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Scientific Notebook #252
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20-1402-871

KTI on Radionuclide Transport

01/13/98

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Comments

David R. Turner / Senior Research Scientist / CNWRA

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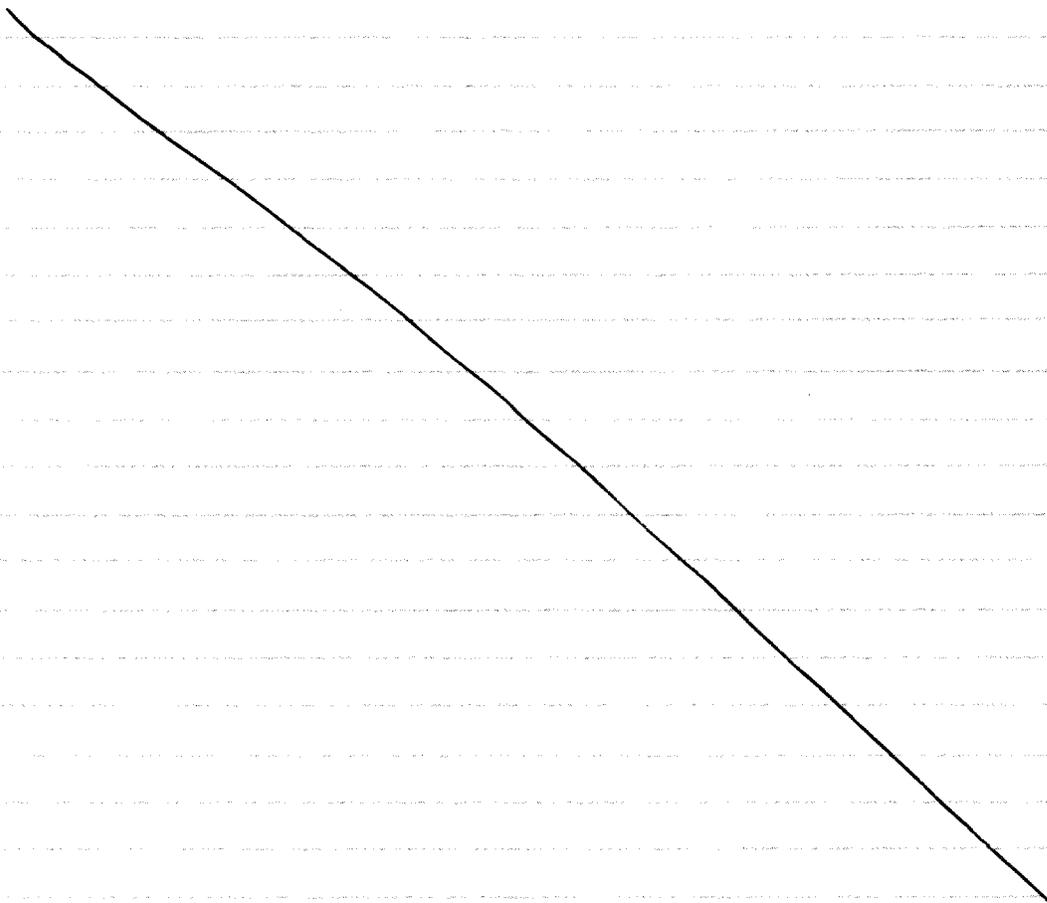
FIRST ENTRY

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Objectives (20-1402-871) - KTI on Radionuclide Transport (01/13/98)

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The purpose of the research discussed in this scientific notebook is to evaluate available geochemical, geological, and hydrologic data in the vicinity of Yucca Mountain to help constrain radionuclide transport from the proposed repository. Uses of these data may include delineation of regional flow patterns, placing constraints on the potential for dilution of radionuclide-bearing waters through mixing between different bodies of groundwater, geochemical information related to retardation (sorption, precipitation, dissolution). Computer codes that may be used include the EPA geochemistry code MINTEQA2, the USGS hydrochemistry code NETPATH, GIS software such as ArcView, and commercial software such as Microsoft Excel, SigmaPlot, and WordPerfect.



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Culling the Perfect et al. (1995) database

A comprehensive source of water chemistry data is found in the USGS report of Perfect et al. (1995).

Perfect, D.L., C.C. Faunt, W.C. Steinkampf, and A.K. Turner. 1995. *Hydrochemical Data Base for the Death Valley Region, California and Nevada*. USGS Open-File Report 94-305. Denver, CO: U.S. Geological Survey.

This report includes compressed Lotus 1-2-3 (*.WK1) files with major and minor element analyses compiled over several decades for the region surrounding Yucca Mountain. One contains the raw data for over 4700 wells and springs from USGS and DOE reports and the USGS National Water Information Service (NWIS) database. A second file has been edited to remove duplicates, make chemical data entries consistent, and calculate charge balance. The "editing" philosophy used by Perfect et al. (1995) is described in the report. These data were not generally collected under a DOE-approved QA program, but are freely used here. The sources referenced in Perfect et al. (1995) should be consulted for determining the quality of the data.

The entire edited database (DATAEDIT.WK1) of Perfect et al. (1995) includes 3733 groundwater analyses. It is necessary to cull these data and develop a more refined database for geochemical models. Based on criteria presented in:

Hitchon, B. and M. Brulotte. 1994. Culling criteria for "standard" formation water analyses. *Applied Geochemistry* 9: 637-645.

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the following series of steps were taken to eliminate water analyses from the entire database. The number of analyses eliminated are also listed.

Elimination Criteria	Number of analyses eliminated	Remaining analyses (from 3733)
No pH; pH <5 or pH >10	1708	2025
No reported Ca	121	1904
No reported Mg	127	1777
No reported Cl	9	1768
No reported SO ₄	21	1747
No reported Na	44	1703
No reported HCO ₃ or CO ₃	405	1298
Not charge balanced (± 10 %)	23	1275
No collection date reported	82	1193
Collected before 1960	165	1027
Mg > Ca	72	956

It is important to note that these criteria are not mutually exclusive, and the number of analyses eliminated for a given step is dependent on the order in which the steps were taken. For example, Perfect et al. (1995) report that 1373 records did not charge balance to within 10 percent. These include analyses without reported pH values as well as analyses missing major elements such as Ca, Mg, Na, and Cl. If charge balance had been the first criterion used, the number of analyses eliminated by the other criteria would have been significantly fewer.

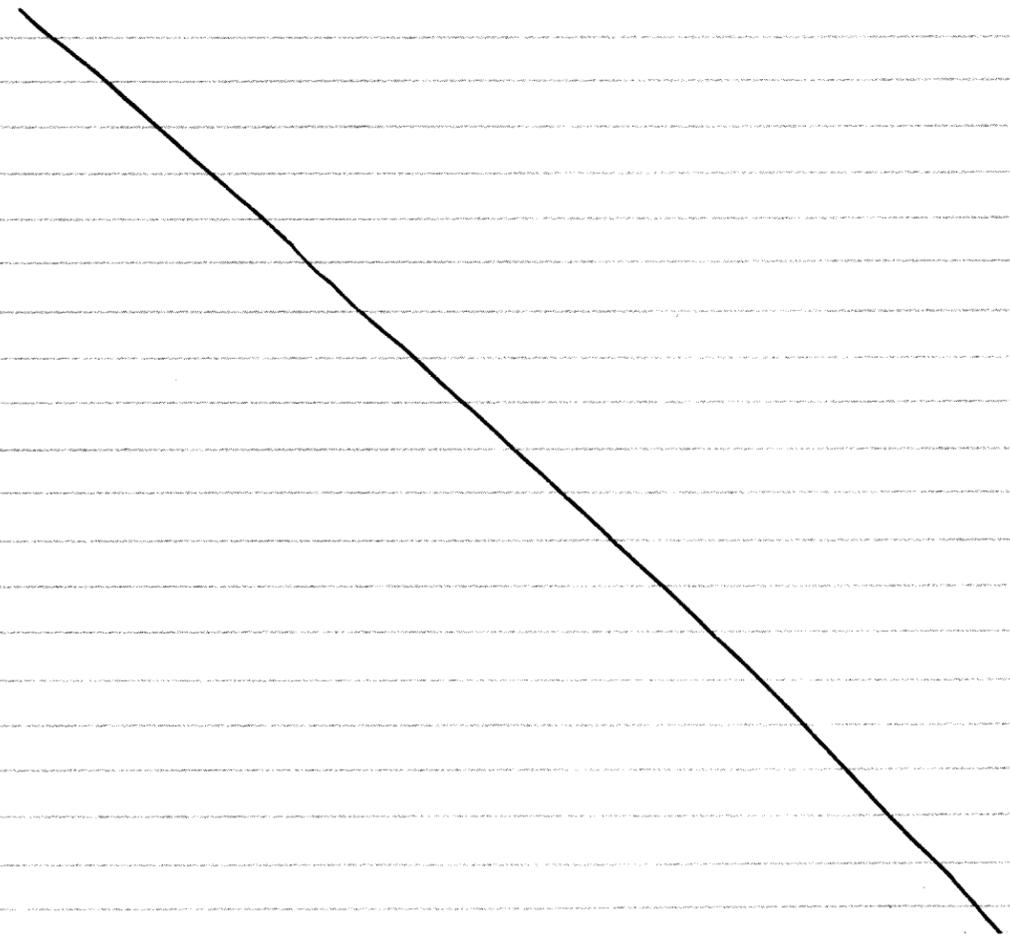
An additional criteria that can be used is to limit the study to those waters within the YM vicinity. A 100 km x 100 km area is considered, with UTM coordinates from 4,000,000N to 4,100,000N and 500,000E to 600,000E. This eliminates an additional 498 analyses for a remaining total of 458. These include multiple analyses for the same well/spring, and no attempt has yet been made to select a preferred analysis or calculate a mean water composition for a given sampling point.

The culled databases has been saved as a Microsoft Excel 5.0 file called PERFCO1.XLS and is included on a 3 1/2-inch floppy disk in the envelop at the back of this scientific notebook.

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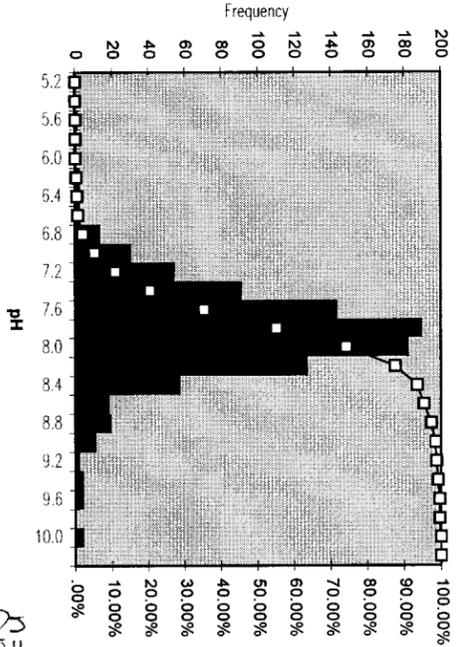
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Bin	Frequency	umulative %
5.2	0	.00%
5.4	0	.00%
5.6	1	.10%
5.8	0	.10%
6.0	0	.10%
6.2	2	.31%
6.4	2	.52%
6.6	2	.73%
6.8	13	2.09%
7.0	30	5.24%
7.2	54	10.89%
7.4	91	20.42%
7.6	143	35.39%
7.8	189	55.18%
8.0	182	74.24%
8.2	127	87.54%
8.4	57	93.51%
8.6	18	95.39%
8.8	19	97.38%
9.0	11	98.53%
9.2	2	98.74%
9.4	4	99.16%
9.6	4	99.58%
9.8	0	99.58%
10.0	4	100.00%
More	0	100.00%

Column1	Mean	Standard E	Median	Mode	Standard D	Sample Va	Kurtosis	Skewness	Range	Minimum	Maximum	Sum	Count	Confidence
Column1	7.801527	0.015672	7.8	7.8	0.484571	0.234809	2.669227	0.473264	4.4	5.6	10	7458.26	956	0.030756



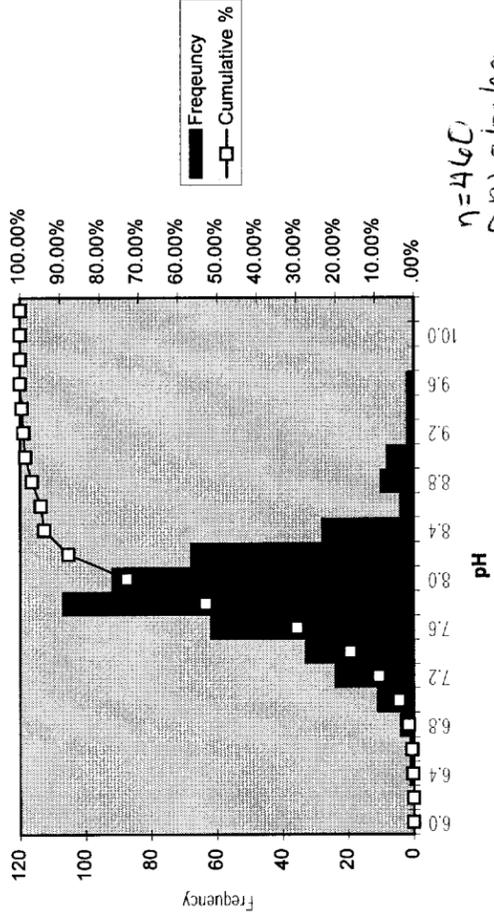
n=956
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Statistics calculated @ 95% Conf. by Excel 5.0

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pH - Saturated Zone (YM Vicinity)



n=460
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Bin	Frequency	umulative %
6.0	0	.00%
6.2	0	.00%
6.4	1	.22%
6.6	1	.44%
6.8	4	1.31%
7.0	11	3.70%
7.2	22	8.93%
7.4	33	12.19%
7.6	62	29.63%
7.8	107	52.94%
8.0	92	72.98%
8.2	68	87.80%
8.4	28	93.90%
8.6	4	94.77%
8.8	10	96.95%
9.0	8	98.69%
9.2	2	99.13%
9.4	2	99.56%
9.6	2	100.00%
9.8	0	100.00%
10.0	0	100.00%
More	0	100.00%

Column1	Mean	Standard E	Median	Mode	Standard D	Sample Va	Kurtosis	Skewness	Range	Minimum	Maximum	Sum	Count	Confidence
Column1	7.832609	0.020773	7.8	7.8	0.445533	0.1985	1.753896	0.433867	3.3	6.3	9.6	3603	460	0.040822

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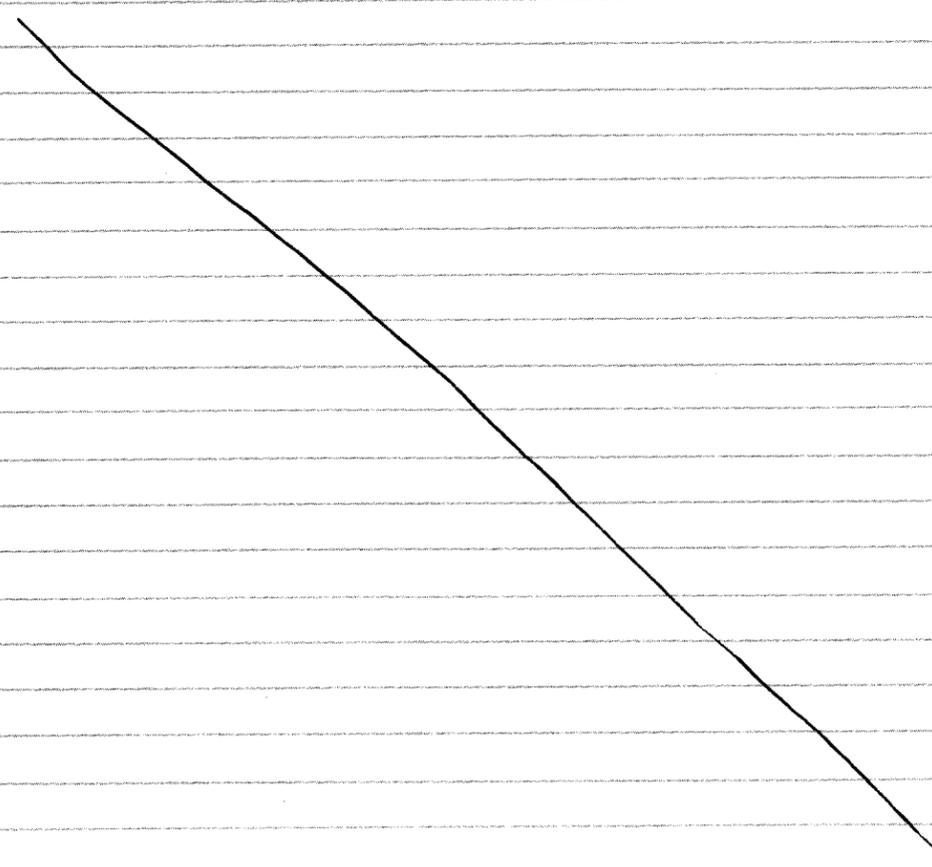
Saturation Indices -

SI Calculated using MINTEQA2, Ver 3.11 with the CNURA radionuclide database (Turner, 1995).

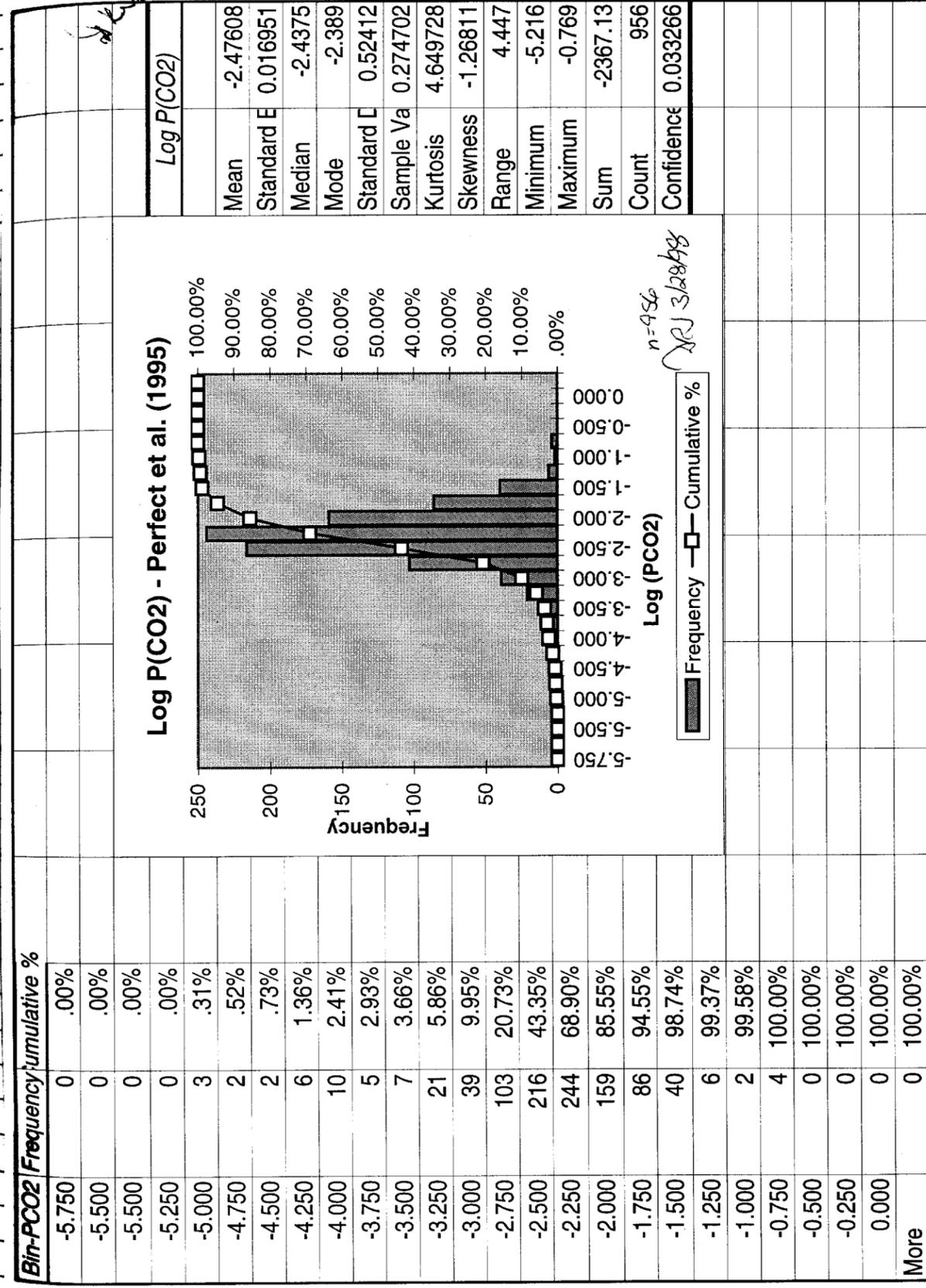
Turner, D.R. 1995. A Uniform Approach to Surface Complexation Modeling of Radionuclide Sorption. CNURA 95-001. San Antonio, TX: CNURA.

Statistics calculated using Excel, Ver 5.0 Data Analysis Tools. Histograms plotted using Excel spreadsheet.

Excel Files stored on a 3 1/2-inch floppy disk in an envelope at the end of this scientific notebook

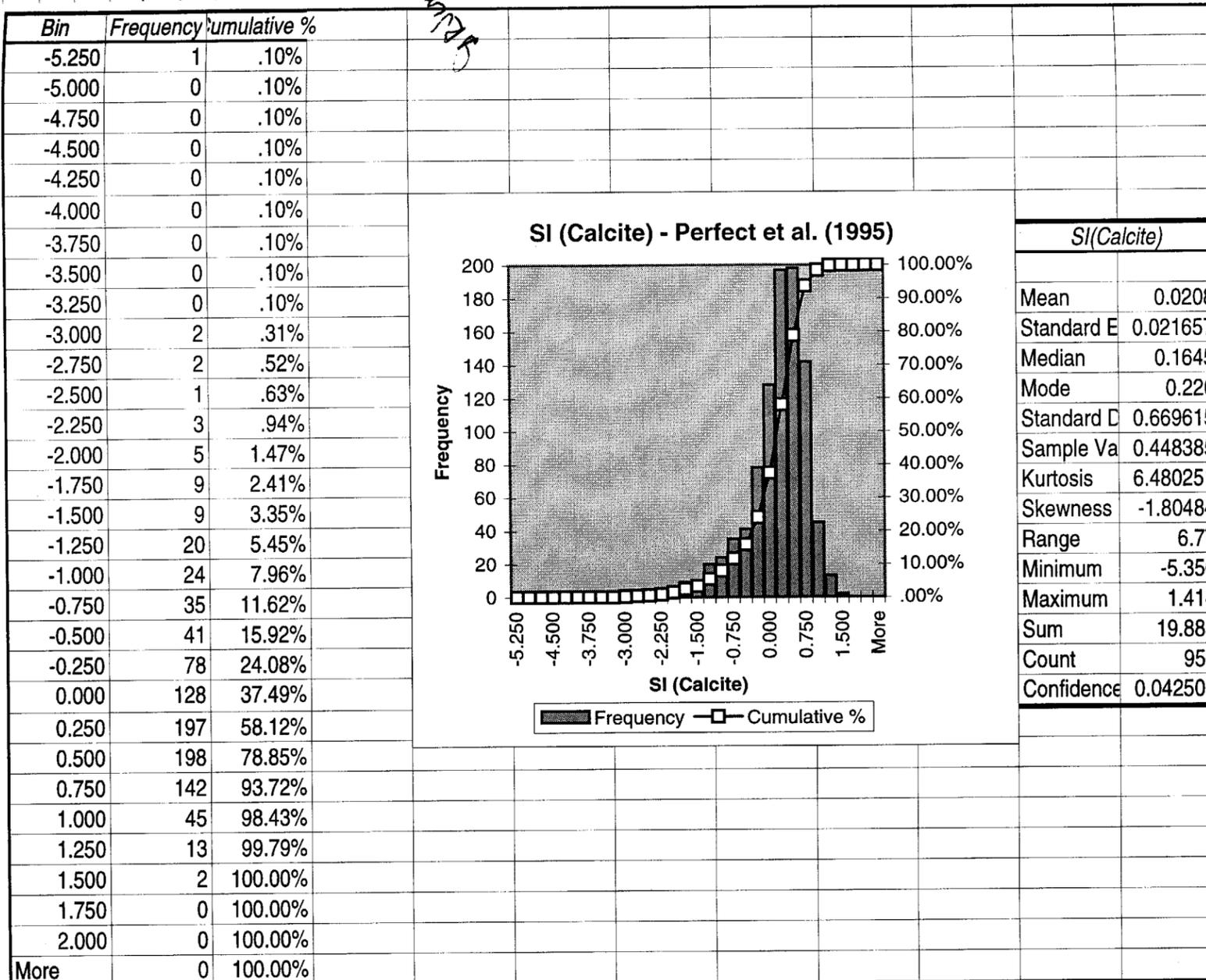
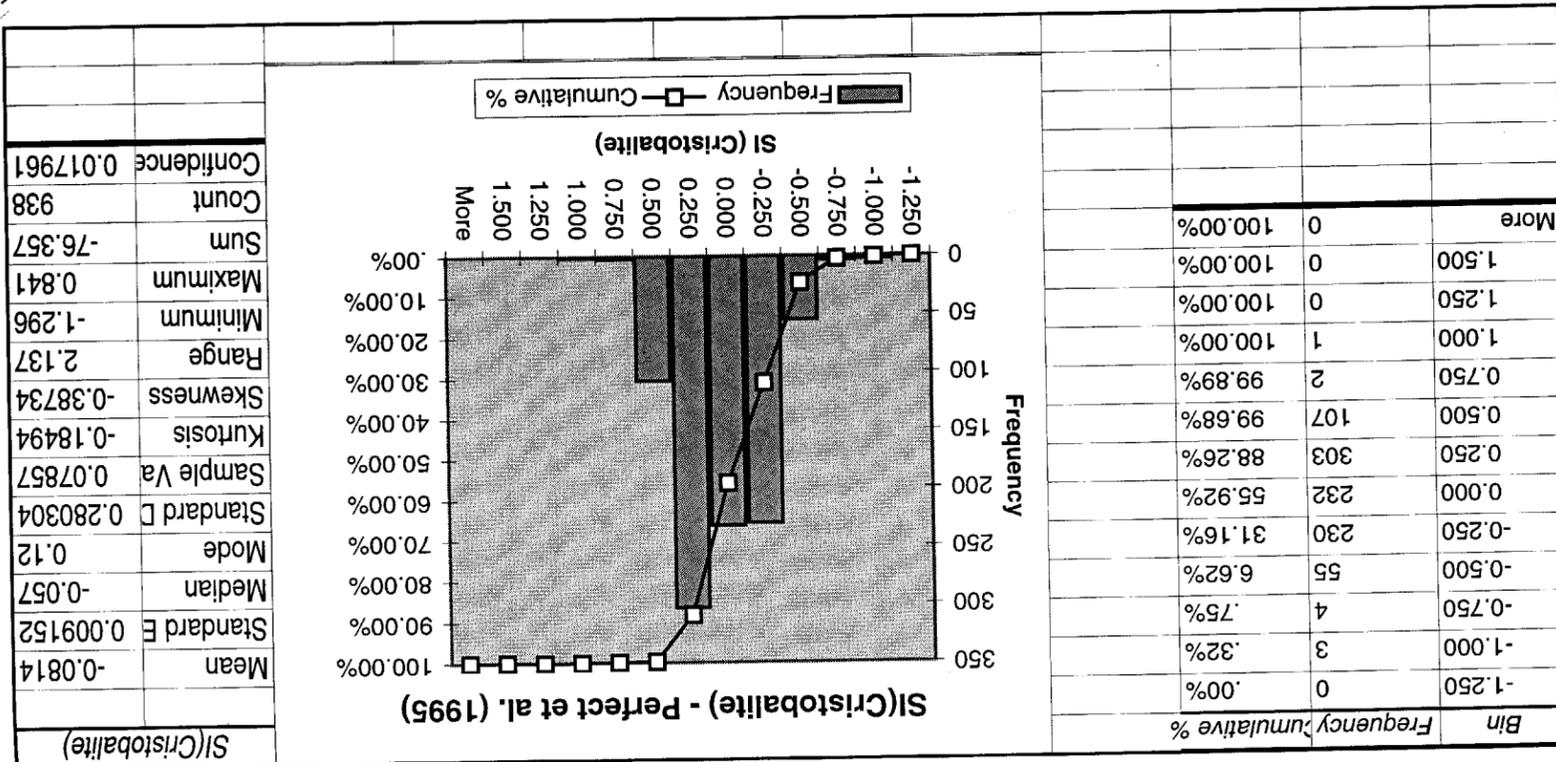


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Other geochemical parameters from Perfect et al. (1995)
Ym vicinity (n=460). SZ groundwater

Table 2. Descriptive statistics of measured groundwater chemical parameters

	pH (standard units)	C _{Total} (mg/L)	SiO ₂ (mg/L)
Mean	7.83	295.76	43.21
Standard Error	0.02	24.52	0.99
Median	7.8	245.0	43.0
Mode	7.8	300.0	22.0
Standard Deviation	0.45	525.99	21.25
Sample Variance	0.20	276665.20	451.61
Kurtosis	1.75	270.67	-1.30
Skewness	0.43	15.03	0.14
Range	3.3	10133.20	78.30
Minimum	6.3	6.80	3.70
Maximum	9.6	10140.0	82.0
Sum	3603.0	136051.80	19747.99
Count	460	460	457
Confidence Level(95.0%)	0.04	48.19	1.95

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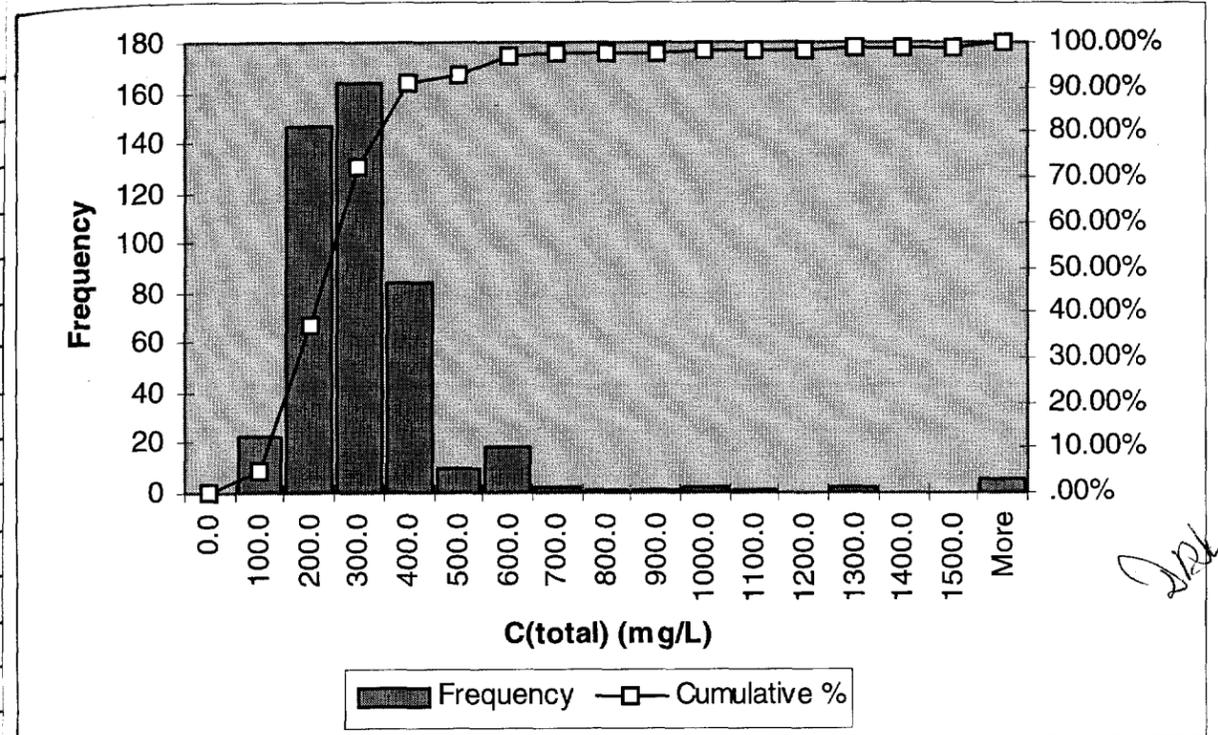


Figure 2. Distribution of total carbon (C_T in mg/L) for saturated zone regional groundwaters (from Perfect et al., 1995; see text for details).

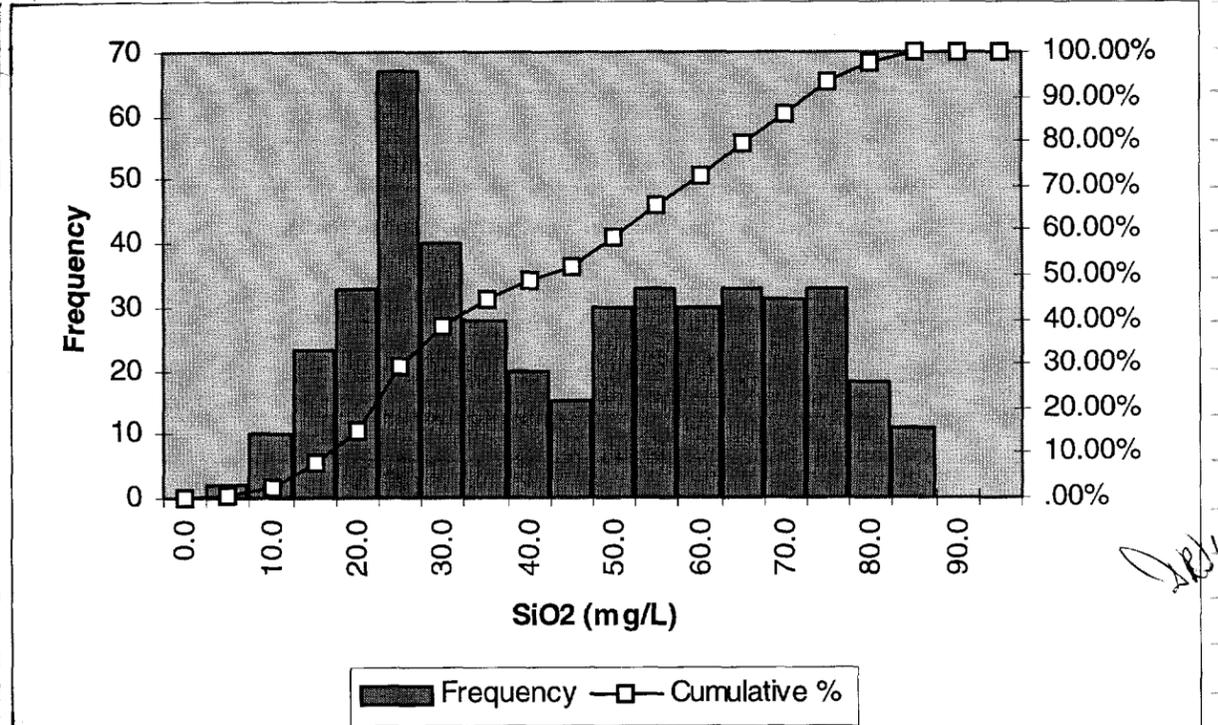


Figure 3. Distribution of SiO₂ (in mg/L) for saturated zone regional groundwaters (from Perfect et al., 1995; see text for details).

Calculated Transport Properties

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Colloidal Particle Concentrations

If actinides and other radioelements can sorb onto natural colloids, the stability of the particles in suspension is of critical importance in colloid-mediated transport. The stability of the colloidal suspension of charged particles varies as a function of ionic strength, groundwater chemistry, and pH. For example, at low ionic strengths, the electrostatic double-layer (EDL) expands outward from particle surfaces, stabilizing the colloids in solution through electrostatic repulsion. At higher ionic strengths, the double layer collapses, and the charged particles begin to flocculate (agglomerate) and come out of suspension due to gravity settling and filtration. Variations in overall solution chemistry (pH, component concentrations) and moisture content of the medium influence the magnitude of the ionic strength effect.

With the major and minor elements provided in the analyses of Perfect et al. (1995), calculated equilibrium ionic strength ranges from to 4.34×10^{-4} molal in the most dilute groundwaters to about 1.3 molal for brines collected in playa deposits. The upper limit should be considered only as approximate since the Davies equation activity coefficient used by MINTEQA2 is not strictly applicable at ionic strength greater than about 0.5 molal (Allison et al., 1991). Most of the groundwaters are dilute, however, with only 3 out of 460 samples exceeding 0.5 molal. The distribution of the ionic strength is approximately log-normal, with a mean value of about 8×10^{-3} molal.

Descriptive statistics of calculated groundwater chemical parameters

	Log (Equil. Ionic Strength) (molal)
Mean	-2.09
Standard Error	0.02
Median	-2.07
Mode	-1.99
Standard Deviation	0.39
Sample Variance	0.15
Kurtosis	8.07
Skewness	1.09
Range	3.48
Minimum	-3.36
Maximum	0.12
Sum	-962.54
Count	460
Confidence Level(95.0%)	0.04

Assuming that ionic strength is the principal control on colloid stability, Triay (1998) presented a relationship between colloid particle concentration (particles/mL) and ionic strength (molal), expressed as:

$$\log[\text{coll}] \text{ (particles/mL)} = 7.0 - 34 * I.S. \text{ (molal)} \quad [1]$$

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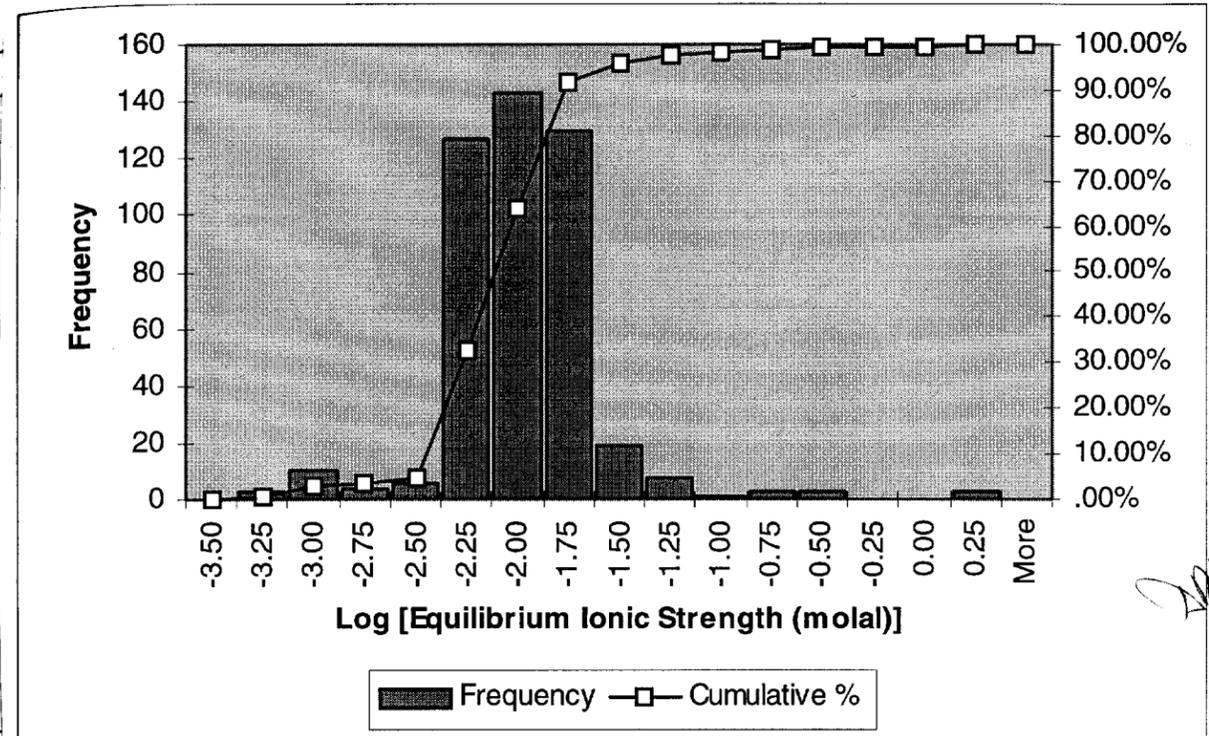


Figure 4. Distribution of the logarithm of the equilibrium ionic strength for saturated zone regional groundwaters (from Perfect et al., 1995; see text for details) calculated using MINTEQA2 (Allison et al., 1991).

This empirical relationship is based on observed colloid particle concentrations and groundwater ionic strength for field sites in a variety of rock/water systems throughout the world. Applying the relationship in Equation [1] with the calculated equilibrium ionic strength for groundwaters in the YM vicinity results in a predicted particle concentration distribution. For example, the mean predicted particle concentration is about $10^{6.3}$ and the median value is about $10^{6.7}$ particles/mL, comparable to the particle concentration of $10^{6.6}$ particles/mL reported for the 100- to 500-nm size fraction in J-13 well water (Triay et al., 1996b). The total predicted range in colloid concentrations is very large, but the distribution is skewed negatively towards the upper end of the range. For example, the maximum estimated particle is about $10^{7.0}$ particles/mL, slightly greater than the mean/median, but many orders of magnitude greater than the estimated minimum ($<10^{-30}$ particles/mL). This narrow range for the bulk of the samples is a direct consequence of the relatively restricted range in equilibrium ionic strength. The relatively few higher ionic strength groundwaters (<10 analyses) result in the very small colloid concentrations.

It is important to remember that the predicted distribution shown is based on a simplified empirical relationship between groundwater chemistry and colloid particle concentrations, and is restricted to only one chemical parameter, ionic strength. Other parameters that may have an effect on $\log[\text{coll}]$, such as pH and relative concentrations of alkali (Na^+) and alkaline earth (Ca^{2+}) elements (Degueldre et al., 1996; Degueldre, 1997), are not included in this analysis. Additional investigation to characterize the relationship between colloid stability and other chemical parameters is necessary to make a more quantitative estimate of $\log[\text{coll}]$. Finally, the estimate of particle concentration is only part of the information needed to evaluate radionuclide colloid transport. Particle size distribution and surface area estimates are necessary to calculate the amount of radionuclides that can be loaded onto the colloid particles and the efficiency with which they are transported through the YM fracture system. The information shown should be considered as a qualitative estimate of the range and distribution type that may be observed for particle concentrations, and therefore colloid stability, in the groundwaters near YM.

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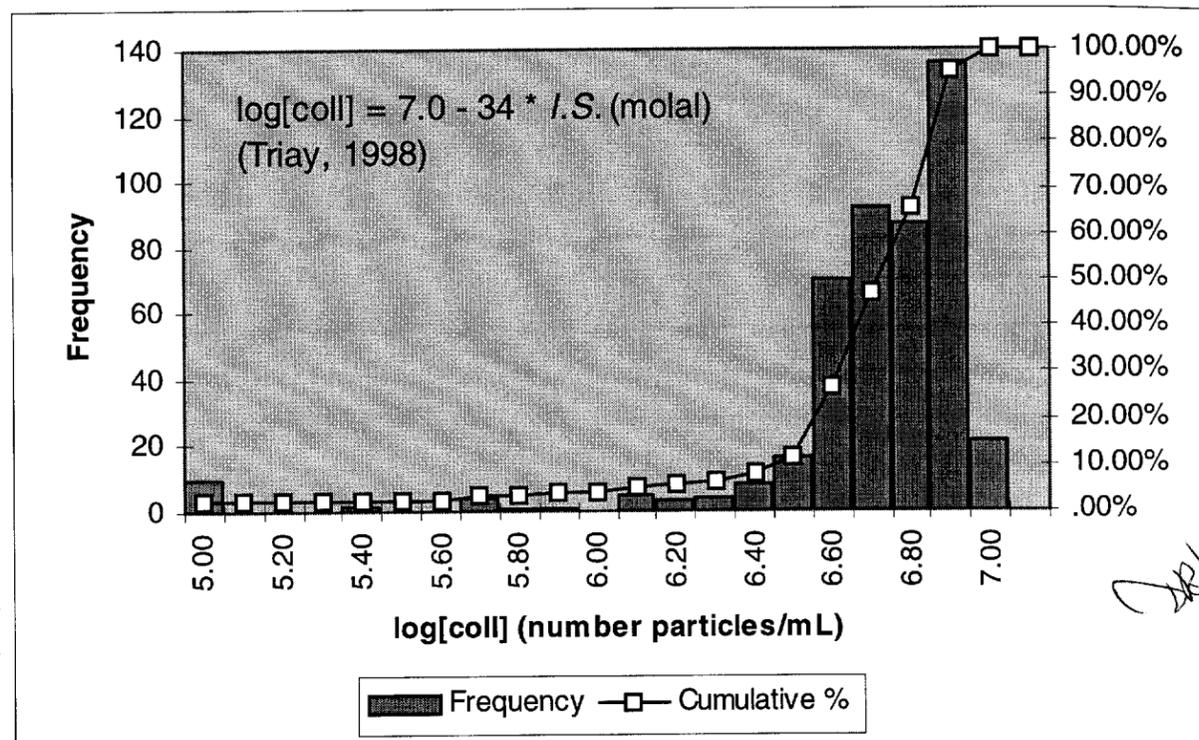
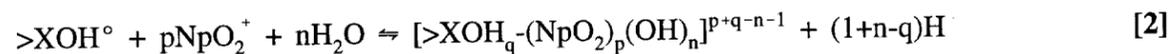


Figure 8. Colloid particle concentration (particles/mL) based on the empirical relationship between colloid particles and ionic strength (molal) presented by Triay (1998). Ionic strength is based on the equilibrium ionic strength (see Figure 4) calculated for water chemistries from Perfect et al. (1995). See section 3.1 for details.

Retardation Parameters

Surface reactions are written for sorbing species, and mass action and mass balance relations are used to determine sorption at the mineral surface as a function of system chemistry. Of the different SCMs, the Diffuse-Layer Model (DLM) is perhaps the simplest, using a one layer representation of the mineral-water interface. A generalized pH-dependent sorption reaction between aqueous actinides and a variably charged surface sorption site can be represented by Np(V) and written in the form:



where q is the protonation state of the sorption site (q=0, 1, or 2 for deprotonated, neutral, and protonated sites, respectively), and p and n are the reaction coefficients for NpO₂⁺ and H₂O, respectively. NpO₂⁺ and [>XOH_q⁻(NpO₂)_p(OH)_n]^{p+q-n-1} represent the aqueous Np(V) species and the Np(V) surface complex, respectively. In the SCM approach, a coulombic correction is incorporated into the mass action expressions for surface reactions to extract the intrinsic equilibrium constants (e.g., K₊^{int}, K₋^{int} and K_{[>XOH_q⁻(NpO₂)_p(OH)_n]^{int})}

that are independent of surface charge. For sorption reactions of the type given in Equation [2], K_{[>XOH_q⁻(NpO₂)_p(OH)_n]^{int} is commonly referred to as the binding constant. Similar reactions can be written for other actinides such as uranium and plutonium.}

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Sorption Modeling - DLM parameters for Np(V)- and U(VI)-montmorillonite sorption

The DLM has been used to simulate Np(V) and U(VI) sorption on montmorillonite (Pabalan and Turner, 1997; Turner et al., 1998). Montmorillonite is a common fracture mineral in volcanic tuffs in the YM vicinity (Bish et al., 1996; Vaniman et al., 1996), and is probably also present in the alluvium derived from volcanic outcrop. The observed dependence of Np(V)- and U(VI)-montmorillonite sorption on pH and P_{CO₂} is a consequence of mass action effects and equilibrium chemistry in the actinide-H₂O-CO₂-montmorillonite system represented by Equation [2]. In a qualitative sense, an increase in the activity of NpO₂⁺ (or similarly UO₂²⁺) drives the equilibrium reaction in Equation [2] forward (increasing sorption). The presence of a complexing ligand such as dissolved carbonate in the presence of a CO₂ atmosphere tends to form aqueous actinide-carbonate complexes in competition with the sorbing clay surface. Carbonate competition for the available actinide increases with increasing pH, reducing the aqueous activity of NpO₂⁺ or UO₂²⁺ and driving the reaction in the opposite direction (decreasing sorption). This explanation is, of course, simplistic due to the synergistic effects between solution chemistry, sorption site protonation state, and speciation of the aqueous and surface complexes.

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Sorption Modeling - DLM Parameters

Modeling for the regional groundwaters in the YM vicinity was performed for both Np(V) and U(VI) sorption on montmorillonite using the DLM. MINTEQA2, Version 3.11 (Allison et al., 1991) was used in the same manner as that described in section 3, with major element water chemistry from the culled data set of Perfect et al. (1995) formatted for MINTEQA2 input (460 analyses). Surface sorption reactions, based on CNWRA experimental work (Pabalan and Turner, 1997; Turner et al., 1998), were added to the input file. Total dissolved and sorbed concentrations were extracted from the output file for ease of handling.

Parameters and reference sources for the model are given. It is important to note that although full aqueous speciation is included in the model, the only surface reactions considered in the model are protonation/deprotonation and radionuclide sorption reactions. No competitive sorption reactions are incorporated in the model due to a general lack of available information on sorption behavior for the H₂O-CO₂-montmorillonite system. Therefore, predicted changes in sorption behavior are limited to those resulting from aqueous speciation of the radionuclide with ligands in the groundwater (Pabalan and Turner, 1997; Turner et al., 1998). DLM results are also dependent upon the thermodynamic data that are used in constructing the model. The thermodynamic data for Np(V) and U(VI) are taken from the MINTEQA2 database modified at the CNWRA (Turner, 1993), with exceptions as noted in Pabalan and Turner (1997) and Turner et al. (1998).

With regard to sorption, only surface complexation reactions are considered; the slightly acid to alkaline pH solutions in the regional groundwater system are above the pH range for likely ion exchange for actinides, even at low ionic strengths (Pabalan and Turner, 1997; Turner et al., 1998). A M/V ratio of 1 g/L is assumed due to a lack of information on effective M/V ratios in the YM system. Modeling over a wide range in M/V ratios (Turner, 1995) suggests that above a threshold value, there is an overabundance of sorption sites relative to radionuclide concentration and further increases in M/V have relatively little effect on the predicted sorption maximum. For this reason, the actual value chosen for M/V may be relatively insignificant as long as the selected value is above this threshold.

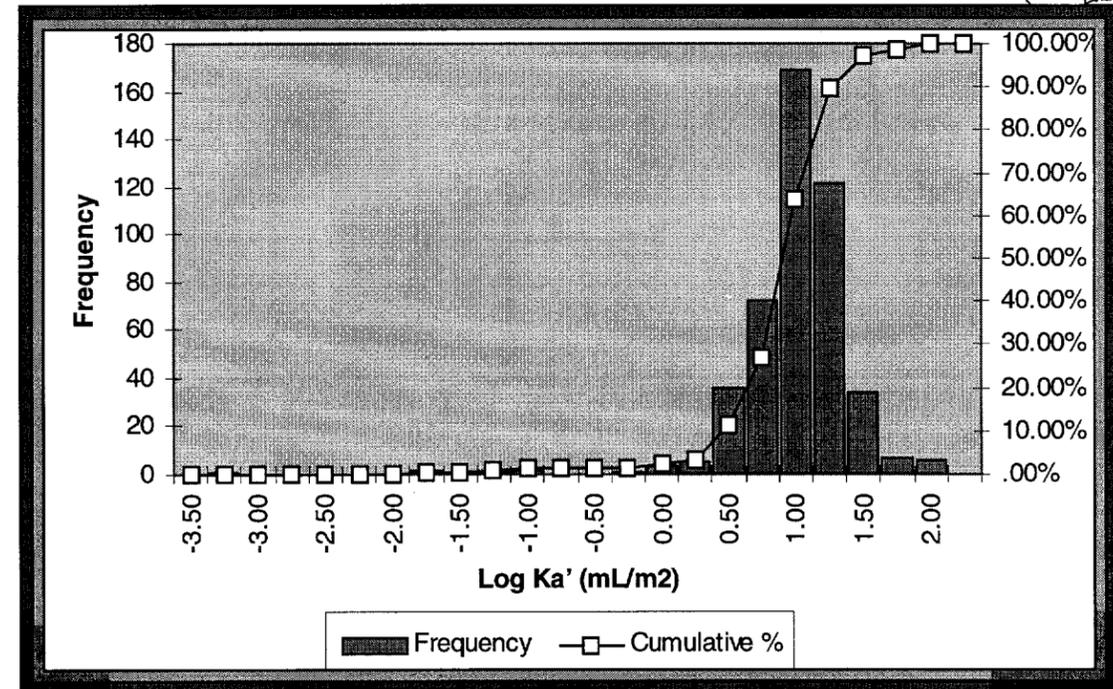
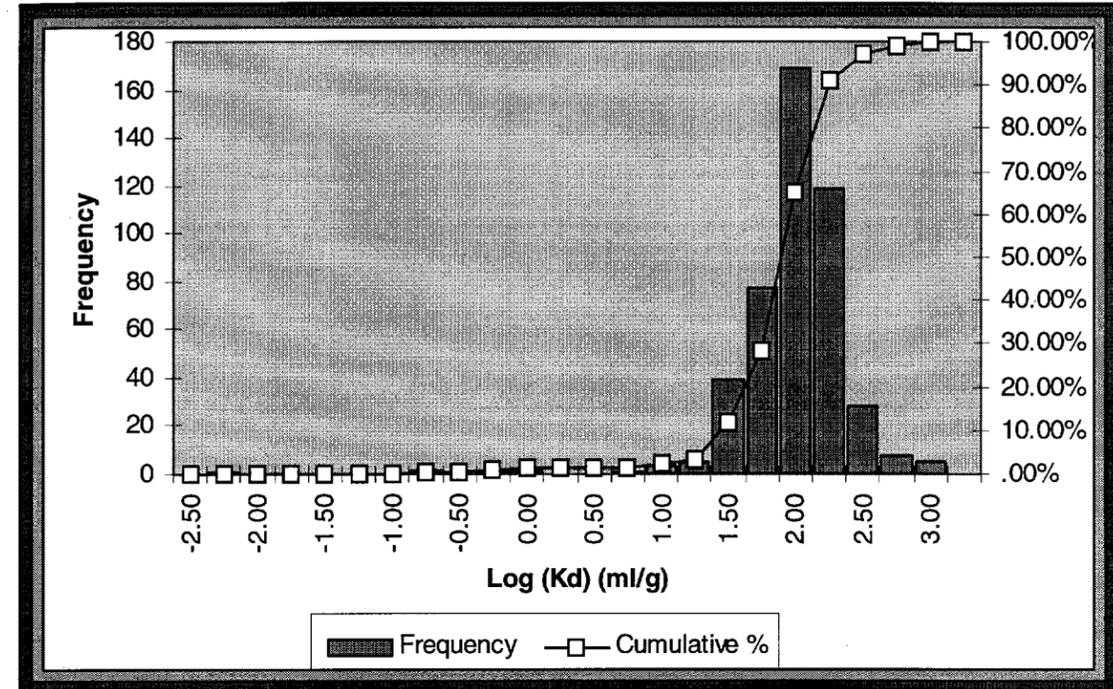
DLM parameters used in modeling Np(V) and U(VI) sorption on montmorillonite.

Model Parameter/Surface Reaction	Mineral/Surface Parameters	
Solid concentration (M/V)	1 g/L	
Site density	2.3 sites/nm ²	
Surface area _a	9.7 m ² /g	
Total site concentration	>AlOH ^o = 1.69 x 10 ⁻⁵ mol sites/L >SiOH ^o = 2.03 x 10 ⁻⁵ mol sites/L	
Ionic strength	Calculated by MINTEQA2	
P _{CO2}	Calculated by MINTEQA2	
Edge-Site Reactions:	Log K	
	Np(V)	U(VI)
Total radionuclide concentration	1x10 ⁻⁶ m	1x10 ⁻⁶ m
>AlOH ^o + H ⁺ ⇌ >AlOH ₂ ⁺	8.33 _b	8.33 _b
>AlOH ^o ⇌ >AlO ⁻ + H ⁺	-9.73 _b	-9.73 _b
>SiOH ^o ⇌ >SiO ⁻ + H ⁺	-7.20 _b	-7.20 _b
>AlOH ^o + NpO ₂ ⁺ + H ₂ O ⇌ >AlO-NpO ₂ (OH) + 2H ⁺	-13.79 _c	--
>SiOH ^o + NpO ₂ ⁺ ⇌ >SiOH-NpO ₂ ⁺	4.05 _c	--
>AlOH ^o + UO ₂ ²⁺ ⇌ >AlO-UO ₂ ⁺ + H ⁺	--	2.70 _d
>SiOH ^o + UO ₂ ²⁺ ⇌ >SiO-UO ₂ ⁺ + H ⁺	--	2.60 _d
>AlOH ^o + 3UO ₂ ²⁺ + 5H ₂ O ⇌ >AlO-(UO ₂) ₃ (OH) ₅ ^o + 6H ⁺	--	-14.95 _d
>SiOH ^o + 3UO ₂ ²⁺ + 5H ₂ O ⇌ >SiO-(UO ₂) ₃ (OH) ₅ ^o + 6H ⁺	--	-15.29 _d

- a. Effective edge site surface area assumed to be 10% of total N₂-BET surface area (97 m²·g⁻¹) (Pabalan et al., 1998).
- b. Acidity constants for am-SiO₂ and α-Al₂O₃ from Turner and Sassman (1996).
- c. Np(V)-montmorillonite binding constants from Turner et al. (1998).
- d. U(V)-montmorillonite binding constants from Pabalan and Turner (1997).

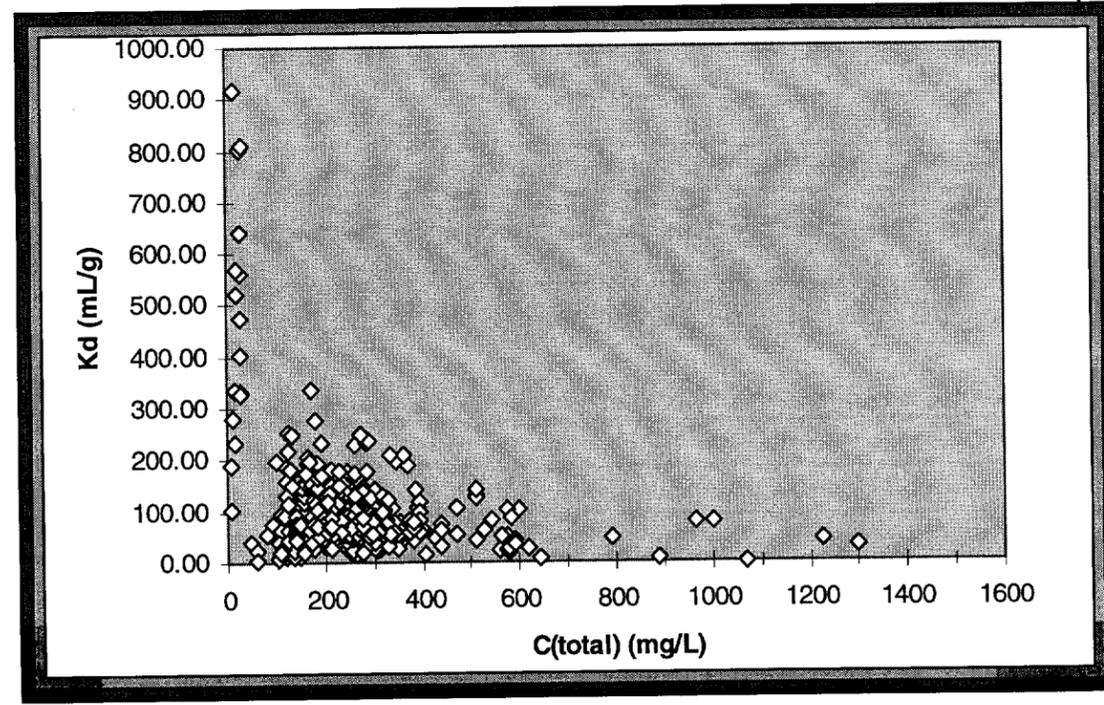
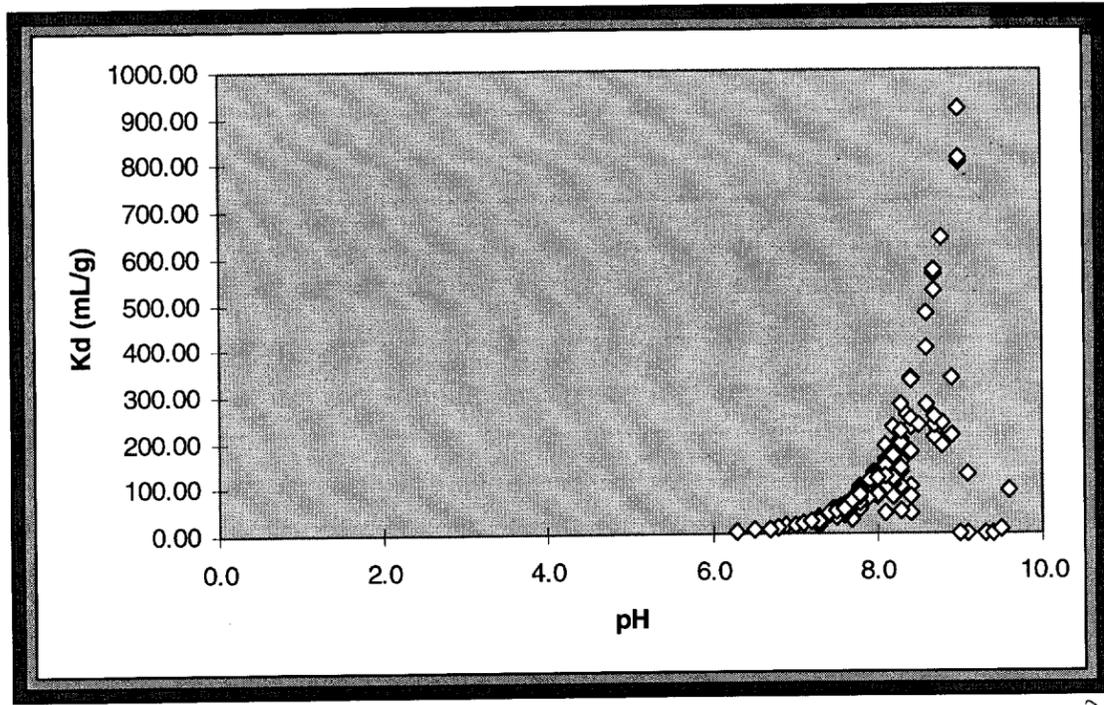
Np-Montmorillonite Sorption Results

Modeling Results - Np(V)-Montmorillonite



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Modeling Results - Np(V)-Montmorillonite



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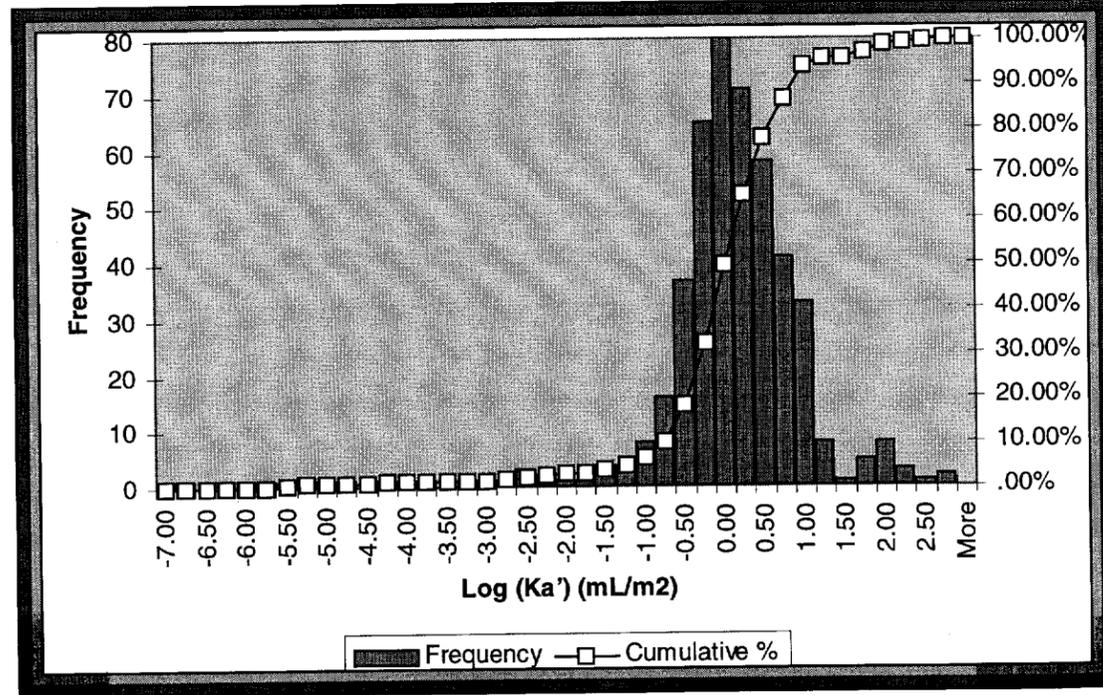
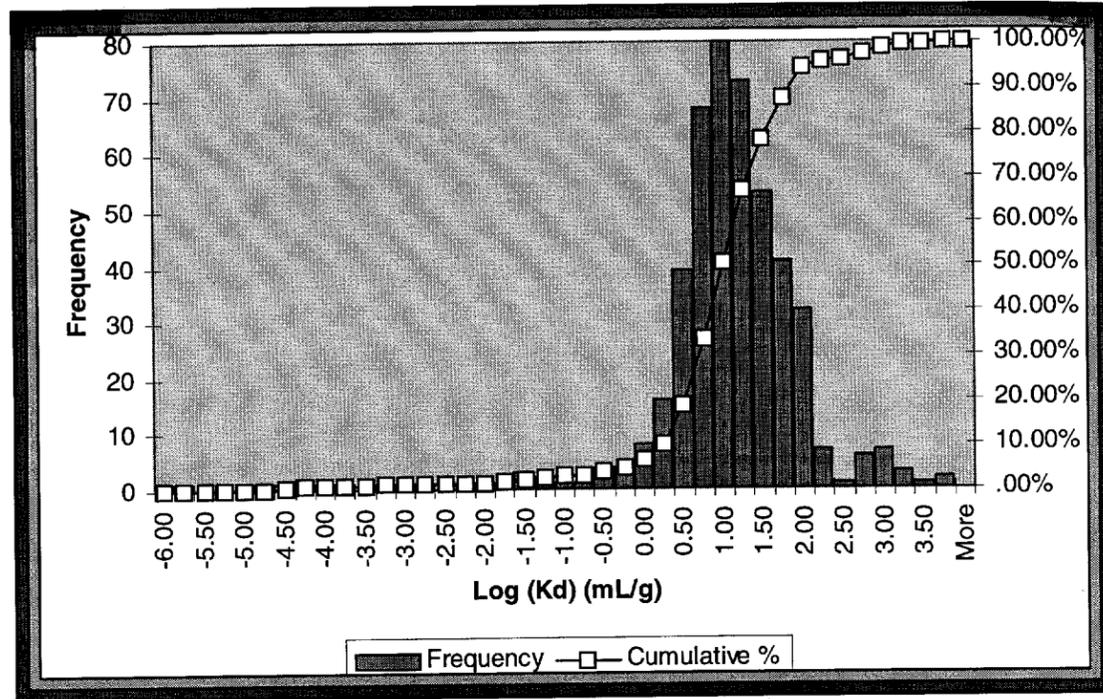
Descriptive statistics of predicted Np(V)-montmorillonite sorption for groundwater chemistries reported in Perfect et al. (1995). Derived using DLM with model parameters given above.

Np(V)-Montmorillonite	Log (K_D) (mL/g)	Log K_A (mL/m ²)
Mean	1.85	0.86
Standard Error	0.02	0.02
Median	1.89	0.90
Mode	2.07	1.09
Standard Deviation	0.44	0.44
Sample Variance	0.20	0.20
Kurtosis	24.53	24.53
Skewness	-3.45	-3.45
Range	5.24	5.24
Minimum	-2.27	-3.26
Maximum	2.96	1.97
Sum	851.15	397.23
Count	460	460
Confidence Level(95.0%)	0.04	0.04

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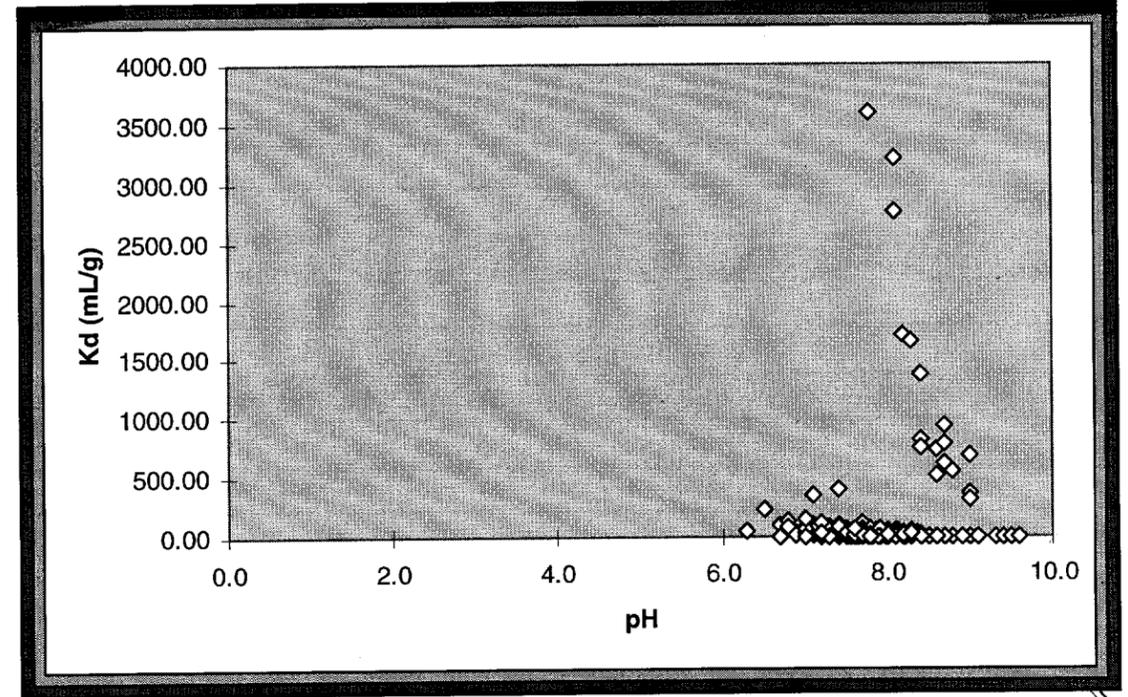
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Modeling Results - U(VI)-Montmorillonite



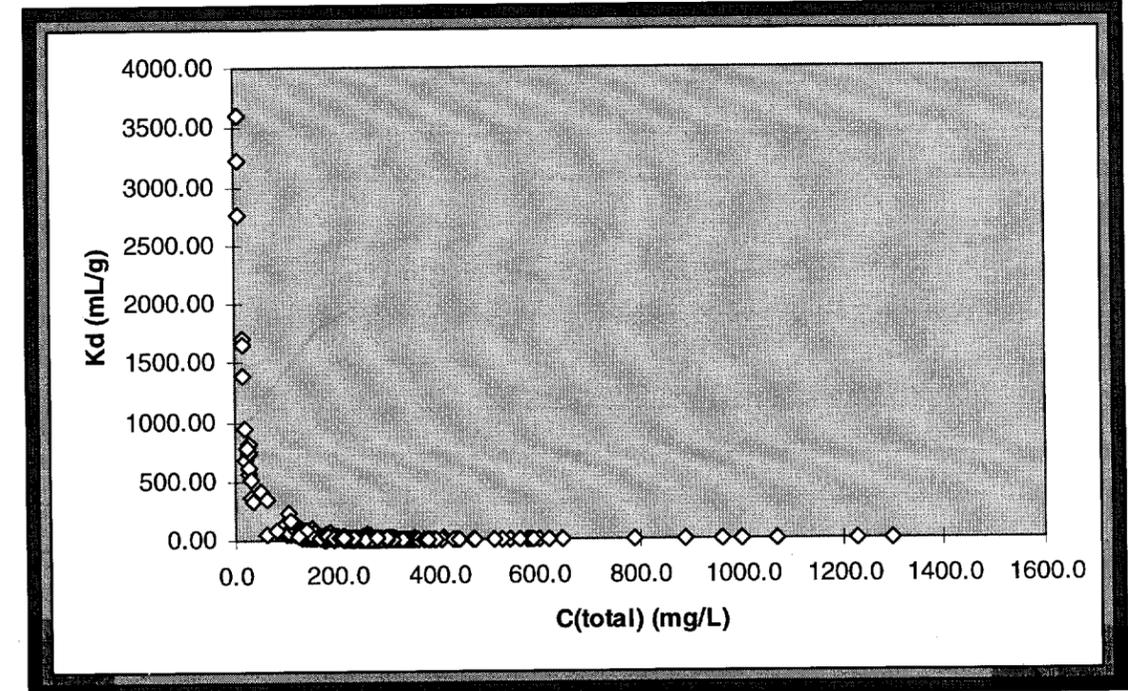
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Modeling Results - U(VI)-Montmorillonite



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Descriptive statistics of predicted U(VI)-montmorillonite sorption for groundwater chemistries reported in Perfect et al. (1995). Derived using DLM with model parameters given above.

U(VI)-Montmorillonite	Log (K_D) (mL/g)	Log K_A (mL/m ²)
Mean	0.96	-0.03
Standard Error	0.05	0.05
Median	0.99	0.002
Mode	0.83	-0.16
Standard Deviation	0.98	0.98
Sample Variance	0.95	0.95
Kurtosis	12.93	12.93
Skewness	-2.32	-2.32
Range	9.41	9.41
Minimum	-5.85	-6.84
Maximum	3.56	2.57
Sum	439.35	-14.56
Count	460	460
Confidence Level(95.0%)	0.089	0.089

Incorporation of Predictive Distributions in PA

The predicted Np(V) and U(VI) sorption behavior represents a means of constraining the development of PA PDFs. Descriptive statistics such as those given for Np(V)- and U(VI)-montmorillonite sorption represent the type of information that can be indirectly incorporated from predictive sorption models, including distribution type, mean values, upper and lower limits, and standard deviations. For Np(V)- and U(VI)-montmorillonite sorption, K_D can be calculated by multiplying K_A by the effective surface area of 9.7 m²/g, a constant (Bertetti et al., 1998; Pabalan et al., 1998). For this reason, although the magnitude of the values is different, the distribution type and descriptive statistics such as standard deviation, sample variance and skewness are the same for the two sorption parameters (e.g., compare columns two and three in the DLM results tables). As demonstrated in Bertetti et al. (1998) and Pabalan et al. (1998), Np(V) and U(VI) sorption on aluminosilicate minerals exhibit a similar pH-dependent behavior that is independent of mineral type. Absolute sorption is instead a function of surface area. This suggests that the distribution developed for sorption on montmorillonite is valid for other aluminosilicate and silicate minerals. The magnitude of K_D for PA transport calculations can therefore be determined for a given mineral by multiplying the distribution in Figure 9 by the effective surface area for that mineral. For example, for quartz, the effective surface area is typically small [$A' = 0.03$ m²/g (Bertetti et al., 1998)]. Assuming that the distribution shown in Figure 9 is valid for Np(V)-quartz sorption, the mean $K_{D,Qz} = K_A * A' = (7.2 \text{ mL/m}^2) * (0.03 \text{ m}^2/\text{g}) = 0.2 \text{ mL/g}$. This value compares well to the low values ($K_{D,Qz} < 1 \text{ mL/g}$) reported for Np(V) batch sorption studies with crushed tuff and J-13 well water (Triay et al., 1996a).

The application of a calibrated sorption model for other radionuclides such as U(VI), Np(V), Pu(V), and Pu(IV) also offers a means of correlating among sorption coefficient distributions that is missing in the current Iterative Performance Assessment code (Wescott et al., 1995). For example, calculating a distribution for U(VI)-montmorillonite sorption for the same set of water analyses used to generate Figure 9 can provide a direct means of testing the correlation between Np(V) and U(VI) sorption that takes into account at least some of the observed ranges in ambient chemistry. For Np(V) and U(VI) sorption on montmorillonite, the calculated correlation, defined by:

$$\rho = \frac{\text{Cov}(x,y)}{\sigma_x \sigma_y} \quad [3]$$

where:

$$\text{Cov}(x,y) = \frac{1}{n} \sum_{j=1}^n (x_j - \mu_x)(y_j - \mu_y) \quad [4]$$

is about 0.5, indicating a positive correlation between calculated sorption coefficients. It is important to remember that these models were calibrated in end-member experimental systems (Pabalan and Turner, 1997; Turner et al., 1998), and do not take into account possible competition among radionuclides for available sorption sites.

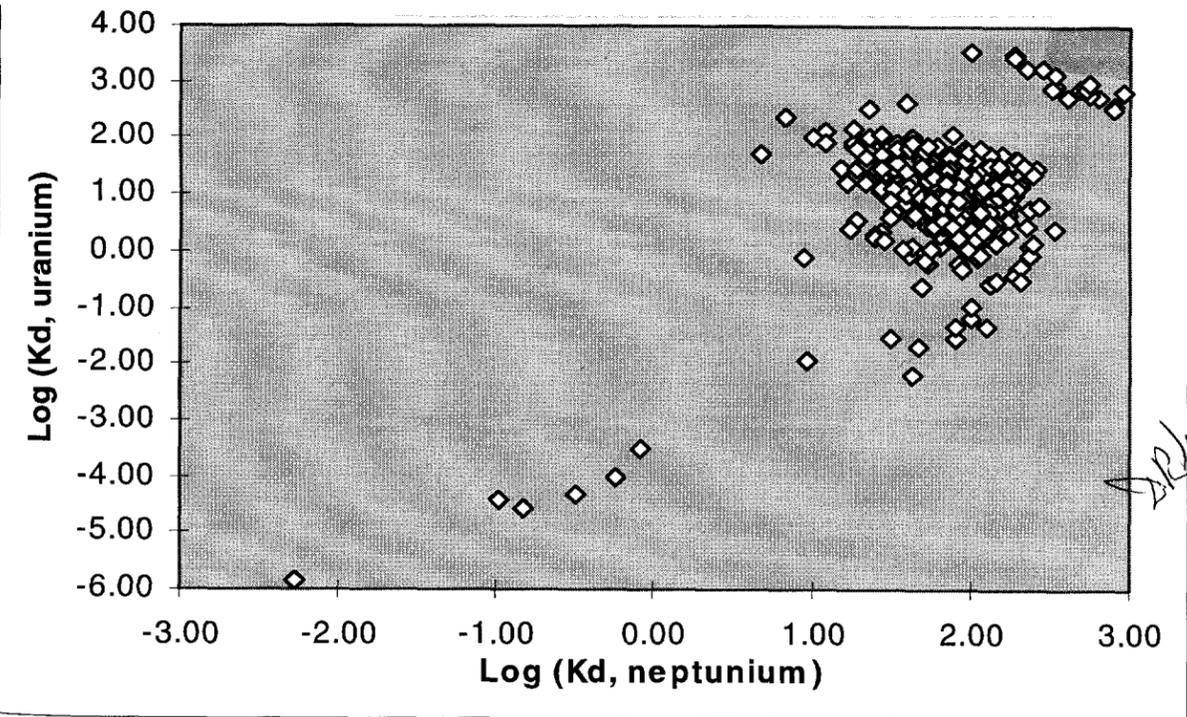


Figure 15. Corresponding Np(V)- and U(VI)-montmorillonite sorption coefficients (K_D in mL/g) calculated with MINTEQA2 (Allison et al., 1991) using a Diffuse-Layer surface complexation model with parameters given in Table 4. Water chemistries are for saturated zone regional groundwaters (from Perfect et al., 1995; see text for details).

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Developing a Response Surface for Np(V)-montmorillonite sorption (5/98)

A summary of the DLM parameters used in generating the Np(V)-montmorillonite response surface is given in the Table.

The model parameters (i.e., binding constant and surface reactions modeled) are based on a limited number of Np(V) sorption experiments conducted over a limited range of PCO_2 , M/V, and Np(V) concentration. The model is also limited to surface complexation reactions only and does not consider any ion exchange reactions. Ion exchange is not considered because the experiments on which the model is based were conducted at relatively high ionic strength in an effort to suppress ion exchange in the montmorillonite at low pH. Thus, the extent of chemical and physical conditions over which the model can be applied should be carefully considered.

Conditions and reaction constants used for the Diffuse-Layer Model in this study.

Site density	2.3 sites/nm ²
Surface area _a	9.7 m ² /g
Edge-Site Reactions:	Log K
$>AlOH^o + H^+ \rightleftharpoons >AlOH_2^+$	8.33 _b
$>AlOH^o \rightleftharpoons >AlO^- + H^+$	-9.73 _b
$>AlOH^o + NpO_2^+ + H_2O \rightleftharpoons >AlO-NpO_2(OH)^- + 2H^+$	-13.79 _c
$>SiOH^o \rightleftharpoons >SiO^- + H^+$	-7.20 _b
$>SiOH^o + NpO_2^+ \rightleftharpoons >SiOH-NpO_2^+$	4.05 _c
Aqueous Speciation Reactions:	Log K
$NpO_2^+ + H_2O \rightleftharpoons NpO_2OH^o + H^+$	-10.0 _d
$NpO_2^+ + 2H_2O \rightleftharpoons NpO_2(OH)_2^- + 2H^+$	-22.4 _{d,e}
$NpO_2^+ + CO_3^{2-} \rightleftharpoons NpO_2CO_3^-$	4.6 _{e,f,g}
$NpO_2^+ + 2CO_3^{2-} \rightleftharpoons NpO_2(CO_3)_2^{3-}$	7.0 _{e,f,g}
$NpO_2^+ + 3CO_3^{2-} \rightleftharpoons NpO_2(CO_3)_3^{5-}$	8.5 _{f,g}
$NpO_2^+ + NO_3^- \rightleftharpoons NpO_2NO_3^o(aq)$	-0.5 _h

a. Effective edge site surface area assumed to be 10% of total N₂-BET surface area (97 m²/g). See text for detailed discussion.

b. Acidity constants for am-SiO₂ and α-Al₂O₃ from Turner and Sassman (1996).

c. Np(V)-montmorillonite binding constants from Turner et al. (1998).

d. Lemire and Garisto (1989)

e. Fuger (1992)

f. Lemire (1984)

g. Lemire et al. (1993)

h. Danesi et al. (1971)

Simulations were conducted over a range of pH, PCO_2 , M/V, and Np(V) concentration. A summary of all MINTEQA2 model runs is provided. In general, for each model simulation PCO_2 was held constant while pH was varied throughout a range from 2 to 11.75. MINTEQA2 reports percent Np(V) remaining in solution and percent Np(V) sorbed on surface sites. These values are then converted to K_D and subsequently, K_A s by applying the known conditions (Np(V) concentration, M/V, and surface area of the mineral) of the model

run. Following the simulations, generated data was analyzed to identify important variables and evaluate significant trends and consistencies. Surface plots of results were generated. An effort was then made to evaluate the best approach to reproducing sorption coefficient or response surface.

Summary of MINTEQA2 simulations.

Run	pH range	PCO_2 (atm)	Np(V) _{total}	M/V (g/L)
1	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ through 10 ⁻² atm in 10 ^{0.5} atm increments	8.9 x 10 ⁻⁷ M	4
2	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ through 10 ⁻² atm in 10 ^{0.5} atm increments	2.0 x 10 ⁻¹¹ M	4
3	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ atm, 10 ^{-3.5} atm	8.9 x 10 ⁻⁷ M	40
4	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ atm, 10 ^{-3.5} atm	8.9 x 10 ⁻⁷ M	400
5	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ atm, 10 ^{-3.5} atm	8.9 x 10 ⁻⁷ M	2000
6	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ atm, 10 ^{-3.5} atm	1.0 x 10 ⁻¹⁴ M	4
7	2 to 11.75 in 0.25 increments	no CO ₂ , 10 ⁻⁷ atm, 10 ^{-3.5} atm	1.0 x 10 ⁻¹⁸ M	4

A comparison of the SCM generated data with experimental data collected under similar Np(V) concentration, M/V and over a range of pH and PCO_2 confirms that the model reliably reproduces the data on which it is based. Fig. 1, pg. 30

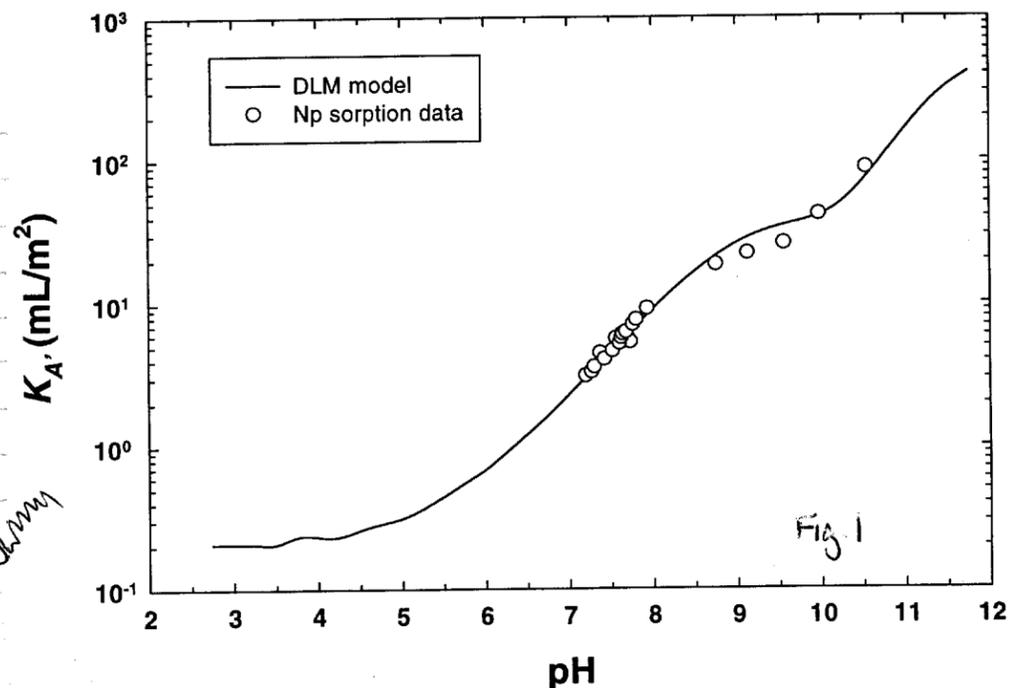
Sensitivity of the Model Results to pH

The DLM used in this study reproduces the pH dependency well. In general, Np(V) sorption follows a trend similar to the hydrolysis of the NpO₂⁺ species in solution (Bertetti et al., 1996). For any set of additional variables (e.g., PCO_2 , M/V, Np(V) concentration) there is a distinct increase in sorption with increasing pH starting at pH-6. For simulations where CO₂ is present, Np(V) sorption continues to increase with increasing pH eventually peaking between pH 7.5 and pH 10. Under CO₂-free conditions, Np(V) sorption continues to increase throughout the simulated pH range up to pH 11.75. For PCO_2 greater than zero, Np(V) sorption decreases with increasing pH after reaching its maximum until it finally reaches a calculated value of zero at high pH values. The actual value is dependent on the PCO_2 used in the model run. Fig. 2, pg. 30

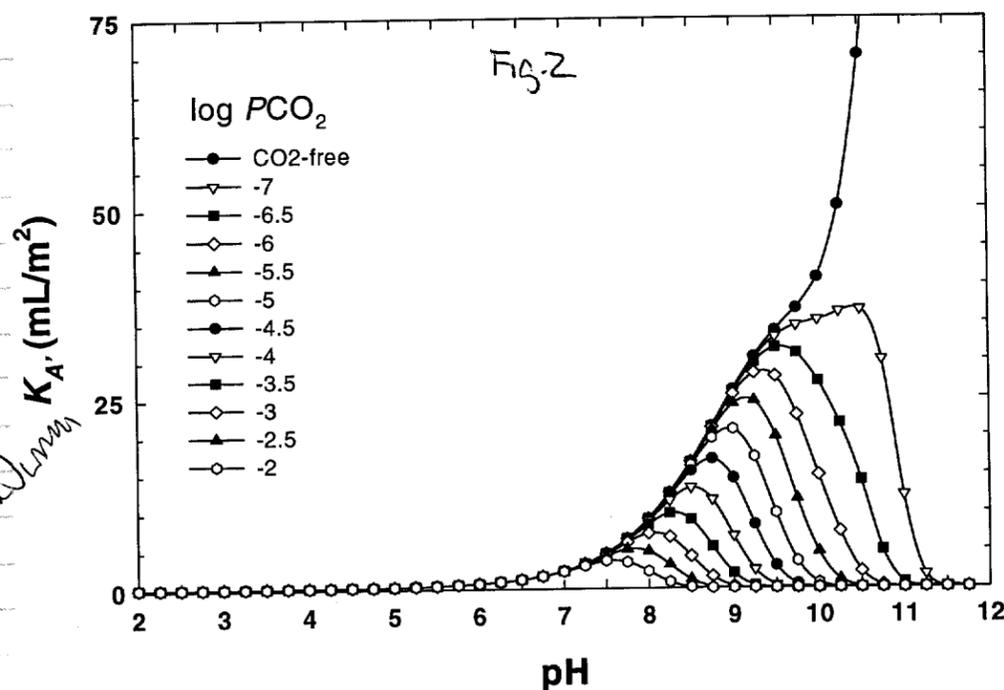
Sensitivity of the model to variation in PCO_2

The pH range of the sorption envelope (region where sorption of Np(V) increases, rises to a maximum and then decreases) decreases as the PCO_2 increases. The peak (maximum magnitude) of Np(V) sorption occurs at progressively lower pH values as the PCO_2 increases. The sorption peak shifts from pH~10.5 or $PCO_2 = 10^{-7}$ atm to pH~7.5 for $PCO_2 = 10^{-2}$ atm. In addition, the magnitude of the sorption maximum decreases from about 40 mL/m² for PCO_2 of 10⁻⁷ atm to less than 5 mL/m² for PCO_2 of 10⁻² atm. In fact, for any given pH

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Comparison of DLM output versus experimental data for Np(V) sorption onto montmorillonite under CO₂-free conditions. Np(V)_{total} ~ 1 × 10⁻⁶ M, M/V = 4 g/L.



DLM results of Np(V) sorption plotted over a range of PCO₂ and pH. Np(V)_{total} ~ 1 × 10⁻⁶ M, M/V = 4 g/L.

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within the sorption envelope, the Np(V) sorption decreases exponentially with an increase in PCO₂. This behavior is not surprising because the PCO₂ is increasing exponentially and Np(V) sorption is linked to Np(V) speciation, including the formation of Np-carbonate species in solution. The results are consistent with previous experimental results (Bertetti et al., 1998; Turner et al., 1998).

Sensitivity of the model to variation in M/V

The model does not appear to be sensitive to changes in M/V when data is represented in terms of K_A. That is to be expected since calculation of K_D, and subsequently K_A, normalizes data for differences in M/V. This has significant implications for application of this approach to field conditions where M/V is difficult, if not impossible, to determine. Fig. 4(a) + 4(b), pg. 33

Sensitivity of the model to variation in Np(V) concentration

Neptunium concentration was varied over a range of 12 orders of magnitude from 10⁻⁶ M to 10⁻¹⁸ M. Nevertheless, the predicted Np(V) sorption was similar over the range of pH and PCO₂ modeled. These model results agree with experimentally derived results of Np(V) sorption at varying Np(V) concentrations (e.g., Righetto et al., 1991; Bertetti et al., 1998). A lack of model sensitivity to changes in Np(V) concentration is significant because it implies that the sorption model does not have to be modified to account for loss of Np(V) due to sorption. Therefore, no re-evaluation of K_Ds would be required in the TPA code due to changes in Np(V) concentration. Only changes in pH and PCO₂ would need to be considered.

Fig. 5, pg. 34

Comparison of the Sorption Model to Experimental Data

Comparisons to data generated in experiments conducted at CNWRA using a variety of minerals (Bertetti et al, 1998; Turner et al, 1998) reveal that the model performs favorably over a limited range of pH, M/V, Np(V) concentration, and PCO₂. Fig. 6 pg. 34

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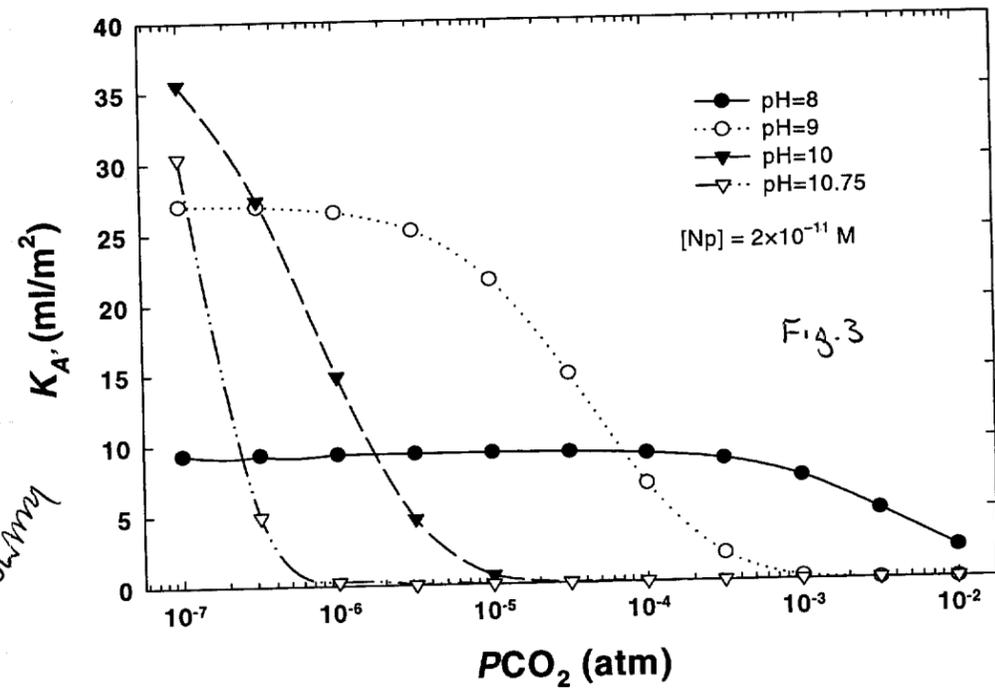
Assuming that the model presented here is correct in its representation of independence with respect to changes in Np(V) concentration and M/V, and since pH is generally reported, only the PCO₂ and mineral surface area need be known to produce a comparison between experiments. However, even mineral surface area may vary significantly depending on the methods used to determine Surface area. Data from several studies (Beall and Allard, 1981; Torstenfelt et al., 1988; Righetto et al., 1991; Legoux et al., 1993) are plotted with the model curves for CO₂-free conditions and PCO₂ of 10^{-3.5} atm. A summary of the experimental conditions is given here. Fig. 7, pg. 36

Summary of experimental conditions for data used as comparison to DLM.

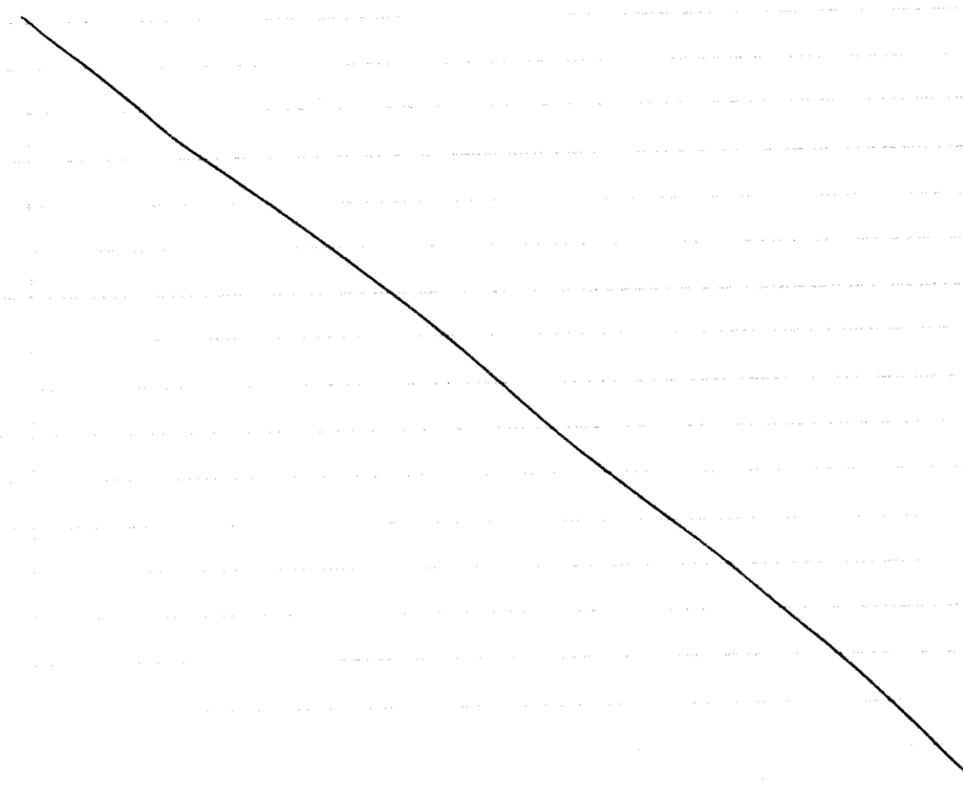
Investigators	Conditions
Beall and Allard (1981)	Np(V) _{total} ~ 2 × 10 ⁻¹¹ M, quartz
Allard et al. (1984)	Np(V) _{total} ~ 2 × 10 ⁻¹¹ M, montmorillonite
Torstenfelt et al. (1988)	Np(V) _{total} < 10 ⁻⁷ M, illite and bentonite, pH ~ 8 and ~ 10, 143 ppm HCO ₃ ⁻
Righetto et al. (1991)	Np(V) _{total} < 10 ⁻¹⁴ M, colloidal silica, pH 2-10, no specified CO ₂ conditions (appear CO ₂ -free)
Legoux et al. (1992)	Np(V) _{total} ~ 5 × 10 ⁻¹² M, mix of quartz, smectite, glauconite, kaolinite. Measured N ₂ -BET surface area, 2.1 - 52.4 ppm CO ₃ ²⁻

Data from Legoux et al. (1992) contain BET surface area measurements and correlate well with the model

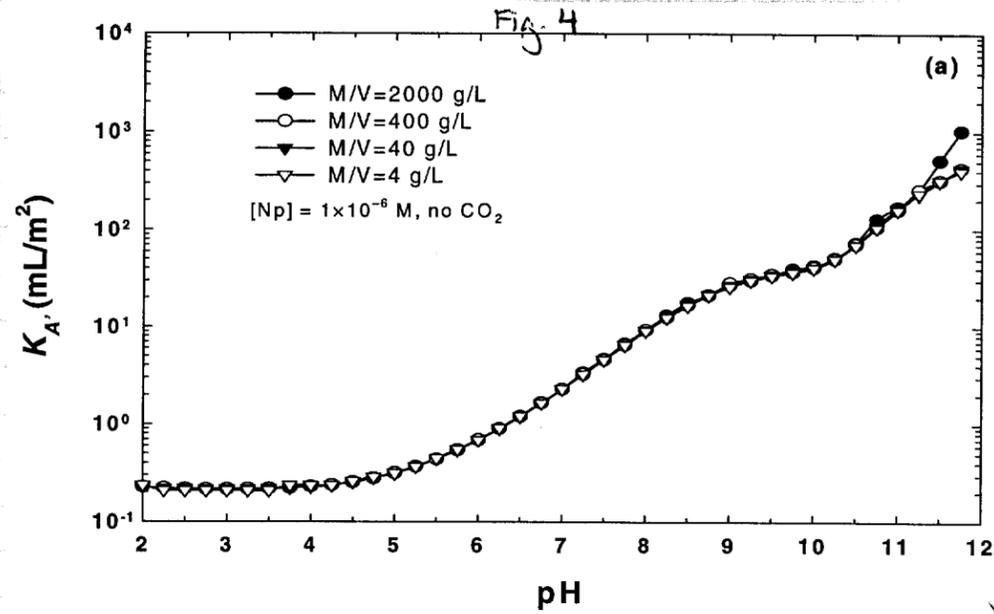
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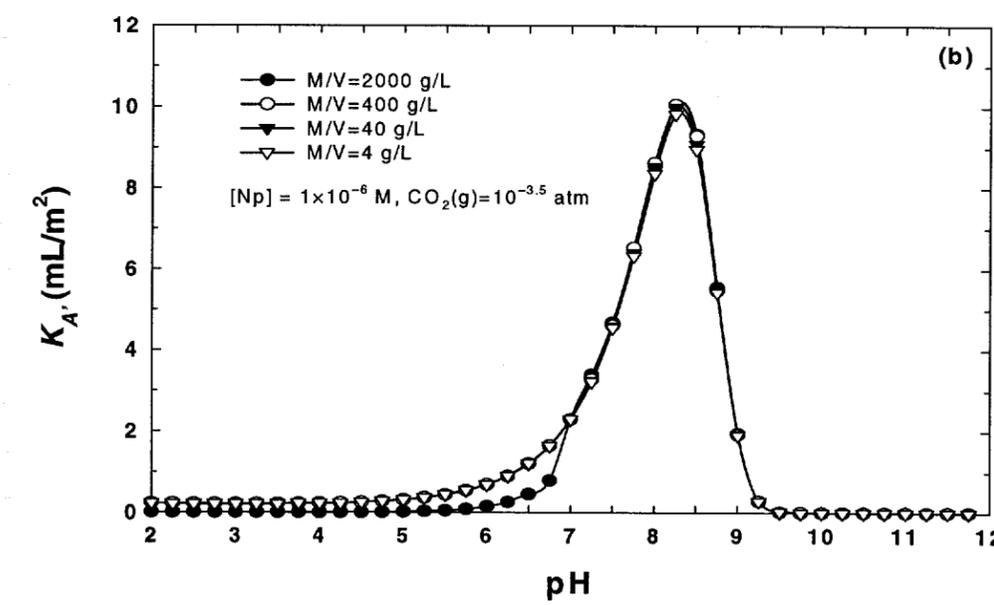
DLM results showing a decrease in Np(V) sorption as a function of PCO_2 for given pH values. $Np(V)_{total} \sim 2 \times 10^{-11} \text{ M}$, $M/V = 4 \text{ g/L}$.



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DLM results for Np(V) sorption at various M/V values under (a) CO_2 -free conditions, and (b) atmospheric CO_2 ($PCO_2 = 10^{-3.5} \text{ atm}$). $Np(V)_{total} \sim 1 \times 10^{-6} \text{ M}$.

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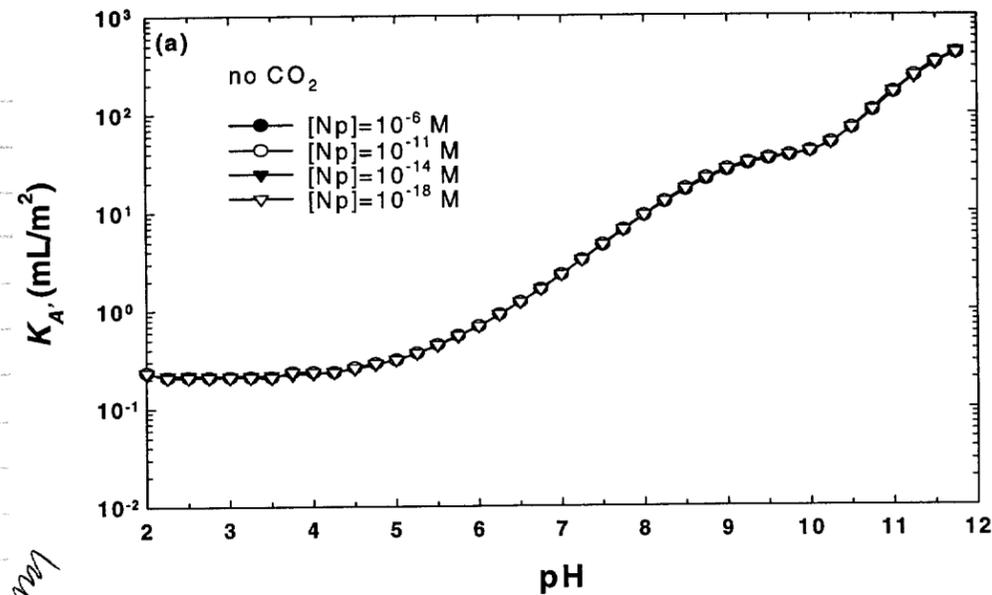
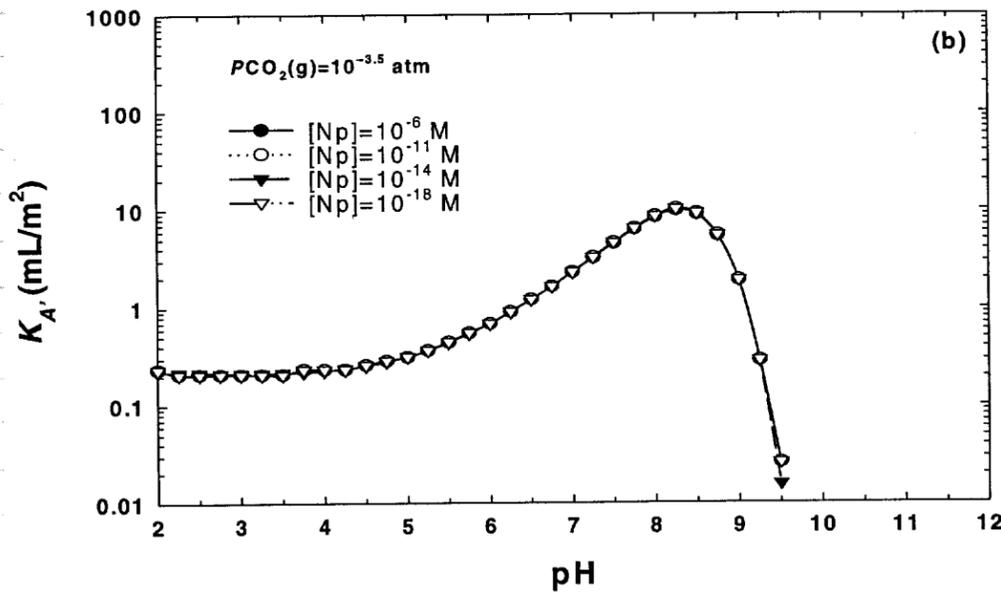


Fig. 5



DLM results for Np(V) sorption at various Np(V) concentrations under (a) CO₂-free conditions, and (b) atmospheric CO₂ (PCO₂ = 10^{-3.5} atm). M/V = 4 g/L.

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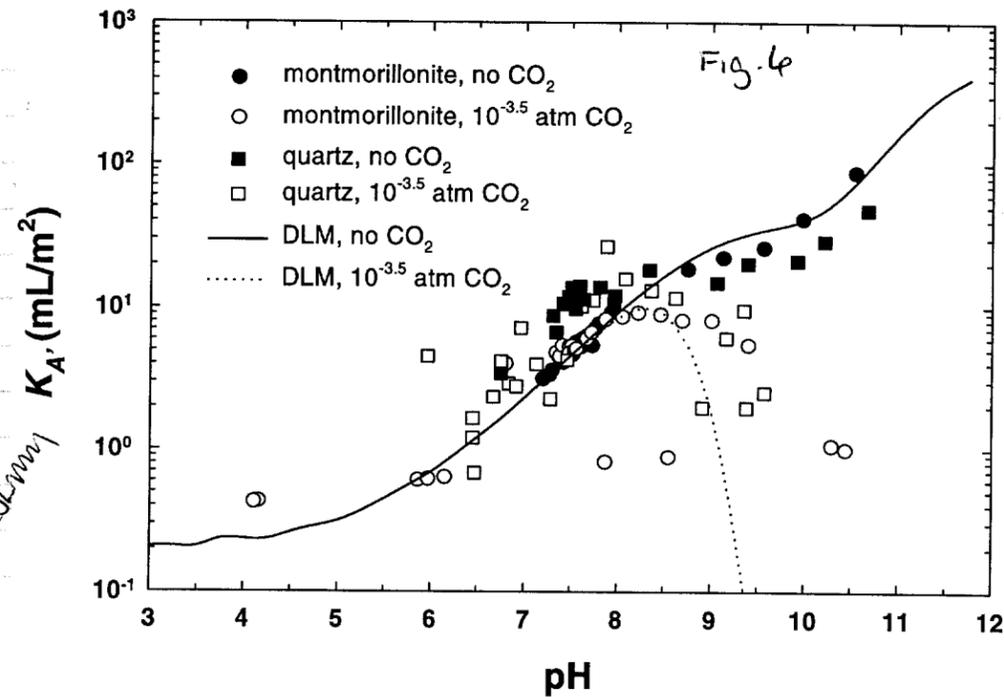
K_A plot. The minerals used by Legoux et al. are mixtures of quartz and clay minerals. Data from Righetto et al. (1991) were collected at $Np(V)_{total} \sim 10^{-14}$ M on colloidal silica. No surface area measurements are available for the colloid, but Righetto et al. (1991) report 1 μ m-sized aggregates of particles. Assuming the average spherical particle size was 50 nm, a reasonable calculated surface area for the colloids would be 20 m^2/g . This would result in a favorable comparison of their data to the model data. Data from Beall and Allard (1981) include experiments using both montmorillonite and quartz at $Np(V)_{total} \sim 2 \times 10^{-11}$ M. Data from the quartz experiments have high K_D , and it is difficult to assume a surface area that might result in K_A s equivalent to the model. However, there is little information on the mineral preparation for quartz used in their studies. If the quartz surfaces were contaminated with Fe-oxides, even in small quantities, the resulting K_D s would be larger than those expected for a pure quartz sorbent. Beall and Allard (1981) also report bicarbonate concentrations of 90-275 mg/L. However, no effect of reduction in sorption due to presence of carbonate is revealed in their Np(V) sorption results. Data from Allard et al. (1984) for montmorillonite also show high K_D s and little or no influence of the presence of carbonate. If a surface area of 100 m^2/g is assumed (A' of 10 m^2/g) for their montmorillonite, their data correlate better with the model used here, although K_A s are still greater than the model prediction at high pH and significant sorption is present, possibly due to ion exchange, at lower pH. To use data from Torstenfelt et al. (1988), a surface area for the clay must also be assumed (surface area = 100 m^2/g or $A' = 10 m^2/g$). When converted to K_A and including the presence of CO₂ reported in their experiments, their data correlate well with the model. Fig. 8, pg. 37

The model performs adequately when compared to experimental data collected by different researchers under a variety of chemical conditions and with a mixture of montmorillonite, quartz and other clay minerals. When the model does falter, it generally predicts K_A values that are lower than values measured experimentally, which is considered to be conservative with respect to PA model results. Broad applicability of the model is limited by:

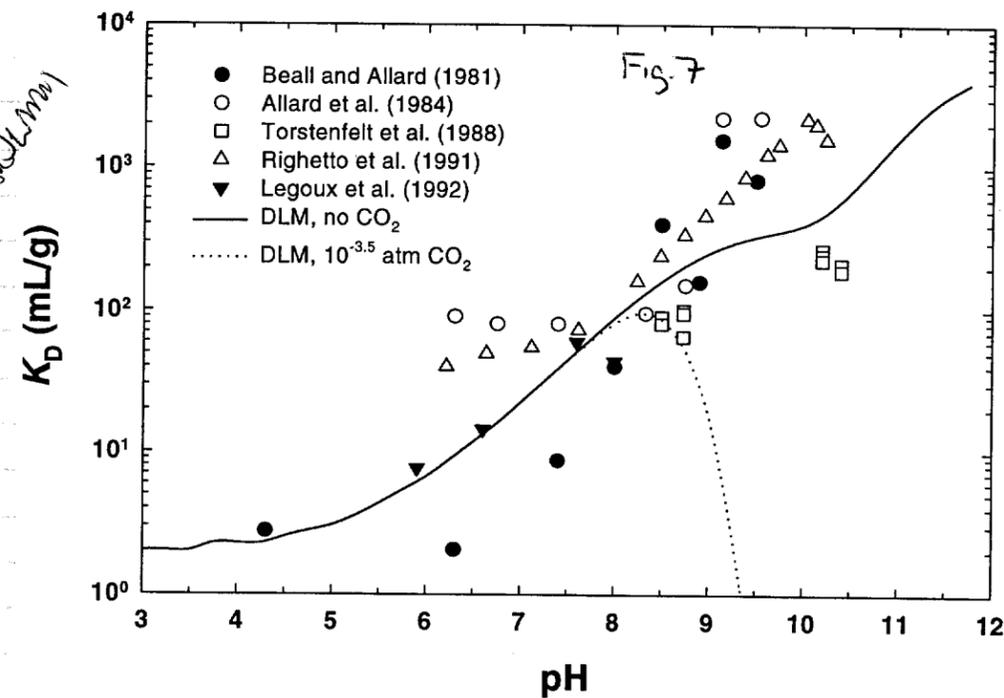
- a consistent underestimation of Np(V) sorption at high pH when PCO_2 is greater than zero
- a failure to account for ion exchange at low pH
- difficulties in comparing data among experiments in order to validate the model over a wide range of conditions due to incomplete information
- an underestimation of sorption for some experimental data, although the problems listed above may contribute greatly to this problem
- a general lack of Np(V) sorption experimental data at high M/V.

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DLM results for Np(V) sorption versus CNWRA experimental Np(V) sorption data (Bertetti et al., 1998) for montmorillonite and quartz.



DLM results for Np(V) sorption plotted versus data from other experimental studies. Minerals include: Quartz (Beall and Allard, 1981); montmorillonite (Allard et al., 1984); illite and bentonite (Torstenfelt et al., 1988); colloidal silica (Righetto et al., 1991); quartz-clay mixture (Legoux et al., 1992).

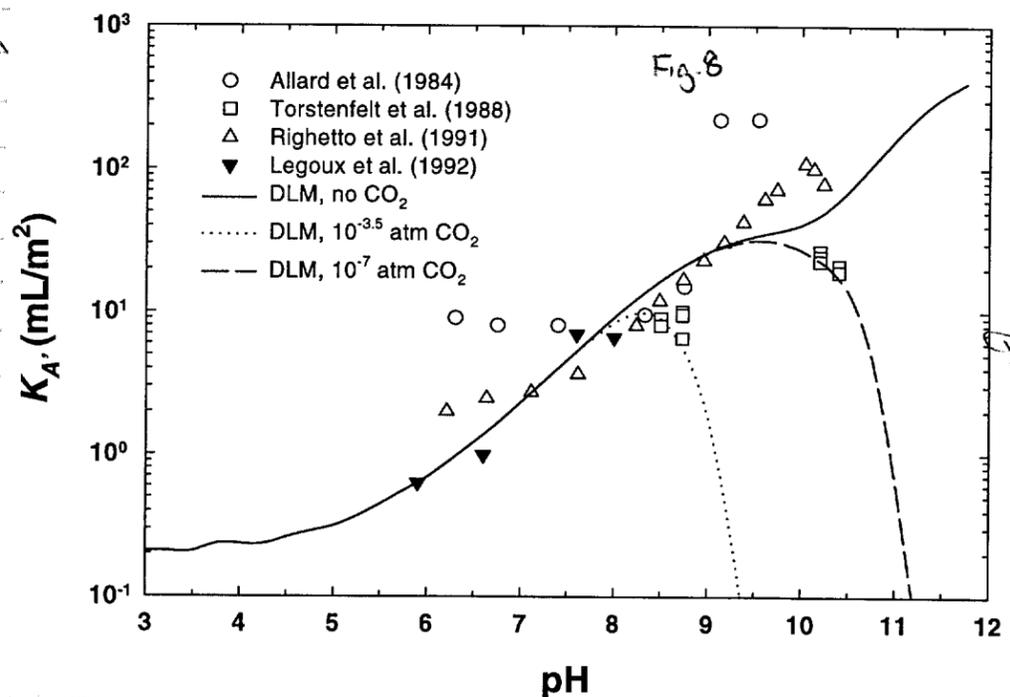
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Look-up Table or Interpolated Surface

The modeled Np(V) sorption surface is a non-interpolated mesh consisting of only of model generated data (mesh intersections). Because of the large range of data generated, especially for PCO_2 , interpolations of the surface, regardless of the methods used (e.g., linear interpolation, kriging, etc.), resulted in inadequate representations of the response surface. Interpolations failed to accurately represent the model surface, even with limited data sets. The only disadvantage of this method of data generation is that it is more time consuming. The data used to create the response surface can be organized into a look-up table format in a form such as the one shown. The primary disadvantages of the look-up table are that it requires some form of interpolation for both pH and PCO_2 since the data is based on discrete indices and that it requires regeneration and replacement of data if a more suitable model for K_A is found.

Three-Dimensional Mathematical Representation

A second approach to incorporating the K_A model would be to generate a mathematical representation of the DLM response surface. The mathematical model would have the advantage of utilizing a continuous range of data without the need for interpolation. A 3-D curve fitting application (TableCurve 3-D, Jandel Scientific) was used in an attempt to mathematically model the response surface. The curve fit routine was unable to generate a function or set of functions that adequately reproduced the response surface. The range of values to be modeled is too large for accurate reproduction over the entire data range considered. In general, the sorption maximum was underpredicted by the mathematical expressions. To improve the fit, the surface was first segregated into components to model only the range in PCO_2 from 10^{-7} to 10^{-2} atm, and further reduced to only the sorption range of pH 6 to 10.5. However, these attempts were also unsuccessful in improving the results.



DLM results for Np(V) sorption plotted versus K_A data from other studies. Data from Allard et al. (1984) and Righetto et al. (1991) are based on an estimated A' for montmorillonite of $10 \text{ m}^2/\text{g}$ (10 percent of a total N_2 -BET surface area of about $100 \text{ m}^2/\text{g}$).

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pH	Log PCO ₂ (atm)											
	no CO ₂	-7.00	-6.50	-6.00	-5.50	-5.00	-4.50	-4.00	-3.50	-3.00	-2.50	-2.00
2.00	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407
2.25	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
2.50	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
2.75	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
3.00	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
3.25	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
3.50	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785	0.20785
3.75	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407
4.00	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407
4.25	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407	0.23407
4.50	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034	0.26034
4.75	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666	0.28666
5.00	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303	0.31303
5.25	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595	0.36595
5.50	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572	0.44572
5.75	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285	0.55285
6.00	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799	0.68799
6.25	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713	0.90713
6.50	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444	1.21444
6.75	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510	1.64510
7.00	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218	2.30218
7.25	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067	3.25067
7.50	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393	4.58393
7.75	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362	6.48362
8.00	9.10258	9.10258	9.10258	9.10258	9.10258	9.05545	9.00844	8.86820	8.36349	7.18486	4.83630	2.21073
8.25	12.46597	12.46597	12.46597	12.46597	12.40932	12.35283	12.12856	11.47131	9.82514	6.68675	3.05588	0.79711
8.50	16.54732	16.54732	16.54732	16.54732	16.40879	16.13444	15.33238	13.15912	8.96157	4.16083	1.10188	0.10351
8.75	21.25818	21.25818	21.17252	21.08716	20.74882	19.68218	16.89766	11.57926	5.42922	1.50003	0.15557	0.00000
9.00	26.08434	25.98021	25.87650	25.46576	24.17486	20.83294	14.37197	6.80986	1.91015	0.20785	0.00000	0.00000
9.25	30.37756	30.13395	29.65303	28.25866	24.56508	17.18213	8.27330	2.36348	0.28666	0.00000	0.00000	0.00000
9.50	33.88698	33.06972	31.62813	27.80933	19.84308	9.77604	2.86369	0.33946	0.02580	0.00000	0.00000	0.00000
9.75	36.78310	34.58558	30.87119	22.67266	11.63347	3.48128	0.44572	0.02580	0.00000	0.00000	0.00000	0.00000
10.00	40.82421	35.30073	27.14918	14.75070	4.65561	0.60674	0.02580	0.00000	0.00000	0.00000	0.00000	0.00000

Sample look-up table for Np(V) sorption response surface (K_A in mL/m², Np(V)_{total} = 10⁻⁶ molal, M/V = 4 g/L)

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Combination of Look-up Table and Mathematical Representation

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Fig. 9, pg. 40

A data set could be constructed that would require interpolation of only one component (in this example PCO₂). This technique would offer the advantage of producing results over a continuous range of pH values while using information for discrete PCO₂ values. The similarities in the shape of the pH curves for PCO₂ suggested that the same type of equation might be capable of representing each curve. pH-sorption curves were fit using TableCurve 2D (Jandel Scientific) to generate equations over a pH range appropriate for each PCO₂ (i.e. for PCO₂ of 10⁻⁶ atm the model was fit from pH 6 to pH 10; likewise, for PCO₂ of 10⁻² atm, the model was fit from pH 6 to pH 8.5 - essentially the pH range of the sorption envelope at a given PCO₂, K_As below pH 6 and above some pH for a given PCO₂ are effectively zero). A relatively simple equation:

$$\ln(y) = a + bx + cx^2 + dx^3 + ex^4 + fx^5$$

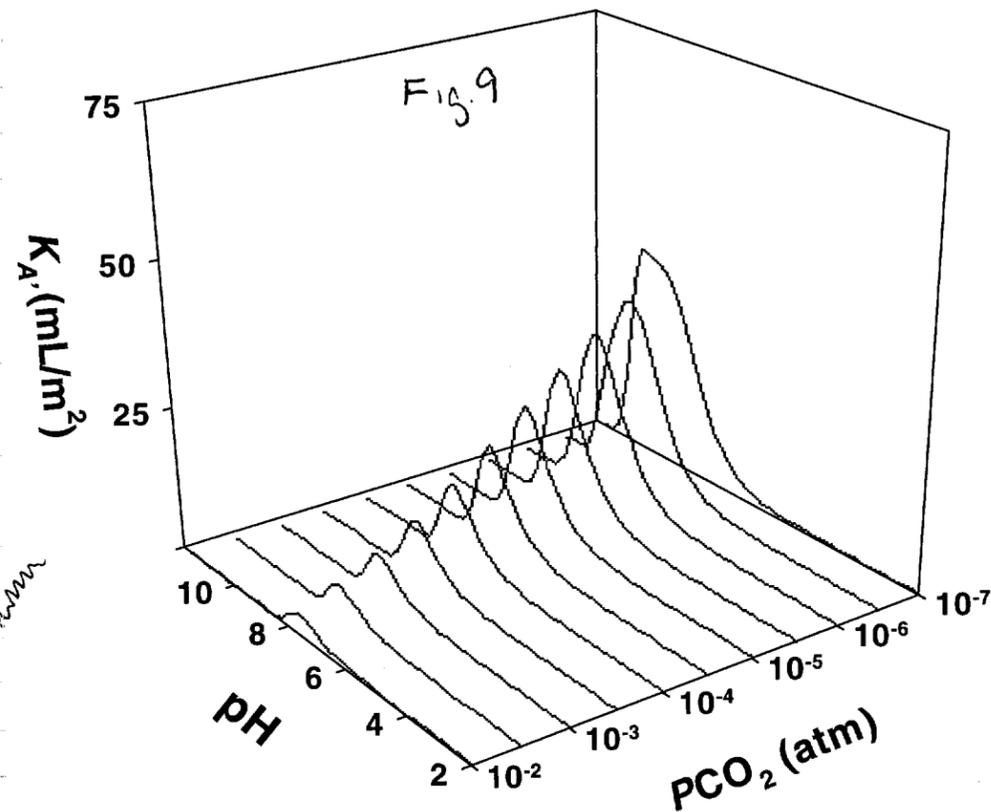
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Fig. 10, pg. 41

(where a, b, c, d, and e are constant coefficients) was found to adequately reproduce the model predicted sorption over the desired pH range for each PCO₂ (including the CO₂-free curve). Coefficient values and goodness-of-fit values for each curve are shown, and sample output is shown graphically for selected values of PCO₂. The equations used here are an example only and alternative equations could be used to improve the fit for each curve, but use of one form of equation would simplify the incorporation of the model into a TPA code by requiring only a matrix for coefficients (i.e., a set for each PCO₂ value) and one equation. Since pH is commonly reported, and PCO₂ may need conversion or calculation depending on the available data, PCO₂ was left for interpolation and/or adjustment here. This method has the advantage of using a continuous range of pH data while reducing the number of discrete points needed for the look-up table.

Parameters and summary of results for model curves at discrete PCO₂. Np(V)_{total} = 10⁻⁶ molal; M/V = 4 g/L.

PCO ₂ (atm)	Coefficients: [ln(K _A , in mL/m ²) = a+bx+cx ² +dx ³ +ex ⁴ +fx ⁵]						r ² value	pH range used for fit
	a	b	c	d	e	f		
10 ^{-2.0}	-323.7345029	151.4136753	-17.3990293	-1.7541185	0.4728224	-0.0247745	0.9999	6 - 9.25
10 ^{-2.5}	-441.4872516	226.8171288	-37.7488848	-1.2089255	0.2378357	-0.0167447	0.9999	6 - 9.25
10 ^{-3.0}	148.2265595	-173.8278793	69.4791195	-12.8694017	1.1394455	-0.0390727	0.9999	6 - 9.50
10 ^{-3.5}	604.4445148	-474.5177627	147.2075461	-22.6668262	1.7364614	-0.0529354	0.9999	6 - 9.50
10 ^{-4.0}	847.1361569	-620.1544804	180.5362481	-26.2031203	1.8992944	-0.0549789	0.9999	6 - 10.00
10 ^{-4.5}	925.7298724	-652.8079406	183.1897645	-25.6433576	1.7939710	-0.0501685	0.9999	6 - 10.25
10 ^{-5.0}	923.2318767	-632.0905821	172.1420527	-23.3803904	1.5872876	-0.0430961	0.9999	6 - 10.50
10 ^{-5.5}	672.7843206	-452.9837012	121.1472289	-16.1548188	1.0777544	-0.0287889	0.9999	6 - 11.00
10 ^{-6.0}	393.8474607	-258.6708687	67.3400912	-8.7496479	0.5711094	-0.0149989	0.9999	6 - 11.25
10 ^{-6.5}	722.6946490	-436.2310889	104.2139278	-12.3723844	0.7340464	-0.0174653	0.9978	6 - 11.50
10 ^{-7.0}	2202.1902289	-1290.5774270	299.2738666	-34.3781522	1.9602212	-0.0444424	0.9816	6 - 11.75
no CO ₂	1211.3978170	-705.8275247	161.4080394	-18.1364167	1.0036927	-0.0219067	0.9996	6 - 11.75

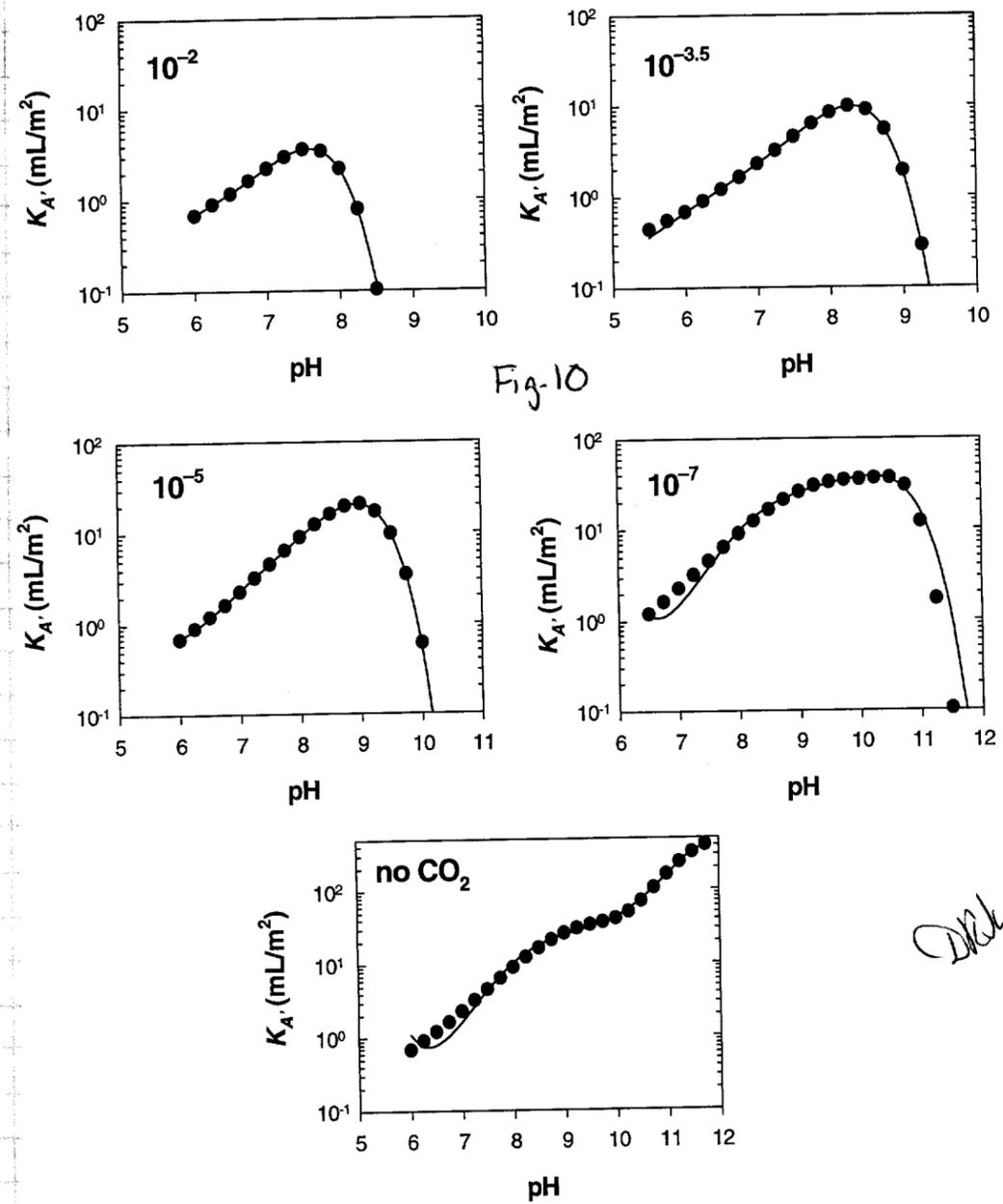
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Plot of DLM predictions of Np(V) sorption at discrete PCO_2 values. $Np(V)_{total} \sim 1 \times 10^{-6} M$, $M/V = 4 g/L$.

DLM

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Plots of polynomial fits to DLM K_A s (points) for selected discrete values of PCO_2 . Values listed in the upper left-hand corner of each plot show PCO_2 in atm for the particular data set and fit.

DLM

Example: Using the Response Surface with YM Site-Specific Data

A response surface for K_D will be ultimately be used with site-specific information. One means of incorporating this information is sampling of PDFs for pH and log PCO_2 that are based on site-specific ambient groundwater chemistry data. Because pH and log PCO_2 are linked through the carbonate aqueous chemistry, selection of one parameter provides constraints on the value of the other.

Descriptive statistics of measured groundwater chemical parameters (from Turner, 1998).

	pH (standard units)	Log PCO_2 (atm)
Mean	7.83	-2.50
Standard Error	0.02	0.03
Median	7.8	-2.45
Mode	7.8	-2.34
Standard Deviation	0.45	0.54
Kurtosis	1.75	3.73
Skewness	0.43	-1.30
Range	3.3	4.311
Minimum	6.3	-5.08
Maximum	9.6	-0.77
Count	460	460

For the purposes of this demonstration, the sampling routine in Microsoft Excel Version 5.0 was used to sample 460 values of pH based on the distribution statistics defined in section 4.2. The correlation between pH and log PCO_2 ($\rho_{\text{Log } PCO_2, \text{pH}} = -0.83$) was then used to define a value for log PCO_2 .

A simple procedure for sampling jointly normal random variables was implemented to demonstrate how correlation between pH and the common logarithm of PCO_2 can be used to preserve correlation among sampled K_D values for selected radionuclides. The vector of mean values for the pH and the logarithm of

PCO_2 is

$$\boldsymbol{\mu} = \begin{pmatrix} \mu_{\text{pH}} \\ \mu_{\text{PCO}_2} \end{pmatrix} = \begin{pmatrix} 7.83 \\ -2.50 \end{pmatrix},$$

and the vector of sample standard deviations for the pH and the logarithm of PCO_2 is

$$\boldsymbol{\sigma} = \begin{pmatrix} \sigma_{\text{pH}} \\ \sigma_{\text{PCO}_2} \end{pmatrix} = \begin{pmatrix} 0.45 \\ 0.54 \end{pmatrix},$$

where bold symbols indicate vectors or matrices of appropriate dimension. Consider a column vector \mathbf{v} consisting of a pair of independent standard normal deviates and a fixed transformation matrix \mathbf{L} such that

$$\boldsymbol{\xi} = \mathbf{L}\mathbf{v},$$

where $\boldsymbol{\xi}$ is jointly normal with zero mean and covariance matrix $\boldsymbol{\Omega}$. Using the correlation coefficient between pH and the logarithm of PCO_2 and the standard deviations estimated from the raw data, the covariance matrix is

$$\boldsymbol{\Omega} = \begin{pmatrix} \sigma_{\text{pH}}^2 & \rho_{\text{PCO}_2, \text{pH}} \sigma_{\text{pH}} \sigma_{\text{PCO}_2} \\ \rho_{\text{pH}, \text{PCO}_2} \sigma_{\text{PCO}_2} \sigma_{\text{pH}} & \sigma_{\text{PCO}_2}^2 \end{pmatrix} = \begin{pmatrix} 0.1985 & -0.2007 \\ -0.2007 & 0.2943 \end{pmatrix}.$$

where the actual numbers will vary slightly depending on rounding. By definition

$$E(\boldsymbol{\xi}\boldsymbol{\xi}^T) = E(\mathbf{L}\mathbf{v}\mathbf{v}^T\mathbf{L}^T) = \mathbf{L}E(\mathbf{v}\mathbf{v}^T)\mathbf{L}^T = \boldsymbol{\Omega},$$

but

$$E(\mathbf{v}\mathbf{v}^T) = \mathbf{I},$$

the identity matrix, therefore

$$\mathbf{L}\mathbf{L}^T = \boldsymbol{\Omega},$$

where \mathbf{L} is the lower triangle of the Cholesky decomposition (Atkinson, 1978) of the covariance matrix $\boldsymbol{\Omega}$. Once \mathbf{L} is determined, jointly normal random values of the pH and the logarithm of PCO_2 are constructed by first generating the vector of independent standard normal deviates, \mathbf{v} , and performing the transformation

$$\boldsymbol{\xi} = \mathbf{L}\mathbf{v} + \boldsymbol{\mu},$$

or

$$\begin{pmatrix} \xi_{\text{pH}} \\ \xi_{\text{PCO}_2} \end{pmatrix} = \begin{pmatrix} 0.45 & 0 \\ -0.4504 & 0.3024 \end{pmatrix} \begin{pmatrix} v_{\text{pH}} \\ v_{\text{PCO}_2} \end{pmatrix} + \begin{pmatrix} 7.83 \\ -2.50 \end{pmatrix}.$$

Comparison of sampled values with measured values indicates that the sampled ranges for both pH and PCO_2 are slightly narrower than the observed ambient groundwater chemistry, particularly at lower values of log PCO_2 . This may be due in part to the assumption of a log-normal distribution of log PCO_2 , when there are a small number (≤ 12) of low PCO_2 waters in the 460 measured groundwaters investigated by Turner (1998).

Sorption coefficients were estimated for Np(V) sorption on montmorillonite ($A' = 10 \text{ m}^2/\text{g}$) by interpolating the K_D response surface defined by the polynomials given above. As might be expected given the narrower ranges in sampled pH and log PCO_2 relative to the measured values, the K_D range estimated using the response surface is narrower than the range calculated directly from the measured water chemistries (Turner, 1998). A 3-D representation of response surface-calculated K_A versus pH and PCO_2 shows a similar trend to that estimated above. The K_D s estimated using the response surface tend to be lower than those calculated directly by the DLM, and in this sense the use of a response surface in PA would be a conservative approach. Larger numbers of sampled pH and PCO_2 might provide more sampling of extreme conditions, but the assumption of a normal distribution does not take into account observed extremes.

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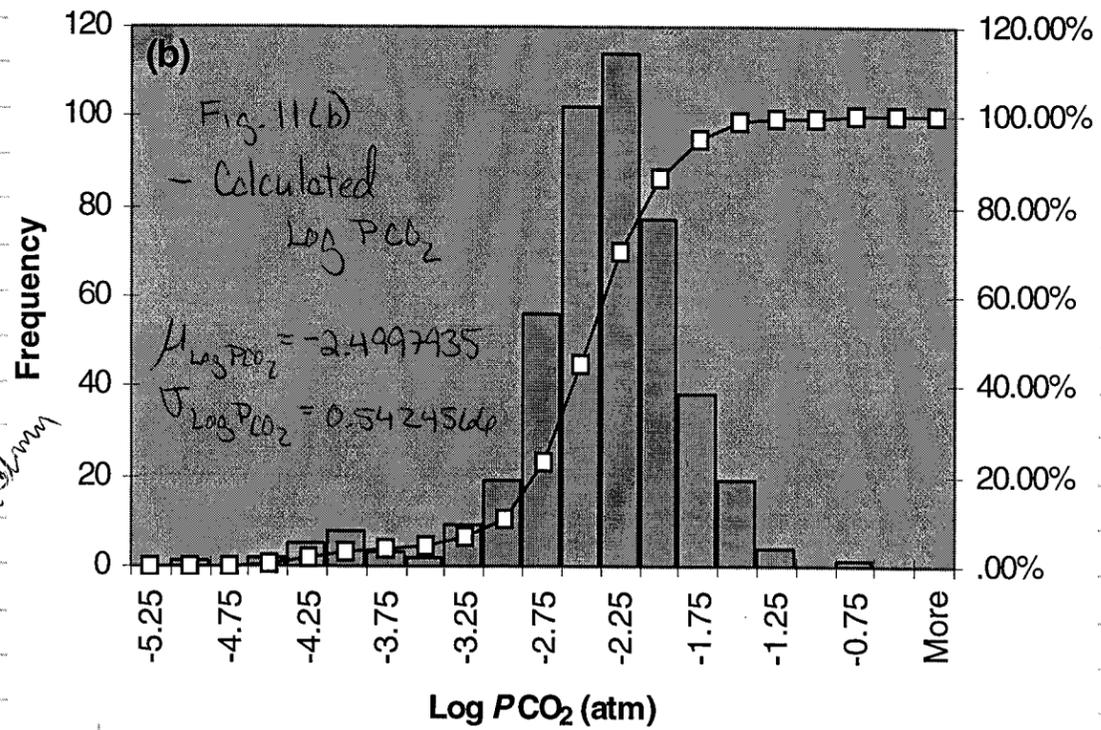
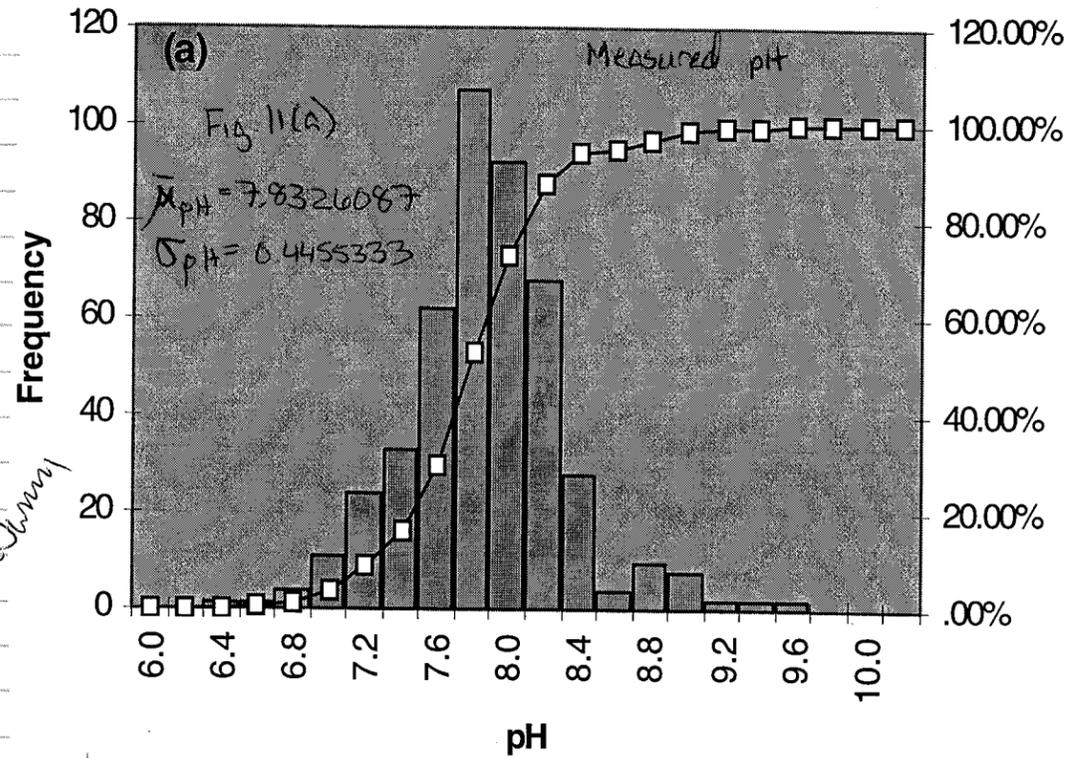
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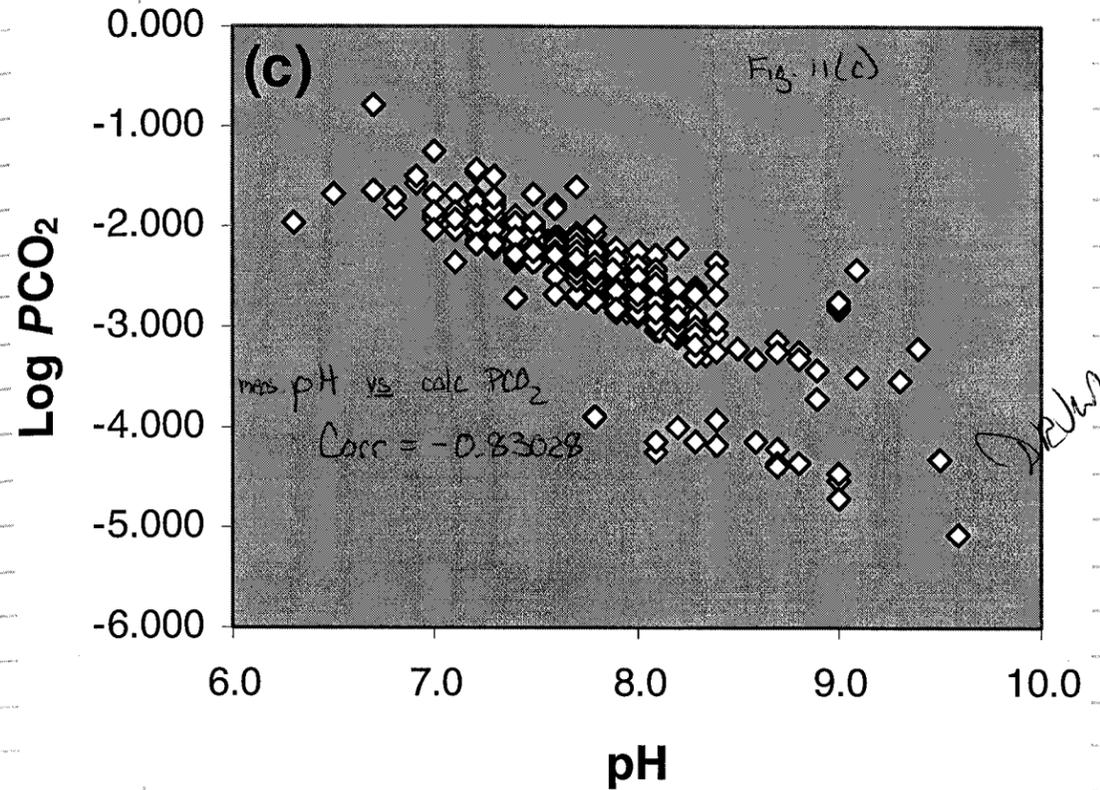
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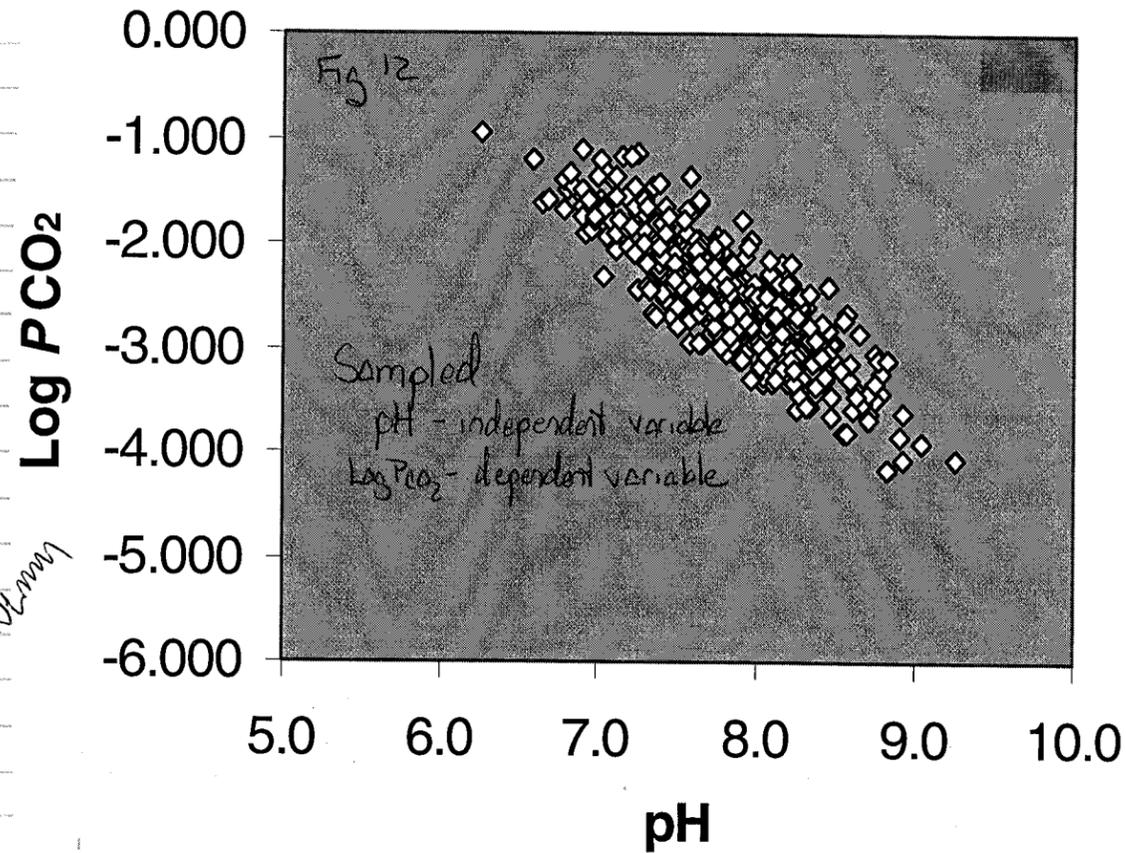


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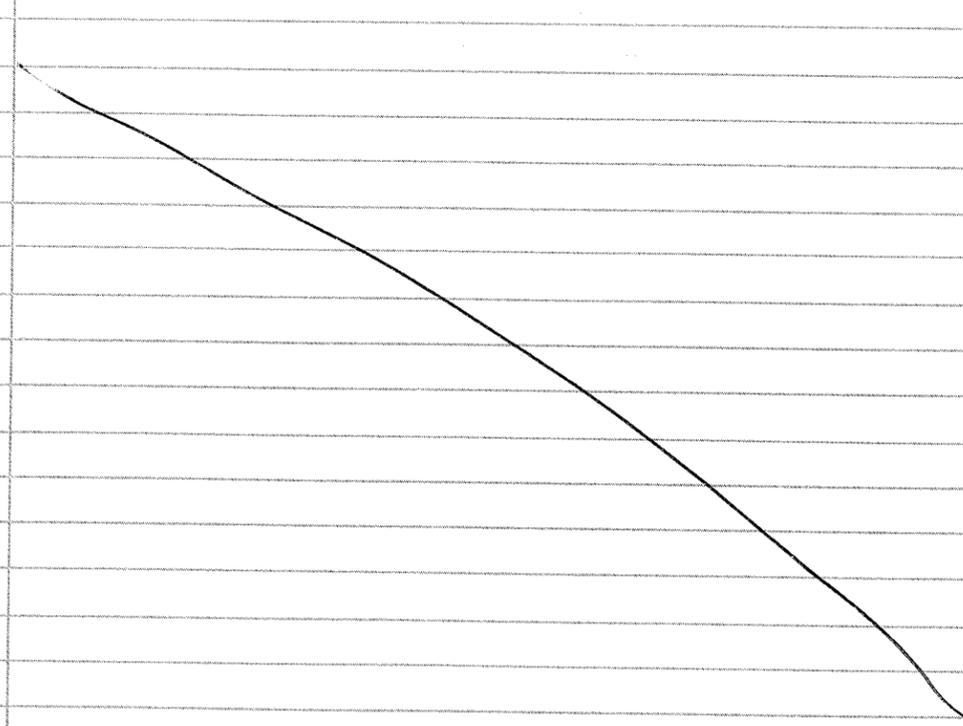


Critical geochemical parameter values for 460 saturated zone regional groundwater samples collected in the vicinity of Yucca Mountain, Nevada (original data from Perfect et al., 1995; see Turner, 1998 for screening criteria). (a) pH, (b) Log PCO₂ calculated using major ion chemical analyses and MINTEQA2, Version 3.11, and; (c) pH versus Log PCO₂.

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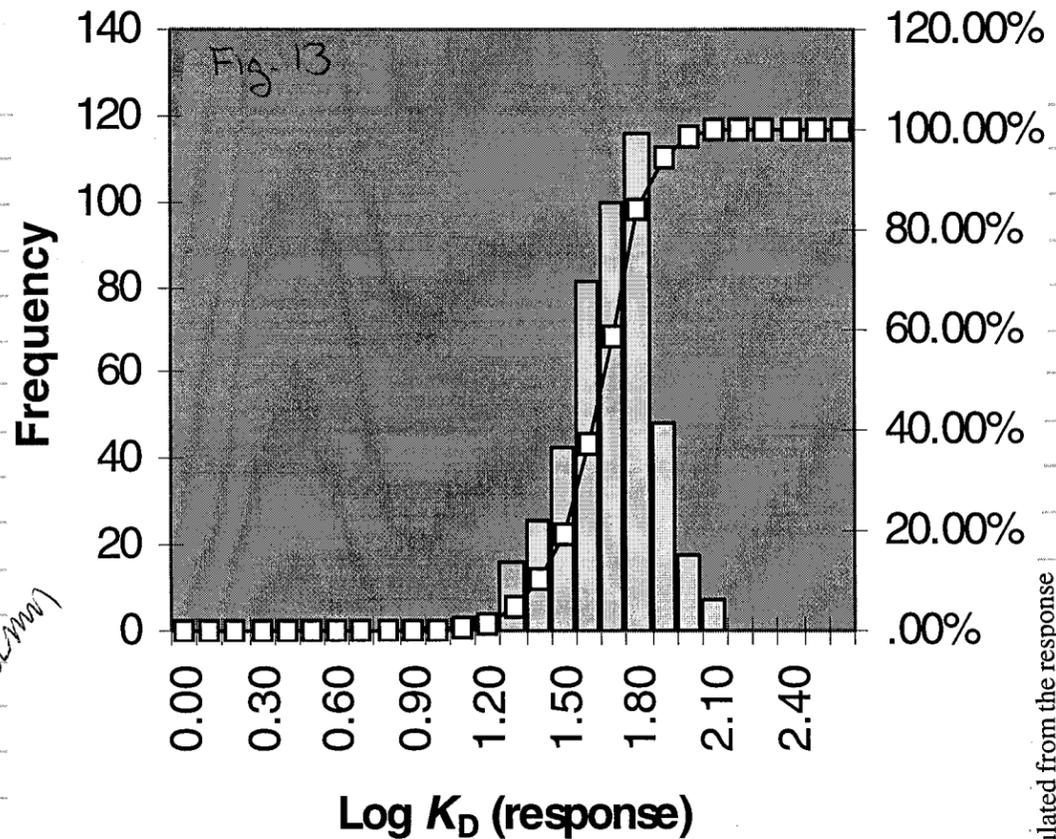


Sampled pH versus Log PCO_2 calculated using the population statistics provided in Turner (1998).



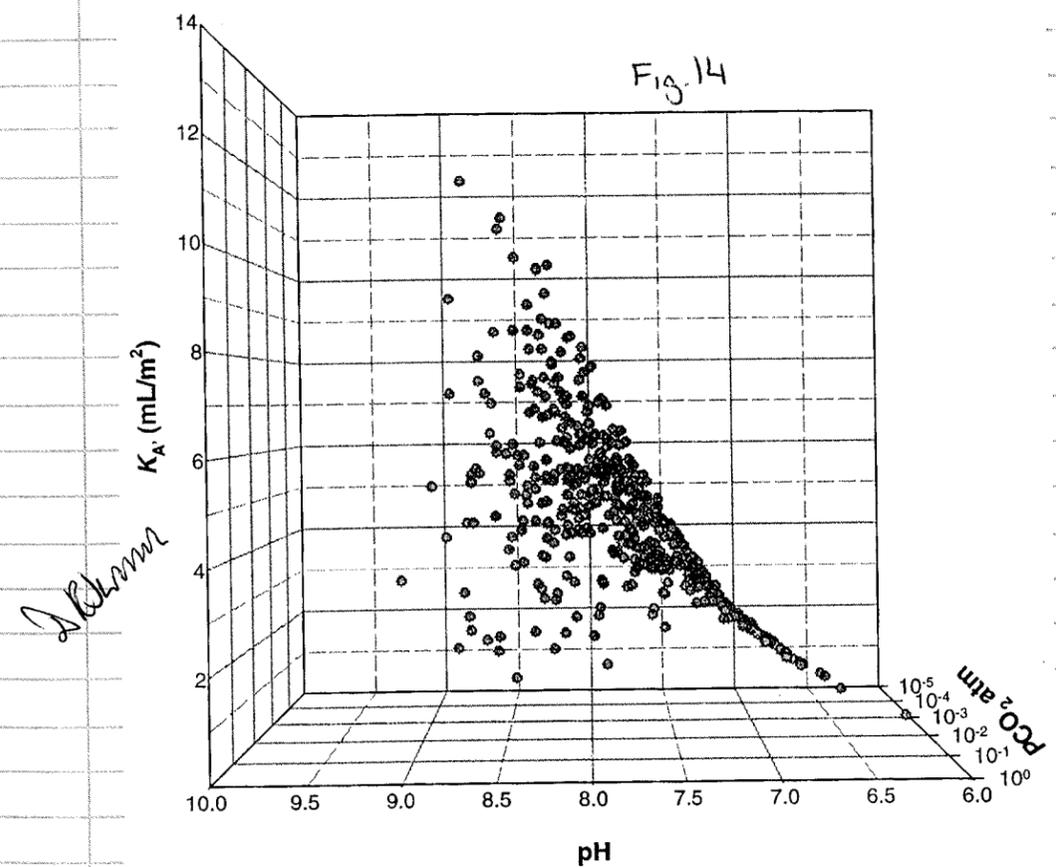
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$Log K_D$ calculated from the response surface defined by the polynomial equations.

DRJ/mmj



Three-dimensional representation of K_A shown as a function of pH and PCO_2 , calculated from the response surface defined by the polynomial equations.

DRJ/mmj

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DEJ
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Calculating Response Surfaces

	A	B	C	D	E	F	G	H
1	[Np]=1e-6; M/V=4 g/L		A'=9.7 m ² /g for montmorillonite					
2	pH	no CO ₂	%Np sol	% Np sorbed	Np sol	Np solid	Kd	Ka
3	2	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
4	2.25	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
5	2.5	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
6	2.75	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
7	3	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
8	3.25	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
9	3.5	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
10	3.75	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
11	4	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
12	4.25	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
13	4.5	552	99	1	8.81E-07	8.9E-09	2.525253	0.260335
14	4.75	552	98.9	1.1	8.8E-07	9.79E-09	2.780586	0.286658
15	5	552	98.8	1.2	8.79E-07	1.07E-08	3.036437	0.313035
16	5.25	552	98.6	1.4	8.78E-07	1.25E-08	3.549696	0.365948
17	5.5	552	98.3	1.7	8.75E-07	1.51E-08	4.323499	0.445722
18	5.75	552	97.9	2.1	8.71E-07	1.87E-08	5.362615	0.552847
19	6	552	97.4	2.6	8.67E-07	2.31E-08	6.673511	0.687991
20	6.25	552	96.6	3.4	8.6E-07	3.03E-08	8.799172	0.907131
21	6.5	552	95.5	4.5	8.5E-07	4.01E-08	11.7801	1.214444
22	6.75	552	94	6	8.37E-07	5.34E-08	15.95745	1.645098
23	7	552	91.8	8.2	8.17E-07	7.3E-08	22.33115	2.302181
24	7.25	552	88.8	11.2	7.9E-07	9.97E-08	31.53153	3.250673
25	7.5	552	84.9	15.1	7.56E-07	1.34E-07	44.46408	4.583925
26	7.75	552	79.9	20.1	7.11E-07	1.79E-07	62.89111	6.48362
27	8	552	73.9	26.1	6.58E-07	2.32E-07	88.29499	9.102577
28	8.25	552	67.4	32.6	6E-07	2.9E-07	120.9199	12.46597
29	8.5	552	60.9	39.1	5.42E-07	3.48E-07	160.509	16.54732
30	8.75	552	54.8	45.2	4.88E-07	4.02E-07	206.2044	21.25818
31	9	552	49.7	50.3	4.42E-07	4.48E-07	253.0181	26.08434
32	9.25	552	45.9	54.1	4.09E-07	4.81E-07	294.6623	30.37756
33	9.5	552	43.2	56.8	3.84E-07	5.06E-07	328.7037	33.88698
34	9.75	552	41.2	58.8	3.67E-07	5.23E-07	356.7961	36.7831
35	10	552	38.7	61.3	3.44E-07	5.46E-07	395.9948	40.82421
36	10.25	552	33.9	66.1	3.02E-07	5.88E-07	487.4631	50.25393
37	10.5	552	26.9	73.1	2.39E-07	6.51E-07	679.368	70.03794
38	10.75	552	19.6	80.4	1.74E-07	7.16E-07	1025.51	105.7227
39	11	552	13.8	86.2	1.23E-07	7.67E-07	1561.594	160.9891
40	11.25	552	9.8	90.2	8.72E-08	8.03E-07	2301.02	237.2186
41	11.5	552	7.4	92.6	6.59E-08	8.24E-07	3128.378	322.5132
42	11.75	552	6	94	5.34E-08	8.37E-07	3916.667	403.7801
43								
44	pH	co2 10-7	%Np sol	% Np sorbed				
45	2	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
46	2.25	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
47	2.5	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
48	2.75	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
49	3	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
50	3.25	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
51	3.5	552	99.2	0.8	8.83E-07	7.12E-09	2.016129	0.207848
52	3.75	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
53	4	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
54	4.25	552	99.1	0.9	8.82E-07	8.01E-09	2.270434	0.234065
55	4.5	552	99	1	8.81E-07	8.9E-09	2.525253	0.260335
56	4.75	552	98.9	1.1	8.8E-07	9.79E-09	2.780586	0.286658
57	5	552	98.8	1.2	8.79E-07	1.07E-08	3.036437	0.313035
58	5.25	552	98.6	1.4	8.78E-07	1.25E-08	3.549696	0.365948
59	5.5	552	98.3	1.7	8.75E-07	1.51E-08	4.323499	0.445722
60	5.75	552	97.9	2.1	8.71E-07	1.87E-08	5.362615	0.552847

$$K_D = \frac{8.01 \times 10^{-9}}{8.82 \times 10^{-7}} \times \frac{1000}{9.7} = 2.27 \text{ mL/g}$$

$$K_A = K_D / A' = \frac{2.27 \text{ mL/g}}{9.7 \text{ m}^2/\text{g}} = 0.23 \text{ mL/m}^2$$

MINTEQA2 results

Np ~ 1e-6
 MN_{TOT} = 4/g/L
 no CO₂

DKmm