898 (I = 0.03 m) pH = 3.406-5.221 (I = 0.10 m)		
Xiong et al. (2017)	(A) 4.4 m NaCl (B) 5.0 m NaCl	pmH ~9.5	$Nd(OH)_3$ (mcr) mcr = microcrystalline	Data from Table 1 of the report provides 14 replicate samples for both ionic strengths, (A) and (B). The average value of pmH and [Nd(III)] was used in the analysis presented in Knerr (2021), and the 4.4 m NaCl was screened out (G10) by Domski and Nemer (2023). For the RPPCR/CRA-2024 the average 5.0 m NaCl data

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Reference	Solution(s) and Ionic Strength(s)	pcH, pmH, or pH	Solubility-Controlling Solid	Evaluation of Literature / Experimental Results
				was included.

A. Sodium trifluoromethanesulfonate, abbreviated as "sodium triflate" or "NaTr," is a noncomplexing salt used to set the ionic strength. In EQ3/6 calculations, the noncomplexing medium, sodium perchlorate, is used as the supporting solution.
B. Sodium trifluoromethanesulfonate, abbreviated as "sodium triflate" or "NaTr," is a noncomplexing salt used to set the ionic strength.

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The An(III) Uncertainty Analysis

This section describes the comparisons of experimentally measured An(III) solubility and the solubilities predicted using the WIPP An(III) speciation and solubility model included in the EQ3/6 thermodynamic database, DATA0.FM6. It includes information similar to that provided for Th(IV) in Subsection 3.1 (see above).

The following studies were included in the RPPCR/CRA-2024 An(III) actinide uncertainty analysis, for a discussion of each study see Domski (2019a) and Knerr (2021).

- One result from Xiong et al. (2017) in 5.0 M NaCl using $Am(OH)_3(am)$ as the solubility-controlling phase in DATA0.FM6.
- Two results in 5.61 molal NaCl, and one result in 1.04 molal $MgCl_2$, for a total of three samples from Herm et al. (2015) using $Am(OH)_3(am)$ as the solubility-controlling phase in DATA0.FM6.
- Five results from Hinz et al. (2015), two in 5.0 M NaCl 0.004 M $[B]_{tot}$, one in 0.25 $MgCl_2$ 0.004 M $[B]_{tot}$, one in 0.25 M $MgCl_2$ 0.04 M $[B]_{tot}$, and one in 1.0 M $MgCl_2$ 0.004 M $[B]_{tot}$. All using $Am(OH)_3(am)$ as the solubility-controlling phase in DATA0.FM6. There are two fewer results included in the current analysis than documented in the analysis of Domski and Nemer (2023) because replicate samples were combined and averaged.
- Eleven results in ERDA-6 ($\log P_{CO2(g)} = -3.5$) from Rao et al. (1999), were included in this analysis using $NaAm(CO_3)_2.6H_2O(cr)$ as the solubility controlling phase.
- Five samples in 1.02 m $MgCl_2$, one sample in 5.61 m NaCl for a total of six results from Neck et al. (2009). $Am(OH)_3(am)$ was used as the solubility-controlling phase in DATA0.FM6.
- Five 5 M NaCl samples of Runde and Kim (1995) were included in this analysis using the FM6 solubility-controlling phase $Am(OH)_3(s)$.

Four of the six studies included in the RPPCR/CRA-2024 An(III) uncertainty analysis used the more soluble amorphous form of Am-hydroxide as the solubility controlling phase. This polymorph Am-hydroxide was a new addition to the WIPP database with the release of DATA0.FM6. Its use in the uncertainty analysis is warranted by the observations of the authors (Xiong et al., 2017; Herm et al., 2015, Hinz et al., 2015; and Neck et al., 2009) of these papers that the solubility behavior of $Nd(OH)_2(s)$ more closely represents the amorphous form of the phase even though the starting material for each was crystalline. This is in agreement with the EPA (2022, pg. 107), where it is stated:

> *"Because the crystallinity of $Am(OH)_3(s)$ in the long-term WIPP repository is uncertain, the +III actinide solubility uncertainty distribution should incorporate the effects related to varying degrees of crystallinity on +III actinide solubilities."*

Combined, the four studies representing $Am(OH)_3(am)$ provide 14 of the 30 data points in the An(III) distribution, and of the remaining 16, five data points are represented by the lower solubility $Am(OH)_3(s)$ phase, and eleven as $NaAm(CO_3)_2.6H_2O(cr)$. Use of these three phases

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provides a broader comparison the WIPP model to experimental data. The addition of the amorphous form of Am-hydroxide to DATA0.FM6 (Domski, 2023b), and using this phase, with two other An(III) phases, in the current actinide uncertainty analysis addresses the concerns of the EPA Issue 10 (Peake, 2022) by using more appropriate phases to simulate experimental data where less-crystalline solid phases may have been controlling solubility.

Figure 3 and 4 provide the frequency histogram and the CDF of the An(III) RPPCR/CRA-2024 D-values. The current comparison includes a total of 30 measured and predicted solubilities. Table 7 summarizes the statistical parameters of the current frequency distribution generated in this analysis with that of CRA-2019 DPA (Domski, 2019a).

Figure 3 shows that the WIPP Am(III) model, using DATA0.FM6 over predicted ($D < 0$) 21 of the 30 measured Nd(III) and Am(III) solubilities, and under predicted ($D > 0$) 9 of 30 experimental solubilities. The mean and median values of the log of the sampled uncertainty factor used to adjust the baseline Am(III) solubilities are -0.509 and -0.245, respectively (Table 7), and the mean and median values of the sampled uncertainty factor are $10^{-0509} = 0.310$ and $10^{-0.245} = 0.569$. WIPP PA uses actinide solubilities that are the products of the baseline solubilities and the sampled uncertainty factors.

Table 8 provides values of the CDF for An(III) actinides for the RPPCR/CRA-2024 , which are also graphically presented in Figure 4. 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 PA calculations. Brush et al. (2005) described the PA implementation of these results in detail. . Appendix A provides a breakdown of D values by author and study the statistics for each data set, and plots of the measured experimental data compared to the EQ3/6 modeled data.

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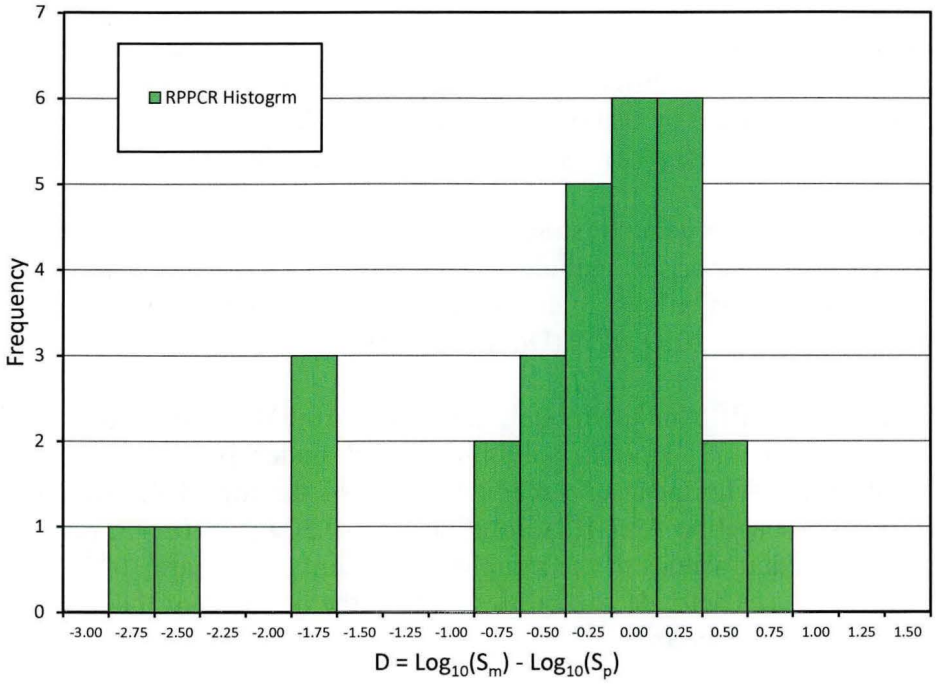


Figure 3. Histogram for the RPPCR/CRA-2024 of the difference D = log10 Sm – log10 Sp An(III).

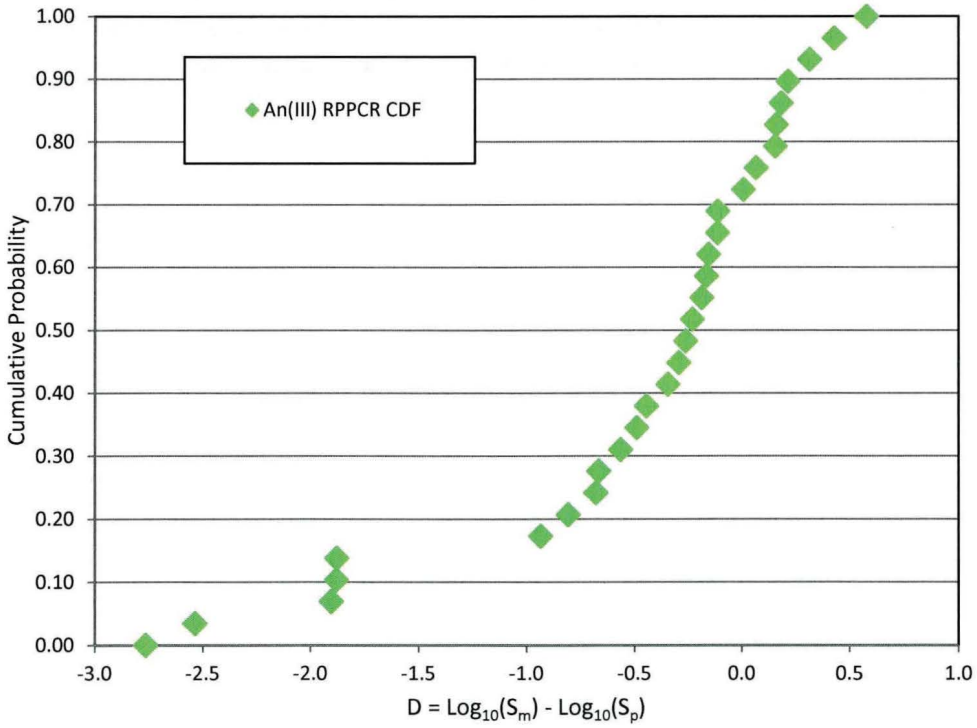


Figure 4. CDF plot for the RPPCR/CRA-2024 of the difference D = log10 Sm – log10 Sp for An(III).

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Table 7. Statistical Properties of the Distribution of D = log10(Sm) – log10(Sp) for An(III) for RPPCR/CRA-2024 compared with the CRA-2019 DPA data (Domski, 2019a).

Analysis	Mean	Median	Standard Deviation	Minimum	Maximum
CRA-2019 DPA	0.651	0.516	1.01	-1.14	2.97
RPPCR/CRA-2024	-0.509	-0.245	0.858	-2.76	0.578

Compared to the CRA-2019 DPA results the RPPCR/CRA-2024 results are shifted to toward negative values. This is direct result of the use of the amorphous form of Am-hydroxide as the solubility-controlling phase which tends provide a better fit to the measured data, albeit the fits may tend to slightly over estimate the An(III) solubility resulting in small negative values.

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Table 8. CDF of the Differences between the Measured and Predicted Nd(III) and Am(III) Solubilities for the RPPCR/CRA-2024 . 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 RPPCR/CRA-2024
-2.76	0.0000
-2.54	0.0345
-1.90	0.0690
-1.88	0.1034
-1.88	0.1379
-0.932	0.1724
-0.806	0.2069
-0.678	0.2414
-0.666	0.2759
-0.563	0.3103
-0.488	0.3448
-0.443	0.3793
-0.343	0.4138
-0.292	0.4483
-0.260	0.4828
-0.229	0.5172
-0.185	0.5517
-0.164	0.5862
-0.155	0.6207
-0.113	0.6552
-0.112	0.6897
0.008	0.7241
0.068	0.7586
0.157	0.7931
0.161	0.8276
0.184	0.8621
0.215	0.8966
0.316	0.9310
0.430	0.9655
0.578	1.0000

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4 CONCLUSIONS

The RPPCR/CRA-2024 actinide uncertainty analysis has been presented in this analysis report. This actinide uncertainty analysis differs from past analysis by the use of an updated WIPP database (Domski, 2023b) which incorporates an additional Am-hydroxide polymorph of higher solubility (Jang et al., 2021) than that used in previous actinide uncertainty analyses (eg., Brush and Domski, 2013; Domski, 2019a; Knerr 2021). Additionally, the current analysis implemented the updated literature screening criteria of Domski and Nemer (2023), which has three significant changes compared to the criteria used in previous analyses:

1) G1: Allows pre-1990 publications and sets the cutoff date to three months prior to the required parameter feed to performance assessment for the current recertification cycle, which was 10/09/2022 for the current analysis.
2) G6: Allows for consideration of additional oxidation-state analogs for the An(III) and An(IV) uncertainty analyses. The An(III) model considered Am(III), Nd(III), Cm(III), and Pu(III), the An(IV) model considered Th(IV), Pu(IV) and U(IV) studies.
3) G10: The WIPP conditions criterion was refined to be more specific to expected WIPP conditions.

An exhaustive literature search was performed for the RPPCR/CRA-2024 , which is summarized in Nemer et al. (2023), the results of which were that eight potential studies were identified for the An(IV) uncertainty analysis, and one for the An(III) analysis. Detailed examination of these experimental studies revealed that none of them qualified under all of the screening criteria, thus no new data were identified for the RPPCR/CRA-2024 uncertainty analysis. The new screening criteria were also applied to literature screened for previous uncertainty analysis, and two additional experimental data points were identified for the An(IV) distribution. The update to screening criterion G1 together with the execution of the literature search for this analysis report addresses EPA Issue 9 (Peake, 2022).

The literature screening identified eight possible An(IV) solubility studies, none of which met all of the screening criteria, however, re-screening of literature identified for previous uncertainty analysis revealed an additional two data points from the paper by Altmaier et al. (2004). The results of the An(IV) RPPCR/CRA-2024 uncertainty analysis showed that the mean and maximum of the distribution were shifted to smaller (more negative in terms of the mean) values relative to the CRA-2019 DPA. The cause of the shift was the application of the update G10 criterion which eliminated data that was underpredicting the Th(IV) solubility, and not changes to the Th(IV) model. The Th(IV) model included in DATA0.FM6 is the same as that in the database used, DATA0.FM4 (Domski, 2019b), therefore, there was no impact of using the new database on the calculated Th(IV) concentrations.

One potential solubility study was identified for the An(III) actinides. The study of Adam et al. (2021) warranted close examination as the study provided solubility data under conditions that qualified under criterion G10. Unfortunately, the authors did not report having performed post-test solids analysis, in violation of the G7 criterion, therefore, no data from this study was included in the RPPCR/CRA-2024 An(III) uncertainty analysis.

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Compared to the An(III) CRA-2019 DPA results the RPPCR/CRA-2024 results are shifted to toward negative values. This is direct result of the use of the amorphous form of Am-hydroxide as the solubility-controlling phase for some of the studies which tends provide a better fit to the measured data, albeit the fits may tend to slightly over estimate the An(III) solubility which leads to negative D values. The maximum of the An(III) uncertain distribution for the RPPCR/CRA-2024 was shifted by ~1 Log_{10} unit to the left compared to the CRA-2019 DPA while the minimum was unchanged.

The addition of the amorphous form of Am-hydroxide to DATA0.FM6 (Domski, 2023b), and the use of this phase, with two other Am(III) phases, in the current An(III) actinide uncertainty analysis addresses the concerns of EPA Issue 10 (Peake, 2022) by using more appropriate phases to simulate experimental data where less-crystalline more soluble solid phases may have been controlling solubility.

Tables 5 and 8 provide values of the empirical CDFs for the An(IV) and An(III) actinides for the RPPCR/CRA-2024 PA, respectively.

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Appendix A. An(IV) and An(III) Uncertainty Outputs

Appendix A provides the statistics for each study included in the An(IV) and An(III) uncertainty distribution. Plots are provided of the measured data as they appear in their respective reports (i.e., some are in terms of pcH and others pmH) compared to the equivalent EQ3/6 WIPP model results.

The An(IV) uncertainty distribution included only a single reference, therefore the values are the same those found in Table 4.

Table A - 1. Studies included in the An(IV) uncertainty distribution with their EQ3NR filenames and D values.

Author	EQ3/6 Filename	D
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), Tbl 2 MgCl2 0.25 M	11Alt04.3o	0.529
Altmaier et al. (2004), Tbl 2 MgCl2 0.25 M	12Alt04.3o	0.029

Table A - 2. Statistics of the D values for each study included in the An(IV) uncertainty distribution.

Reference	Maximum	Minimum	Mean	Median	n
Altmaier et al (2004)	0.529	-2.01	-0.758	-0.947	7

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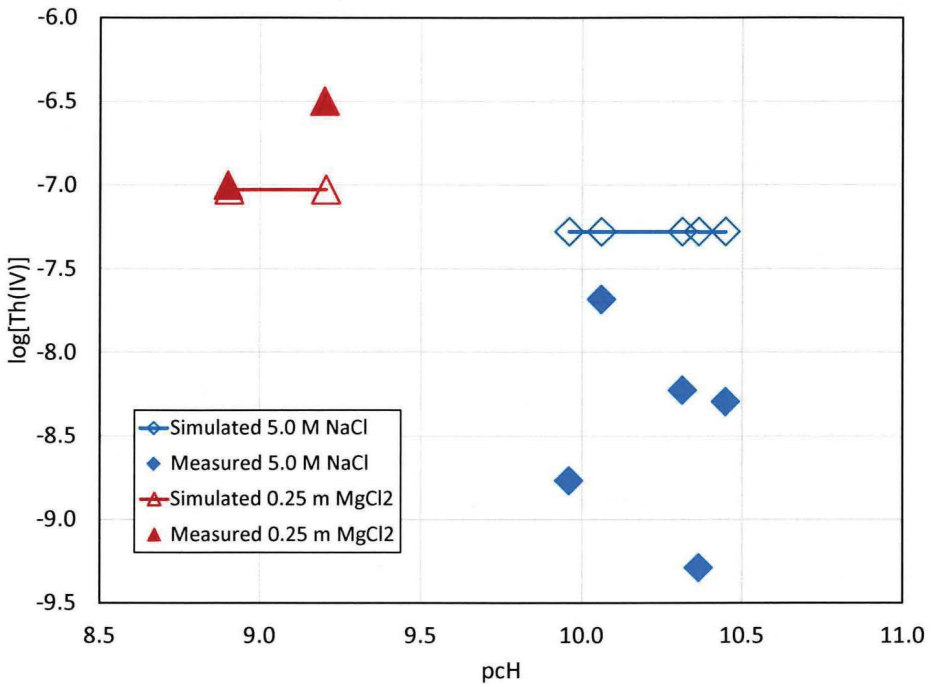


Figure A - 1. Graph of the Altmaier et al. (2004) measured data and EQ3NR model response using the DATA0.FM6 database with ThO_2(am) as the solubility-controlling phase.

Table A - 3. Studies included in the An(III) uncertainty distribution with their EQ3/6 filenames and D values.

Author	EQ3/6 Filename	D
Herm et al (2015) 1.04 m MgCl2	MgHerm1.3o	-0.343
Herm et al (2015) 5.61 m NaCl	NaHerm2.3o	-1.902
Herm et al (2015) 5.61 m NaCl	NaHerm3.3o	-2.536
Xiong et al., (2017) 5.0 m NaCl	Xio50ave.3o	-1.878
Hinz et al (2015) 5M NaCl, 0.004 Btot	NaHinz1.3o	-1.88
Hinz et al (2015) 5M NaCl, 0.004 Btot	NaHinz2.3o	-2.76
Hinz et al (2015) 0.25M MgCl2, 0.004 Btot	MgHinz1.3o	0.316
Hinz et al (2015) 0.25M MgCl2, 0.04 Btot	MgHinz1.3o	0.578
Hinz et al (2015) 1.0 M MgCl2, 0.004 Btot	MgHinz3.3o	-0.185
Neck et al. (2009) 1.04 m MgCl2	17NeckMg.3o	0.430
Neck et al. (2009) 1.04 m MgCl2	18NeckMg.3o	0.184
Neck et al. (2009) 1.04 m MgCl2	19NeckMg.3o	0.068
Neck et al. (2009) 1.04 m MgCl2	20NeckMg.3o	-0.155
Neck et al. (2009) 1.04 m MgCl2	21NeckMg.3o	-0.260
Runde & Kim (1995), 5 M NaCl	04Run95.3o	0.215
Runde & Kim (1995), 5 M NaCl	05Run95.3o	-0.443
Runde & Kim (1995), 5 M NaCl	06Run95.3o	-0.666
Runde & Kim (1995), 5 M NaCl	07Run95.3o	-0.488
Runde & Kim (1995), 5 M NaCl	08Run95.3o	-0.932
Rao et al. (1999), ERDA6, logpCO2=-3.5	05rao99.6o	-0.164
Rao et al. (1999), ERDA6, logpCO2=-3.5	06rao99.6o	-0.113
Rao et al. (1999), ERDA6, logpCO2=-3.5	07rao99.6o	0.157
Rao et al. (1999), ERDA6, logpCO2=-3.5	08rao99.6o	0.008

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Author	EQ3/6 Filename	D
Rao et al. (1999), ERDA6, logpCO2=-3.5	09rao99.6o	-0.112
Rao et al. (1999), ERDA6, logpCO2=-3.5	10rao99.6o	0.161
Rao et al. (1999), ERDA6, logpCO2=-3.5	11rao99.6o	-0.229
Rao et al. (1999), ERDA6, logpCO2=-3.5	12rao99.6o	-0.292
Rao et al. (1999), ERDA6, logpCO2=-3.5	13rao99.6o	-0.563
Rao et al. (1999), ERDA6, logpCO2=-3.5	14rao99.6o	-0.678
Rao et al. (1999), ERDA6, logpCO2=-3.5	15rao99.6o	-0.806

Table A - 4. Statistics of the D values for each study included in the An(III) uncertainty distribution.

Reference	Maximum	Minimum	Mean	Median	n
Herm et al (2015)	-0.343	-2.54	-1.59	-1.90	3
Xiong et al., (2017)	-1.88	-1.88	-1.88	-1.88	1
Hinz et al 2015	0.578	-2.764	-0.787	-0.185	5
Neck 2009	0.430	-0.260	0.053	0.068	5
Runde & Kim (1995)	0.215	-0.932	-0.463	-0.488	5
Rao et al. (1999)	0.161	-0.806	-0.239	-0.164	11

Figures A – 2 – A – 6 present the An(III) simulated WIPP model fits using DATA0.FM6 to the measured experimental data that met the updated criteria (Domski and Nemer, 2023).

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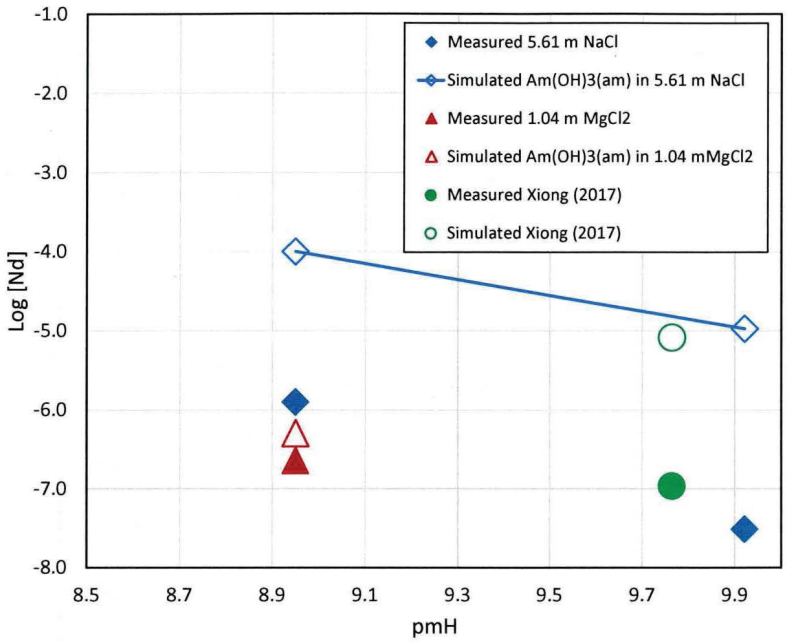


Figure A - 2. Graph of the Herm et al. (2015) and Xiong et al. (2017) measured data and EQ3NR model response using the DATA0.FM6 database with $Am(OH)_3(am)$ as the solubility-controlling phase.

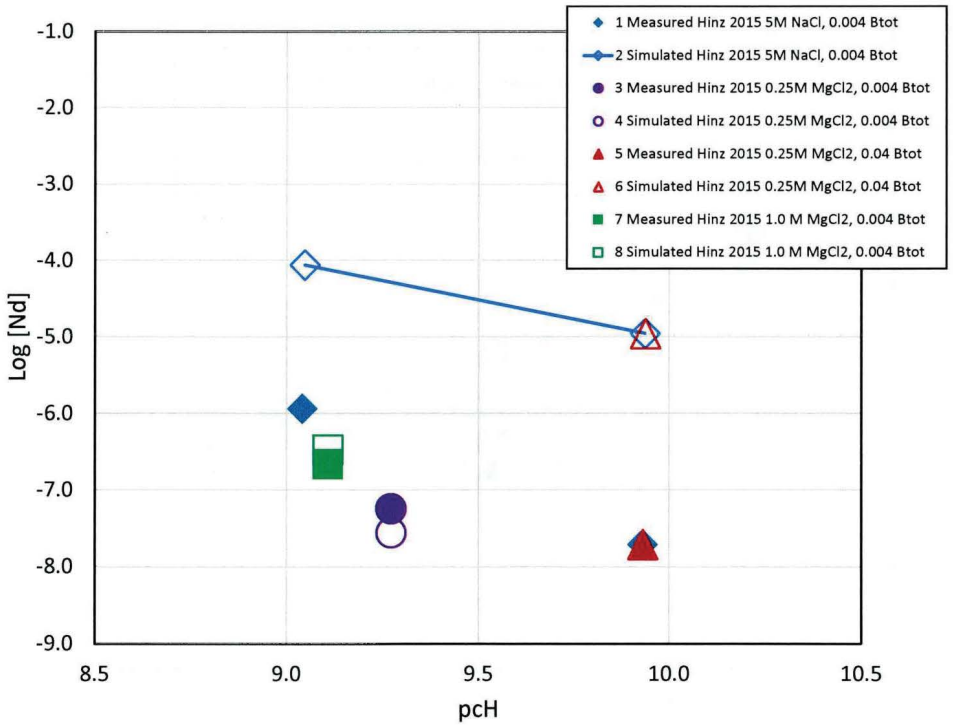


Figure A - 3. Graph of the Hinz et al. (2015) measured data and EQ3NR model response using the DATA0.FM6 database with $Am(OH)_3(am)$ as the solubility controlling phase.

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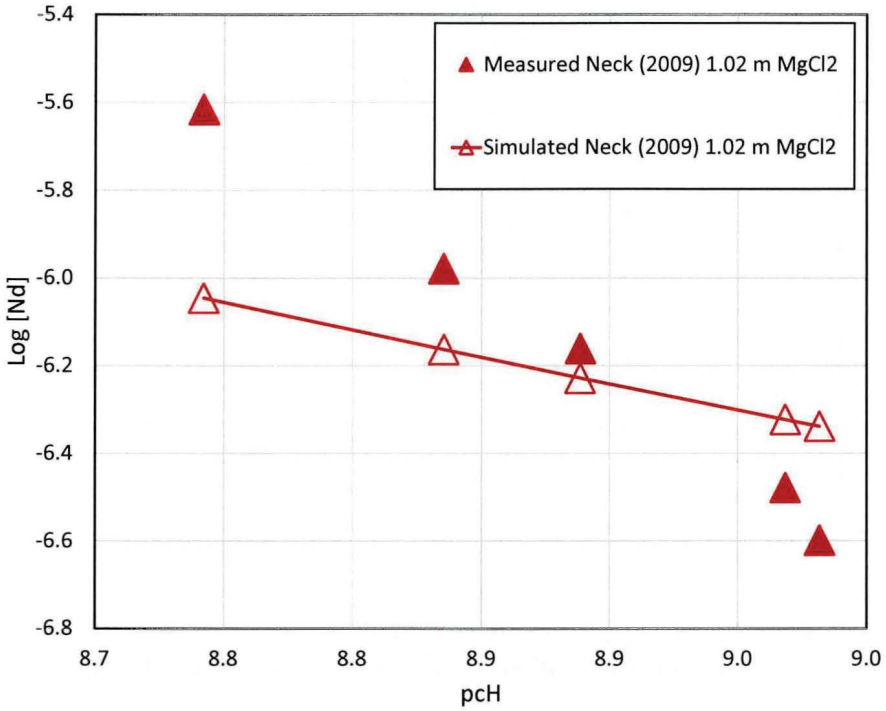


Figure A - 4. Graph of the Neck et al. (2009) measured data and EQ3NR model response using the DATA0.FM6 database with $Am(OH)_3(am)$ as the solubility controlling phase.

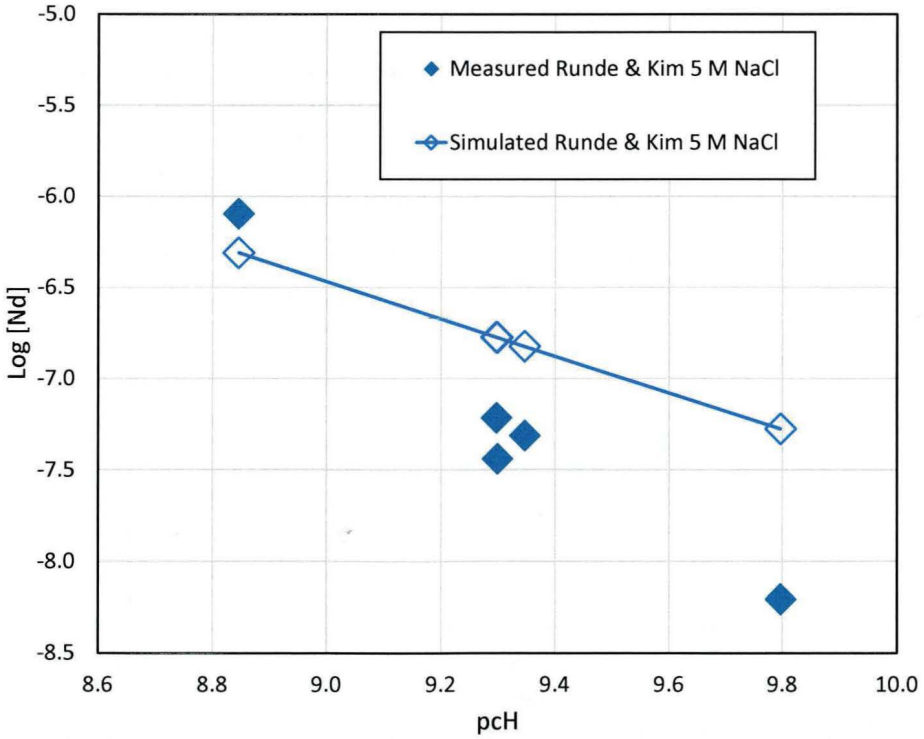


Figure A - 5. Graph of the Runde and Kim (1995) measured data and EQ3NR model response using the DATA0.FM6 database with $Am(OH)_3(s)$ as the solubility-controlling phase.

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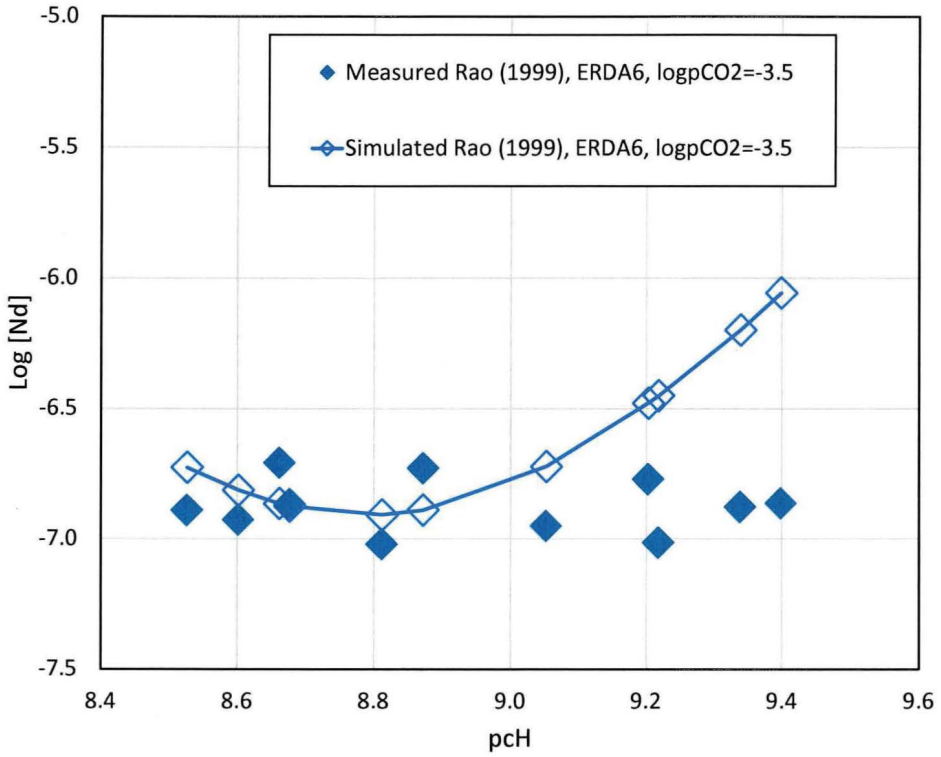


Figure A - 6. Graph of the Rao et al. (1999) measured data and EQ3NR model response using the DATA0.FM6 database with $NaAm(CO_3)_2.6H_2O(cr)$ as the solubility-controlling phase.

Results

Figures A – 1 – A – 6 show the WIPP model fits to the measured experimental data. Overall the visual fit of the WIPP model is subjectively fair compared to the measured data. By which it may be said that the WIPP model, both An(IV) and An(III), fits some data better than others, and taken as a whole, may be described, in the opinion of the author, as a "fair" overall fit to the experimental data.

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