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Sandia National Laboratories
Operated for the U.S. Department of Energy by
Sandia Corporation

4100 National Parks Highway
Carlsbad, NM 88220

Phone: (575) 234-0056
Fax: (575) 234-0061
Internet: psdomsk@sandia.gov

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to: SNL WIPP Records Center
Defense Waste Management Programs

from: Paul S. Domski Jenny Jones for Paul S. Domski 6/28/18

subject: Recommended Ranges of pcH, Ionic Strength, Total Inorganic Carbon (TIC), and Carbon Dioxide Fugacity for WIPP Baseline Solubility Uncertainty Calculations

Introduction

This memo examines 31 WIPP relevant brine compositions and uses EQ3/6 Version 8a and statistical methods to provide recommended ranges for pcH, ionic strength (m), total inorganic carbon (TIC), and carbon dioxide fugacity (f_{CO2}). Both unequilibrated and equilibrated brines from the Salado and Castile formations are considered. The conceptual model for the near-field chemistry at WIPP considers all brines to be in equilibrium with the primary Salado Formation mineral halite, and with the secondary mineral anhydrite. Based on the conceptual model all the equilibrated brine compositions from the baseline solubility calculations beginning with the CCA and up to and including the CRA-2014 meet the mineral saturation criteria, and are included in the analyses shown below. These brines are also equilibrated with MgO and the waste.

In addition, several other native Salado Formation brines (Brush, 1990, Deal et.al., 1991, and Roberts et. al., 1999) are included. The native brines are worth examining when trying to quantify uncertainty in chemical conditions because although the conceptual model for chemical conditions assumes a well-mixed repository at equilibrium, micro-environments are also a possibility. A micro-environment in this context is a section of the repository where fresh (unequilibrated) brine is in contact with the waste.

Methods

For the native brines, their compositions were input into EQ3NR and the code was executed using DATA0.FM1 and the resulting fCO_2, pH, I, and c_{TIC} values were used in the evaluation. The variables of interest were compiled in Microsoft Excel and their descriptive statistics (count, max, min, mean, median, and standard deviation) were calculated, based on these values it was possible to calculate, for plotting purposes, the probability density function, cumulative distribution function, quantile – quantile, and the probability – probability functions. The purpose of calculating and plotting these properties was to test the data for normal behavior. None of the parameters exhibited behavior typical of a normal distribution, therefore, it was not possible to assign ± 2σ rule to the mean to obtain the 95% confidence interval. Rather, the uncertainty ranges were assigned based on knowledge of the chemical system and expert opinion.

Results

Ionic Strength Results

Table 1 provides the ionic strength data and the data sources, Table 2 provides the descriptive statistics for the ionic strength (*Ionic_strgth.xlsx*)[1]. Figure 1 shows the probability density function (pdf) and the cumulative distribution function (CDF), and Figure 2 shows the probability – probability plot (P-P plot), and the quantile – quantile plot (Q-Q plot) for ionic strength. Based on Figures 1 and 2 it is apparent that the data do not obey a normal distribution because they do not fall close to the ideal normal distribution, therefore, it was not possible to assign ± 2σ rule to the mean to obtain the 95% confidence interval. Halite saturation is the greatest contributor to the ionic strength of these brines. All 31 brines that were examined are saturated with respect to halite, and also anhydrite. The observed variation in ionic strength from 5.9 to 8.2 molal is caused by the "ionic strength effect" of ions other than Na^+ and Cl^- in the brine, namely Mg^{2+}, SO_4^{2-}, and K^+. Thus, it can be argued, based on expert opinion, that the ionic strength range should be set at approximately +/- 0.5 molal around the maximum and minimum values. This would make a proposed ionic strength range of 5.5 to 9 m.

Table 1. Brine ionic strength data.

Brine Type	I, mol·kg^{-1}
ERDA-6 (unequilibrated)[A]	5.95
Union[A]	6.22
CCA (ERDA-6, Magnesite, without Organics, All Vectors)[B]	6.66
PAVT (ERDA-6, Hydromagnesite, without Organics,All Vectors)[D]	6.67
CRA-2004 PA (ERDA-6, Hydromagnesite, with Organics, Microbial Vectors)[E]	6.73
WIPP-12[A]	6.73
CRA-2009 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[G]	6.77
CRA-2004 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[F]	6.80
CRA-2014 PA (ERDA-$6_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	6.88
CCA (SPC,Magnesite, w/o Organics, All Vectors)[B]	7.40
PAVT (SPC,Hydromagnesite without Organics, All Vectors)[D]	7.41
G090[I]	7.42
H090[I]	7.44
CRA-2009 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[G]	7.52
CRA-2004 PA (GWB,Hydromagnesite, with Organics, Microbial Vectors)[E]	7.54
OH23[I]	7.55
0H45[I]	7.58
L4P51[J]	7.59
G Seep[A]	7.61
CRA-2014 PA (GWB$_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	7.64
DH36[A]	7.65
OH26[I]	7.66
CRA-2004 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[F]	7.66
DHP402a[I]	7.72
DH28[I]	7.74
DH30[I]	7.76
DH32[I]	7.79
0H20[I]	7.81
SB-1[A]	8.18
SB-3[A]	8.20
SB-2[A]	8.21

[A] Calculated for this document based on the data given in Tables 2.1, 2.2, 2.3 and 2.4 from Brush (1990); [B] Novak et al. (1996); [C]From Novak et al. (1996); [D]From Novak (1997); [E]From Brush and Xiong (2003a, 2003b, 2003c, 2003d) and U.S. DOE (2004, Appendix SOTERM; [F]Brush and Xiong (2005a, 2005b) and Brush (2005); [G]Brush et al. (2009); [H]Brush and Domski (2013); [I]Deal et al. (1991); [J]Roberts et al. (1999).

[1] files may found at: /nfs/data/CVSLIB/WIPP_EXTERNAL/APPENDIXGEOCHEM/Files

Table 2. Descriptive statistics for ionic strength.

Property	I (m)	Property	I (m)
n	31	Mean	7.37
Maximum	8.2	Median	7.55
Minimum	5.9	Standard deviation	0.56

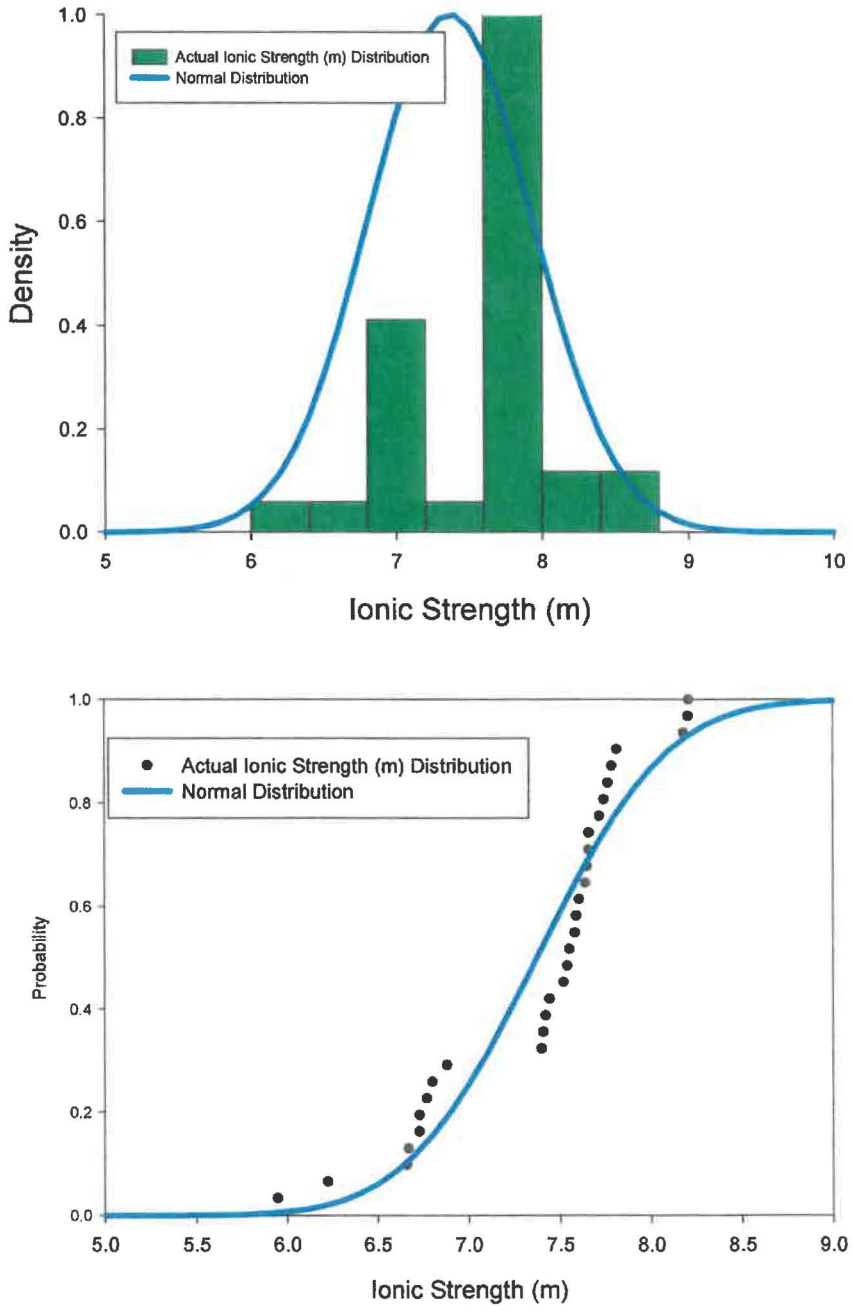


Figure 1. PDF and CDF plots for Ionic Strength.

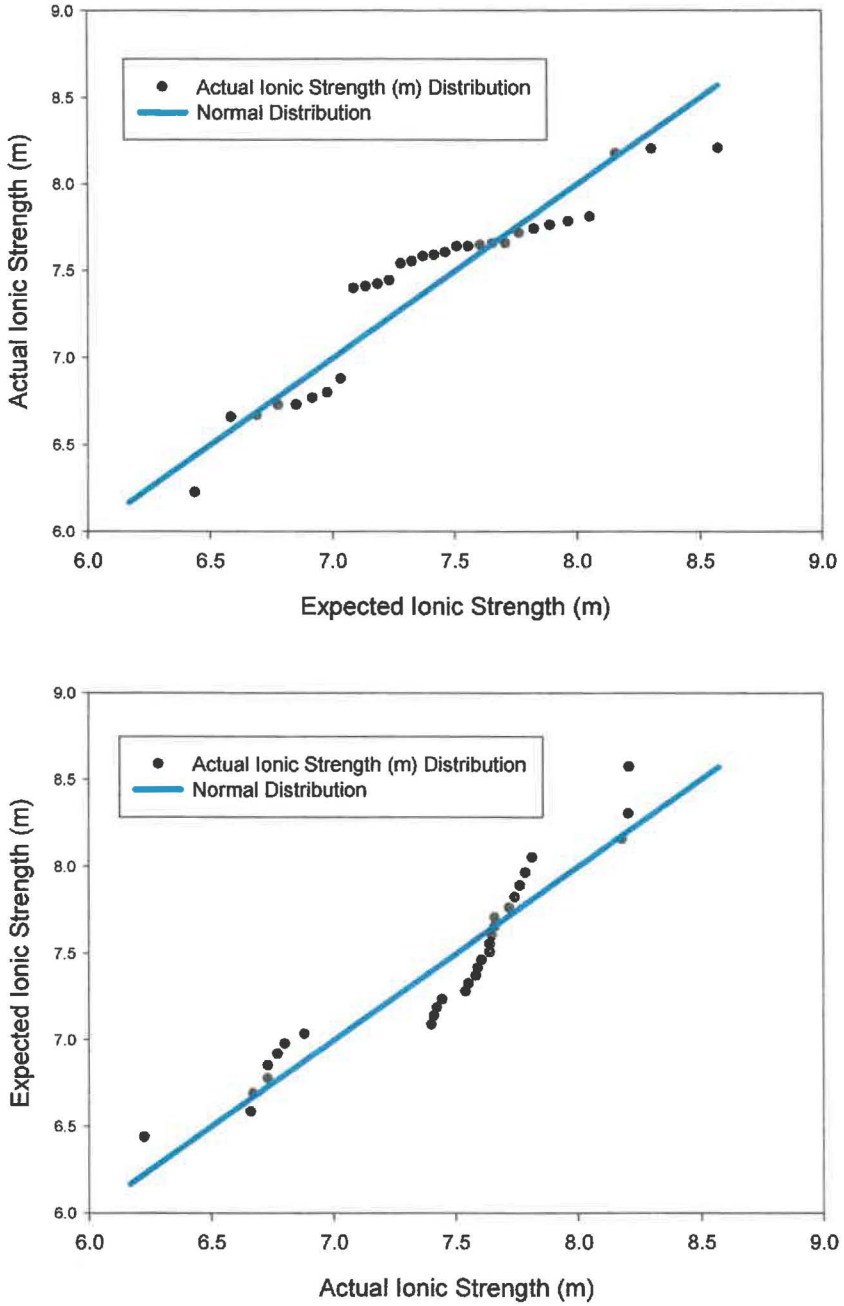


Figure 2. P-P and Q-Q plots for Ionic Strength.

pcH Results

Table 3 provides the pcH data and the data sources, Table 4 provides the descriptive statistics for the pcH (*pcH_stats.xlsx*)[2]. Figure 3 shows the probability density function (PDF) and the cumulative distribution function (CDF), and Figure 4 shows the probability – probability plot (P-P plot), and the quantile – quantile plot (Q-Q plot) for pcH. Based on Figures 3 and 4 it is apparent that the data do not obey a normal distribution because they do not fall close to the ideal normal distribution, therefore, it was not possible to assign ± 2σ rule to the mean to obtain the 95% confidence interval. The average pcH of the fresh brines is close to neutral at 7.3 and is buffered by borate and carbonate interactions with dissolved cations. The average pcH of the reacted brines is

[2] files may found at: /nfs/data/CVSLIB/WIPP_EXTERNAL/APPENDIXGEOCHEM/Files

9.6 which is controlled by the equilibrium between brucite and either magnesite, or hydromagnesite. These two groups of brines are representative of brines that will enter the repository in the case of fresh brines, and those which have entered and reacted with repository materials. Examination of the pcH data reveals a minimum of 6.87 and a maximum of 9.94, therefore, based on expert opinion application of approximately +/- 1 pcH unit is recommended for pcH, making the range $6.0 \leq pcH \leq 11.0$.

Table 3. Brine pcH data.

Brine Type	pcH
ERDA-6 (unequilibrated)[A]	7.01
Union[A]	6.87
CCA (ERDA-6, Magnesite, without Organics, All Vectors)[B]	9.94
PAVT (ERDA-6, Hydromagnesite, without Organics,All Vectors)[D]	9.94
CRA-2004 PA (ERDA-6, Hydromagnesite, with Organics, Microbial Vectors)[E]	9.72
WIPP-12[A]	8.06
CRA-2009 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[G]	9.68
CRA-2004 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[F]	9.64
CRA-2014 PA ($ERDA\text{-}6_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	9.69
CCA (SPC,Magnesite, w/o Organics, All Vectors)[B]	9.41
PAVT (SPC,Hydromagnesite without Organics, All Vectors)[D]	9.41
G090[I]	7.22
H090[I]	7.22
CRA-2009 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[G]	9.39
CRA-2004 PA (GWB,Hydromagnesite, with Organics, Microbial Vectors)[E]	9.40
OH23[I]	8.77
0H45[I]	7.09
L4P51[J]	7.15
G Seep[A]	7.15
CRA-2014 PA ($GWB_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	9.54
DH36[A]	7.10
OH26[I]	7.10
CRA-2004 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[F]	9.39
DHP402a[I]	7.10
DH28[I]	7.21
DH30[I]	7.21
DH32[I]	7.11
0H20[I]	7.13
SB-1[A]	7.28
SB-3[A]	7.27
SB-2[A]	7.37

[A] Calculated for this document based on the data given in Tables 2.1, 2.2, 2.3 and 2.4 from Brush (1990); [B] Novak et al. (1996); [C]From Novak et al. (1996); [D]From Novak (1997); [E]From Brush and Xiong (2003a, 2003b, 2003c, 2003d) and U.S. DOE (2004, Appendix SOTERM; [F]Brush and Xiong (2005a, 2005b) and Brush (2005); [G]Brush et al. (2009); [H]Brush and Domski (2013); [I]Deal et al. (1991); [J]Roberts et al. (1999).

Table 4. Descriptive statistics for pcH.

Property	pcH	Property	pcH
n	31	**Mean**	8.18
Maximum	9.94	**Median**	7.28
Minimum	6.87	**Standard deviation**	1.20

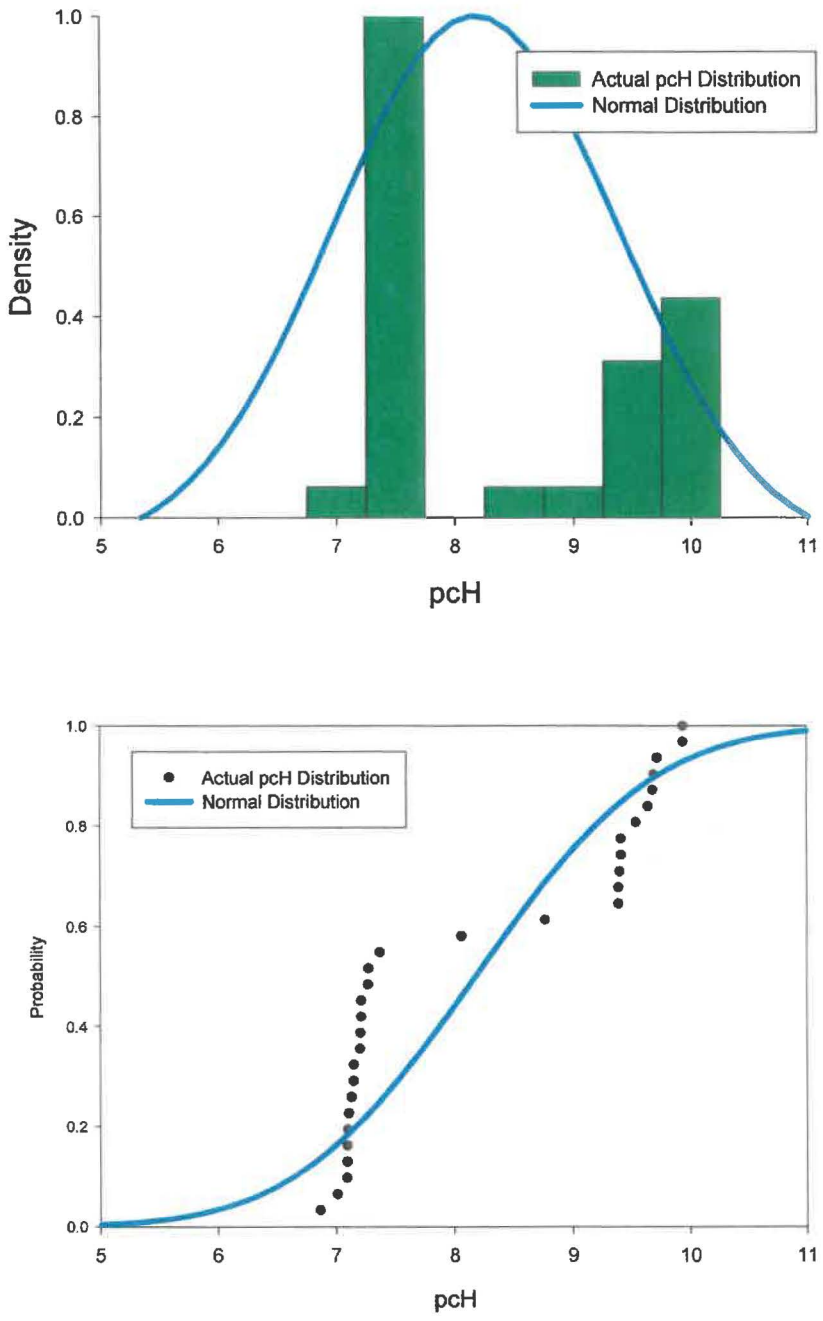


Figure 3. PDF and CDF for pcH.

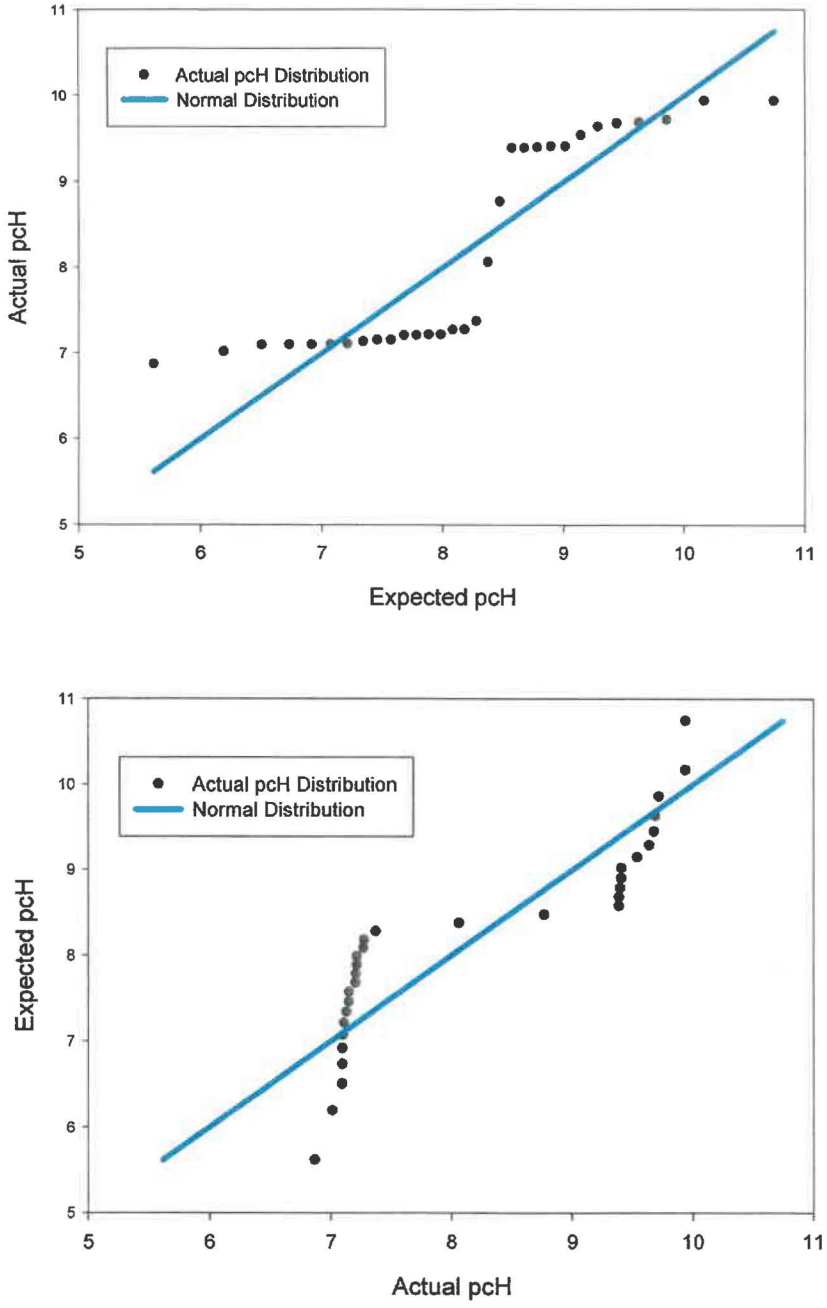


Figure 4. P-P and Q-Q plots for pcH

Total Inorganic Carbon (TIC) Results

Table 5 provides the TIC data and the data sources, Table 6 provides the descriptive statistics for the TIC (*TIC_stats.xlsx*)[3]. Figure 5 shows the probability density function (PDF) and the cumulative distribution function (CDF), and Figure 6 shows the probability – probability plot (P-P plot), and the quantile – quantile plot (Q-Q plot) for TIC. Based on Figures 5 and 6 it is apparent that the TIC data do not obey a normal distribution because they do not fall close to the ideal normal distribution, therefore, it was not possible to assign ± 2σ rule to the mean to obtain the 95% confidence interval. The total inorganic carbon is the total dissolved carbon fraction originating from all inorganic aqueous species. In other words it represents the sum of the primary carbonate

[3] files may found at: /nfs/data/CVSLIB/WIPP_EXTERNAL/APPENDIXGEOCHEM/Files

species, $CO_2(aq)$, HCO_3^- and CO_3^{2-}, as well as all of the carbonate that may be complexed with solution cations, primary among which, but not limited to, are $MgCO_3(aq)$ and $CaCO_3(aq)$. Comparison of the fresh brine TIC values with those of the reacted brines show TIC is greater for all of the reacted brines, maximum ~ 8×10^{-4} m, thus, this data determines the maximum of the range. The minimum TIC will be set to zero because of the necessity to include solubility studies in the actinide uncertainty analysis that did not include carbonate in the experiments. The range for TIC is set to $0 \leq TIC \leq 2 \times 10^{-2}$ m, the upper limit is about 25 times the maximum of the reacted brines TIC to account for uncertainty, and to ensure inclusion of the greatest number of studies in the actinide uncertainty analysis.

Table 5. TIC data.

Brine Type	TIC (m)
ERDA-6 (unequilibrated)[A]	1.80×10^{-5}
Union[A]	N/A
CCA (ERDA-6, Magnesite, without Organics, All Vectors)[B]	3.15×10^{-5}
PAVT (ERDA-6, Hydromagnesite, without Organics,All Vectors)[D]	7.77×10^{-4}
CRA-2004 PA (ERDA-6, Hydromagnesite, with Organics, Microbial Vectors)[E]	5.32×10^{-4}
WIPP-12[A]	6.39×10^{-6}
CRA-2009 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[G]	5.09×10^{-4}
CRA-2004 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[F]	4.86×10^{-4}
CRA-2014 PA (ERDA-$6_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	4.55×10^{-4}
CCA (SPC,Magnesite, w/o Organics, All Vectors)[B]	4.08×10^{-5}
PAVT (SPC,Hydromagnesite without Organics, All Vectors)[D]	4.13×10^{-4}
G090[I]	2.36×10^{-4}
H090[I]	1.80×10^{-4}
CRA-2009 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[G]	4.02×10^{-4}
CRA-2004 PA (GWB,Hydromagnesite, with Organics, Microbial Vectors)[E]	4.02×10^{-4}
OH23[I]	8.52×10^{-5}
0H45[I]	1.32×10^{-4}
L4P51[J]	1.25×10^{-4}
G Seep[A]	1.31×10^{-5}
CRA-2014 PA ($GWB_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	3.79×10^{-4}
DH36[A]	1.58×10^{-5}
OH26[I]	7.61×10^{-5}
CRA-2004 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[F]	4.02×10^{-4}
DHP402a[I]	9.45×10^{-5}
DH28[I]	1.41×10^{-4}
DH30[I]	5.58×10^{-5}
DH32[I]	9.49×10^{-5}
0H20[I]	6.71×10^{-5}
SB-1[A]	4.28×10^{-4}
SB-3[A]	N/A
SB-2[A]	N/A

[A] Calculated for this document based on the data given in Tables 2.1, 2.2, 2.3 and 2.4 from Brush (1990); [B] Novak et al. (1996); [C]From Novak et al. (1996); [D]From Novak (1997); [E]From Brush and Xiong (2003a, 2003b, 2003c, 2003d) and U.S. DOE (2004, Appendix SOTERM; [F]Brush and Xiong (2005a, 2005b) and Brush (2005); [G]Brush et al. (2009); [H]Brush and Domski (2013); [I]Deal et al. (1991); [J]Roberts et al. (1999).

Table 6. Descriptive statistics for TIC.

Property	c_{TIC} (m)	Property	c_{TIC}(m)
n	28	**Mean**	2.36×10^{-4}
Maximum	7.77×10^{-4}	**Median**	1.37×10^{-4}
Minimum	6.39×10^{-6}	**Standard deviation**	2.11×10^{-4}

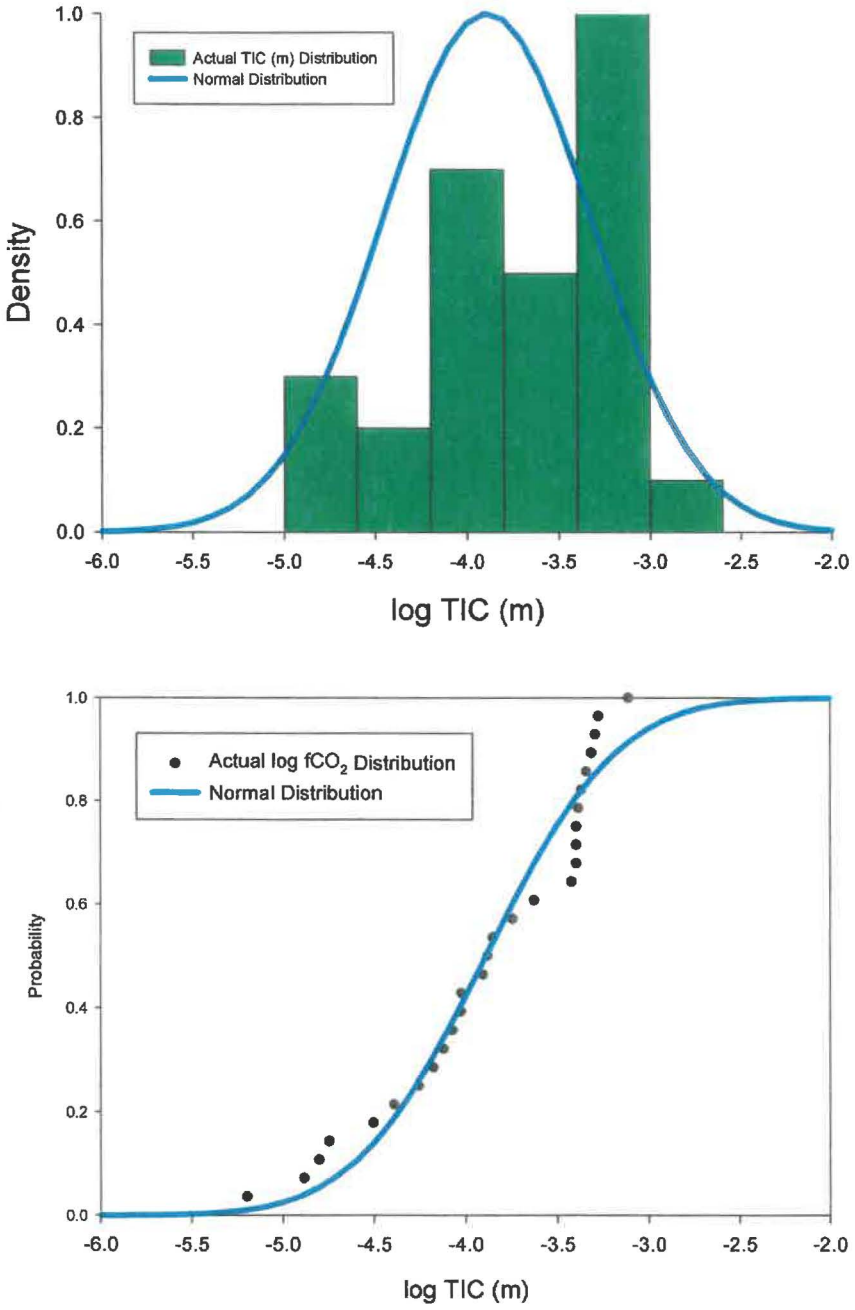


Figure 5. PDF and CDF for TIC.

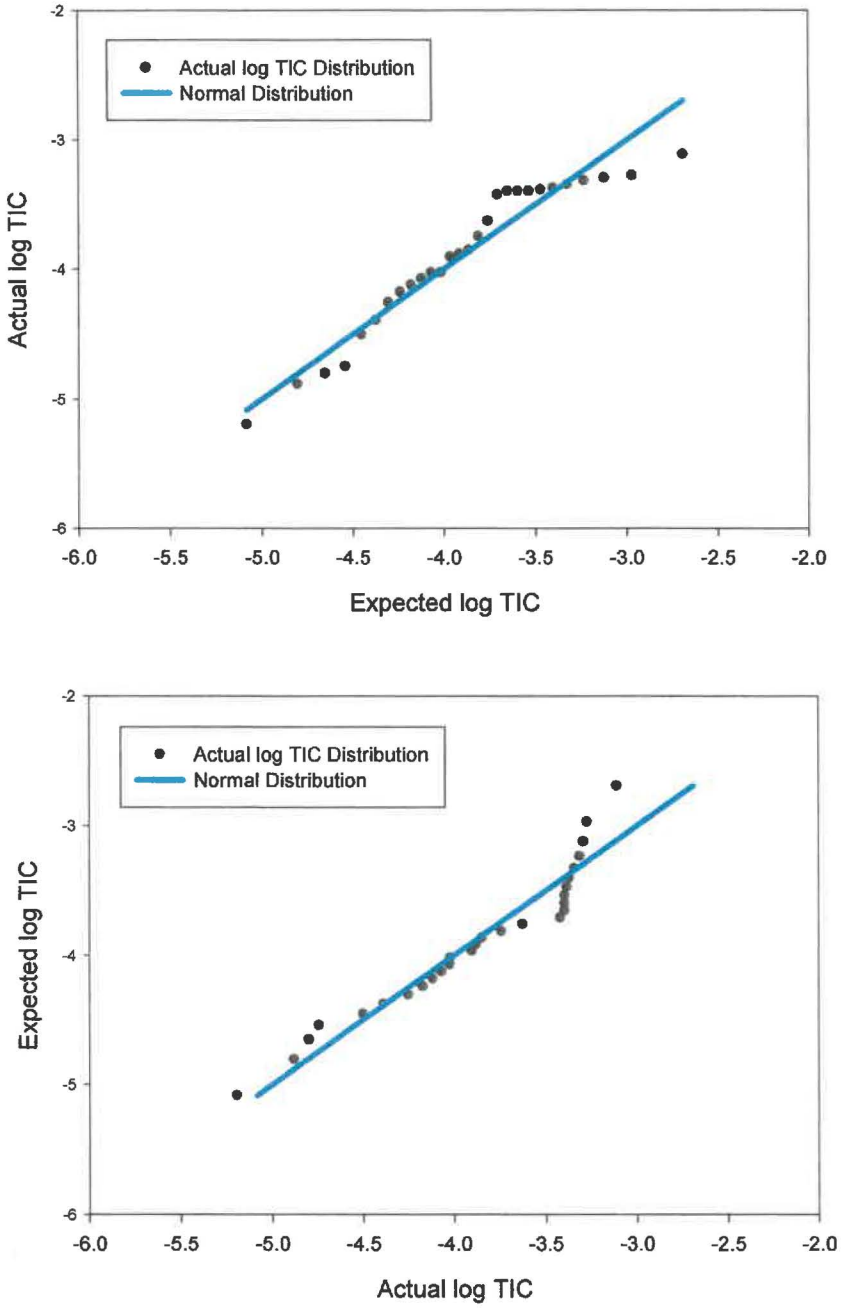


Figure 6. P-P and Q-Q plots for TIC.

Fugacity of CO_2 Results

Table 7 provides the fCO_2 data and the data sources, Table 8 provides the descriptive statistics for the fCO_2 (*fCO2_stats.xlsx*)[4]. Figure 7 shows the probability density function (PDF) and the cumulative distribution function (CDF), and Figure 8 shows the probability – probability plot (P-P plot), and the quantile – quantile plot (Q-Q plot) for fCO_2. Based on Figures 7 and 8 it is apparent that the data do not obey a normal distribution because they do not fall close to the ideal normal distribution, therefore, it was not possible to assign $\pm 2\sigma$ rule to the mean to obtain the 95% confidence interval. Table 8 clearly shows that there is a bi-modal distribution of fCO_2 values, the reacted brines have low values with a minimum of ~1 x 10^{-7} atm, while the native brines tend to

[4] files may found at: /nfs/data/CVSLIB/WIPP_EXTERNAL/APPENDIXGEOCHEM/Files

have high values of fCO_2, with the maximum at ~3 x 10^{-3} atm. The reacted brine values are low due the equilibrium between MgO, or it's hydrated form, brucite, and the carbonate phase hydromagnesite, and the native brines tend to have higher fCO_2 which reflects the conditions in the Salado Formation. The recommended range for fCO_2 is $0 \leq fCO_2 \leq 5 \times 10^{-3}$.

Table 7. CO_2 Fugacity data.

Brine Type	f_{CO_2} (atm)
ERDA-6 (unequilibrated)[A]	1.91×10^{-4}
Union[A]	N/A
CCA (ERDA-6, Magnesite, without Organics, All Vectors)[B]	1.29×10^{-7}
PAVT (ERDA-6, Hydromagnesite, without Organics,All Vectors)[D]	3.14×10^{-6}
CRA-2004 PA (ERDA-6, Hydromagnesite, with Organics, Microbial Vectors)[E]	3.16×10^{-6}
WIPP-12[A]	6.83×10^{-6}
CRA-2009 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[G]	3.14×10^{-6}
CRA-2004 PABC (ERDA-6, Hydromagnesite, with Organics, All Vectors)[F]	3.16×10^{-6}
CRA-2014 PA (ERDA-$6_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	3.14×10^{-6}
CCA (SPC,Magnesite, w/o Organics, All Vectors)[B]	1.29×10^{-7}
PAVT (SPC,Hydromagnesite without Organics, All Vectors)[D]	3.14×10^{-6}
G090[I]	1.76×10^{-3}
H090[I]	1.32×10^{-3}
CRA-2009 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[G]	3.14×10^{-6}
CRA-2004 PA (GWB,Hydromagnesite, with Organics, Microbial Vectors)[E]	3.16×10^{-6}
OH23[I]	4.98×10^{-6}
0H45[I]	1.41×10^{-3}
L4P51[J]	1.10×10^{-3}
G Seep[A]	1.14×10^{-4}
CRA-2014 PA (GWB$_{min.\ vol.}$, Hydromagnesite, with Organics, All Vectors)[H]	3.14×10^{-6}
DH36[A]	1.63×10^{-4}
OH26[I]	7.82×10^{-4}
CRA-2004 PABC (GWB, Hydromagnesite, with Organics, All Vectors)[F]	3.16×10^{-6}
DHP402a[I]	9.46×10^{-4}
DH28[I]	1.14×10^{-3}
DH30[I]	4.49×10^{-4}
DH32[I]	9.45×10^{-4}
0H20[I]	6.78×10^{-4}
SB-1[A]	3.12×10^{-3}
SB-3[A]	N/A
SB-2[A]	N/A

[A] Calculated for this document based on the data given in Tables 2.1, 2.2, 2.3 and 2.4 from Brush (1990); [B] Novak et al. (1996); [C]From Novak et al. (1996); [D]From Novak (1997); [E]From Brush and Xiong (2003a, 2003b, 2003c, 2003d) and U.S. DOE (2004, Appendix SOTERM; [F]Brush and Xiong (2005a, 2005b) and Brush (2005); [G]Brush et al. (2009); [H]Brush and Domski (2013); [I]Deal et al. (1991); [J]Roberts et al. (1999).

Table 8. Descriptive statistics for fCO_2

Property	f_{CO_2} (atm)	Property	f_{CO_2} (atm)
n	28	**Mean**	5.06×10^{-4}
Maximum	3.12×10^{-3}	**Median**	6.05×10^{-5}
Minimum	1.29×10^{-7}	**Standard deviation**	7.46×10^{-4}

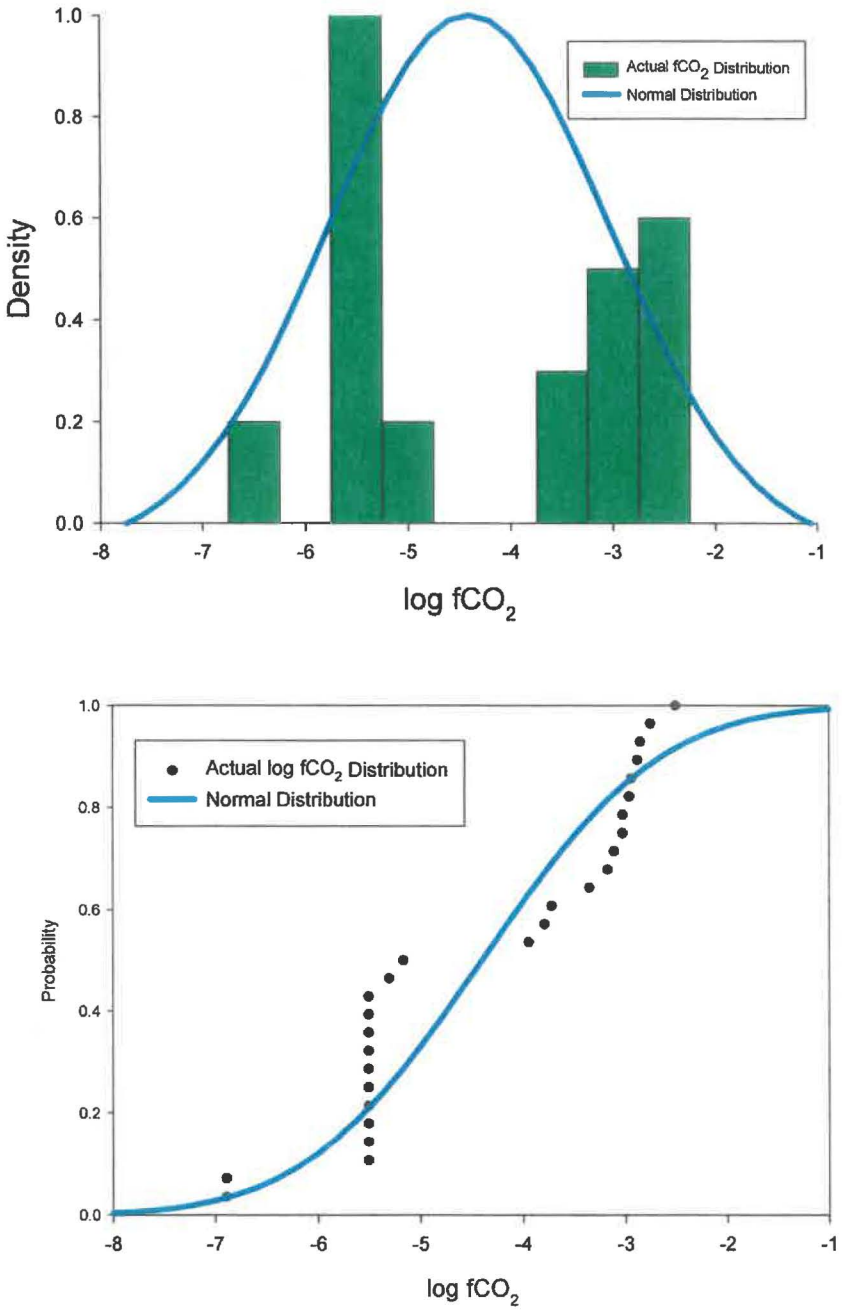
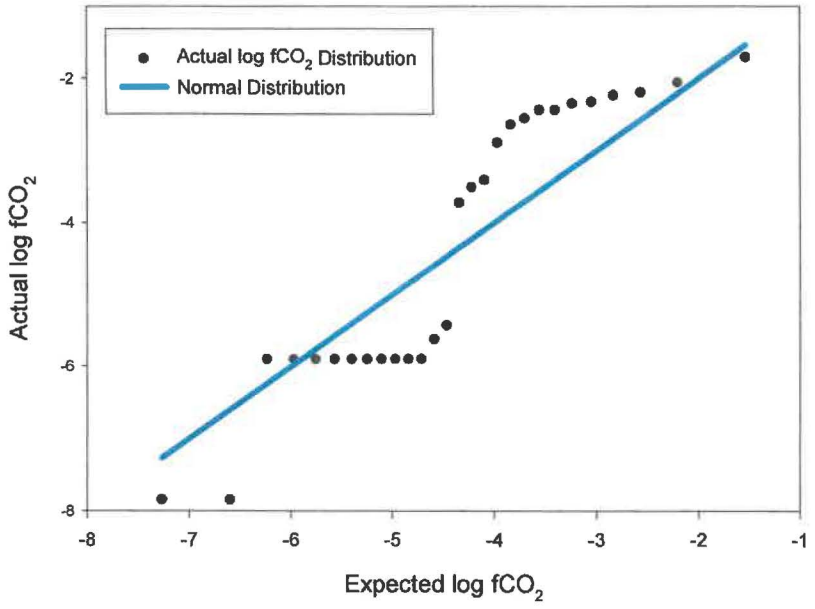


Figure 7. PDF and CDF for fCO_2.



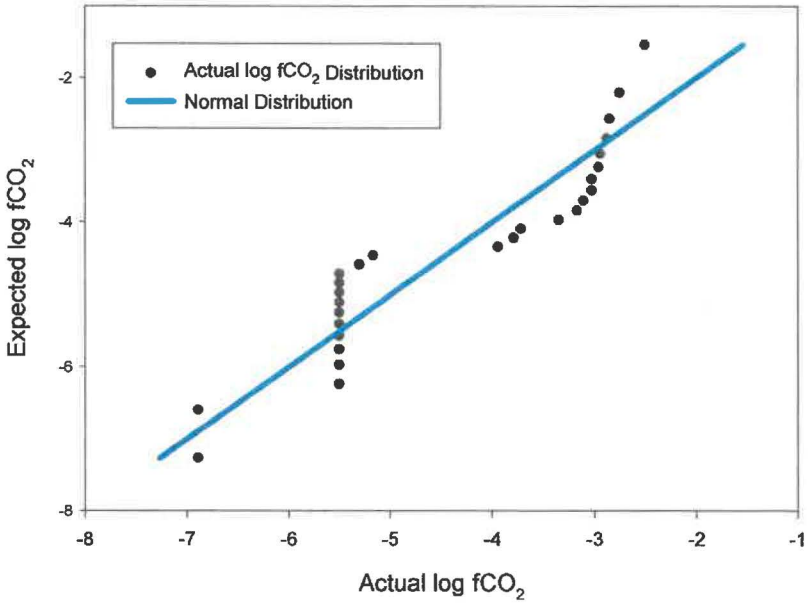


Figure 8. Q-Q and P-P plots for fCO_2.

Summary

Table 9. Summary of recommended parameter ranges.

Property	Minimum	Maximum
Ionic Strength	5.5 molal	9.0 molal
pcH	6.0	11.0
Total Inorganic Carbon	0.0 molal	2×10^{-2} molal
CO_2 Fugacity	0.0 atm	5×10^{-3} atm

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