MODELING ARSENITE SORPTION ON

NEW JERSEY SOILS

by

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ABSTRACT

During the 1990s, the New Jersey Department of Environmental Protection and Energy funded the University of Delaware's Department of Civil and Environmental Engineering to conduct a study of the sorption of six heavy metals: cadmium, copper, lead, chromium, arsenic, and mercury, on fifteen soils found in NJ. The results of the arsenite, AsIII, adsorption were used to develop a model to predict sorption behavior as a function of soil characteristics. A new model fitting procedure was developed that successfully estimated the dependence of the Freundlich isotherm parameters on soil characteristics and produced a model that included the effects of soil characteristics on both the Freundlich constant and exponent. This new procedure expands the ability to determine the effects of soil characteristics on sorption isotherm parameters. The resulting arsenite sorption model demonstrates the importance of soil metal oxides in determining the extent of arsenite sorption.

Chapter 1

INTRODUCTION

Inorganic arsenic poses a threat to human and environmental health. It has been listed as the toxin of highest priority by the Agency for Toxic Substances and Disease Registry on the 2007 Comprehensive, Environmental, Response, Compensation and Liability Act Priority List of Hazardous Substances (Hughes et al., 2011). Ingestion of acute amounts of arsenic results in poisoning while long term effects manifest on the skin (World Health Organization, 2010). Arsenic, a known carcinogen, has been linked to health issues such as cardiovascular illnesses and diabetes (Sparks, 2003). Arsenic transport occurs both in areas with high and low levels of natural arsenic in soils through weathering as well as introduction of historical anthropogenic uses, including medicine, pigments, and pesticides (Hughes et al., 2011; Sparks, 2003). The movement of arsenic through groundwater can introduce this pollutant directly to drinking water sources, as well as through crops that have been irrigated with contaminated water. Of the arsenic compounds, arsenite, AsIII is more toxic than arsenate, AsV (Hughes et al., 2011). Arsenic contamination limits are currently being established and are difficult to set with a limited amount of research on its fate and transport (Dias et al., 2009; Sparks, 2003). Studying the sorption of arsenic onto soils can determine the mobility of the toxin and potential for introduction to groundwater systems.

Arsenic sorption can be linked to the complexation of arsenic to aluminum and iron oxides in a soil, as well as the oxidation and complexation of arsenite to arsenate

by manganese oxide, and clay components (Carbonell-Barrachina et al., 2000; Goldberg and Glaubig, 1988; Goldberg, 2002; Smith et al., 1999; Sparks, 2003). Other soil characteristics influencing sorption include pH and cation exchange capacity (Carbonell-Barrachina et al., 2000).

Carbonnell-Barrachina et al. (2000) reported that most arsenite sorption evaluations are limited to specific clays, oxides, and other ideal components. Correlations between sorption parameters and soil properties have been studied for arsenic and cadmium sorption (Dias et al., 2009; Lee et al., 1996). A common procedure to evaluate correlations is to use a multi-component linear regression model between the soil parameters and the soil partition coefficients can be used to determine the stability constants of the binding constant or other isotherm parameters (Lee et al., 1996). Dias et al. followed a similar procedure, using a forward linear regression of soil parameters to determine Langmuir isotherm adsorption capacity of arsenite (2009). Their model achieved an R2 of 0.895 and determined that the soil properties relevant to arsenite sorption are, in order of importance, iron oxides, organic matter, aluminum oxides, clay, cation exchange capacity, surface area, and manganese oxides (Dias et al., 2009). Historically, arsenite sorption experiments on pure minerals resulted in a Langmuir isotherm. However, it was noted that arsenite sorption on soils rather than pure minerals follows a Freundlich model, consistent with the results of sorption of other anions onto soil sediments (Elkhatib et al., 1984). Similar to the work of Dias et al. (2009), Elkhatib et al. (1984) conducted a stepwise regression to correlate the soil characteristics to the linearized Freundlich isotherm parameters for arsenite sorption on five West Virginian soils. This model was successful in describing the sorption of arsenite on the five different soils. The authors noted that in

addition to adsorption, the oxidation of AsIII to the adsorption of AsV, and precipitation, could be removing arsenite from the equilibrium phase (Elkhatib et al., 1984). The study notes that there a positive correlation between iron and manganese oxides and the Freundlich parameter. The best prediction can be achieved with a linear regression of organic carbon, iron oxide, and pH (Elkhatib et al., 1984). The inclusion of soil parameters into the prediction of adsorption capacity aligned with the understanding of arsenic sorption as described in literature.

This study evaluated the sorption behavior of arsenite onto a variety of soil types with different characteristics, representative of the soils found in New Jersey. Each soil characteristic was investigated by evaluating a variety of regression models. A new parameter estimation method was introduced to determine the dependency of Freundlich parameters on soil characteristics.

Chapter 2

MATERIALS & METHODS

During the 1990s, the New Jersey Department of Environmental Protection and Energy funded the University of Delaware's Department of Civil and Environmental Engineering to develop a Sorption Report of six different heavy metals, including AsIII and AsV, on 15 different soils found in NJ. The metals evaluated were cadmium, mercury, lead, cooper, chromium, CrIII and CrVI, and arsenic, AsIII and AsV. The experiments included determination of adsorption edges and isotherms, desorption experiments, and kinetic investigations. The collected soils, referred to as A – O, were determined to be representative of the major soil types of New Jersey (Lee et al., 1996). The soils were air dried, sieved through a 2 mm sieve, and homogenized before determining the soils' physical and chemical properties (Dias et al., 1997; Lee et al., 1996). The adsorption/desorption experiments were conducted following standard protocol for a batch method sorption experiment at the soil's natural pH and ionic strength of 0.01 M, equilibrated for 48 hours, with the colorimetric method for arsenic analysis to obtain the sorption isotherms (Dias et al., 2009).

Chapter 3

RESULTS

Soil properties are summarized in Table 1 including total and amorphous oxide concentration, pH, and organic matter content (Dias et al., 1997; Lee et al., 1996). Additional soil characteristics, including the soil names can be found in Appendix A.

Table 1. Soil Properties as Reported by Lee et al., 1996

Soil Letter	pH	Organic Carbon (g/kg)	Total Metal Oxide (mg/kg)			Amorphous Metal Oxide (mg/kg)		
			Iron	Aluminum	Manganese	Iron	Aluminum	Manganese
A	5.7	12.8	4.723	3.697	0.077	1.035	0.714	0.025
B	5.1	30.7	3.859	4.541	0.057	1.563	1.321	0.013
C	5.1	49.9	2.834	3.866	0.243	0.773	1.447	0.065
D	4.7	4.6	0.753	4.878	0.018	0.076	0.12	0.0003
E	5.6	11	4.915	4.71	0.062	0.435	0.355	0.011
F	4.8	7	2.065	1.165	0.042	0.578	0.105	0.012
G	5.2	1.2	3.538	3.697	0.037	0.519	0.442	0.009
H	6.4	13.9	1.553	2.009	0.028	0.178	0.052	0.001
I	6.0	18	4.307	4.372	0.116	1.117	0.574	0.034
J	4.2	2.9	0.657	0.321	0.008	0.344	0.096	0.0002
K	4.7	7.5	4.851	6.397	0.077	0.743	0.593	0.022
L	4.7	28.4	3.538	5.047	0.096	0.832	1.181	0.028
M	5.8	3.5	2.674	3.191	0.018	0.699	0.461	0.005
N	6.0	16.8	5.556	7.072	0.136	0.9	0.684	0.038
O	6.2	13.3	3.282	5.047	0.023	0.736	0.388	0.004

Figure 1 represents the correlation between the soil parameters. There is a general correlation between the oxides, although soil C, or soil D for iron and aluminum, do not follow the trend. There is a week positive correlation between organic matter and iron and aluminum oxide, but there is a strong positive correlation between organic matter and total manganese oxide content. The soils' pH ranges from

4.2 to 6.4, total iron oxide content ranges from 0.66 to 5.56%, total aluminum oxide content from ranges 0.32 to 7.1%, and total manganese oxide content ranges from less than 0.01 to 0.24%. Soil C is a soil of particular interest as it contains the highest concentration of organic matter at 49.9 g/kg, as well as the highest concentration of total manganese oxide at 0.24%.
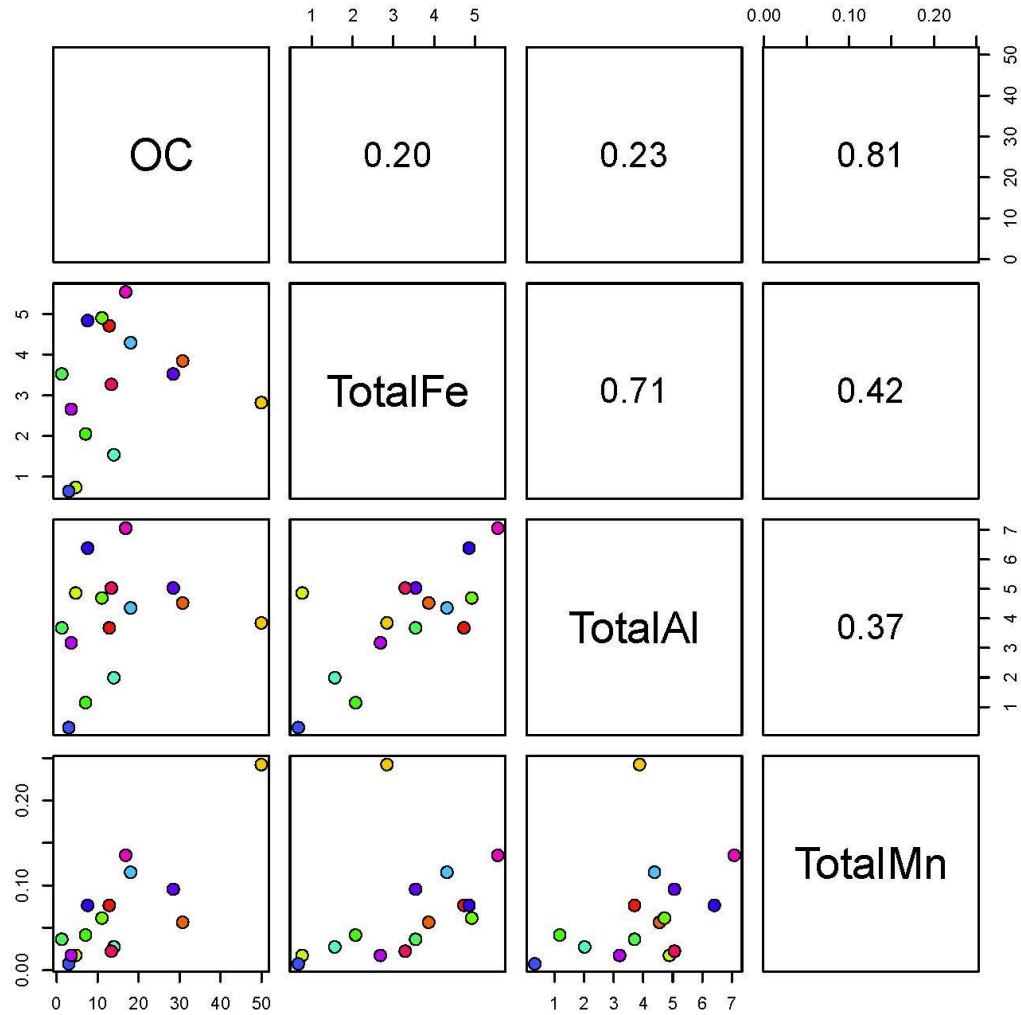
Figure 1. Pairs plot of total metal oxides and organic carbon content of NJ soils. Data and units in Table 1. X axis label at the head of the column, Y axis label at the right of the row. Upper panels contain the Pearson's correlation coefficient. If the lower panel has row index i and column index j, the correlation coefficient is in the j,i panel.

The sorption data are plotted using linear and log scales, in Figures 2 and 3, respectively. In Dias et al.'s (2009) evaluation of this dataset, they suggested that the sorption follows and L-shaped curve, indicating Langmuir isotherm behavior. The linear plots, Figures 2a and 2b, suggest some flattening at the higher concentrations. However, the log-log plot, Figure 3, indicates that the arsenite isotherms do not level off at a maximum level of sorbed arsenite so that a Langmuir isotherm could be fit reliably. However, the sorption data can be fit using a Freundlich isotherm, which are straight lines in Figure 3.
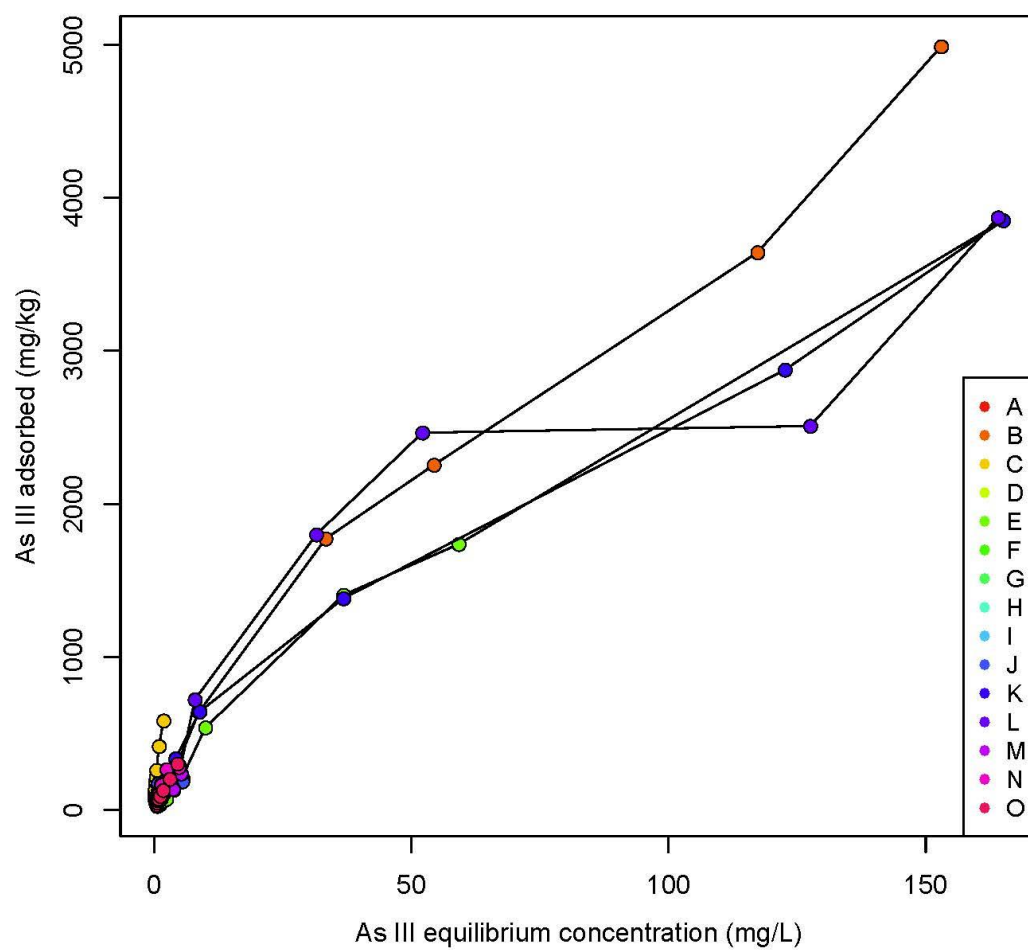
Figure 2a. Arsenite adsorption isotherm on a linear scale, with equilibrium
concentrations ranging from 0-150 mg/L. Soils identified in legend and
Table 1.
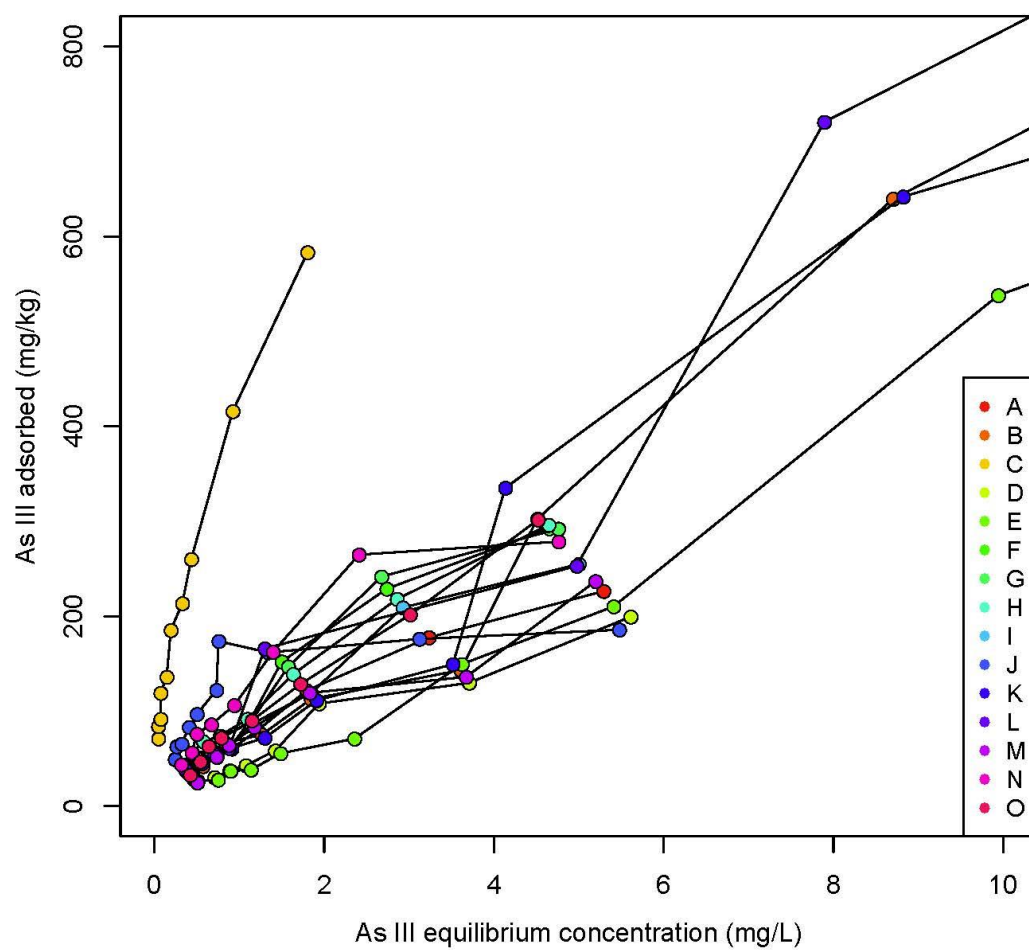
Figure 2b. Arsenite adsorption isotherm on a linear scale, with equilibrium
        concentrations ranging from 0-10 mg/L. Soils identified in legend and
        Table 1.
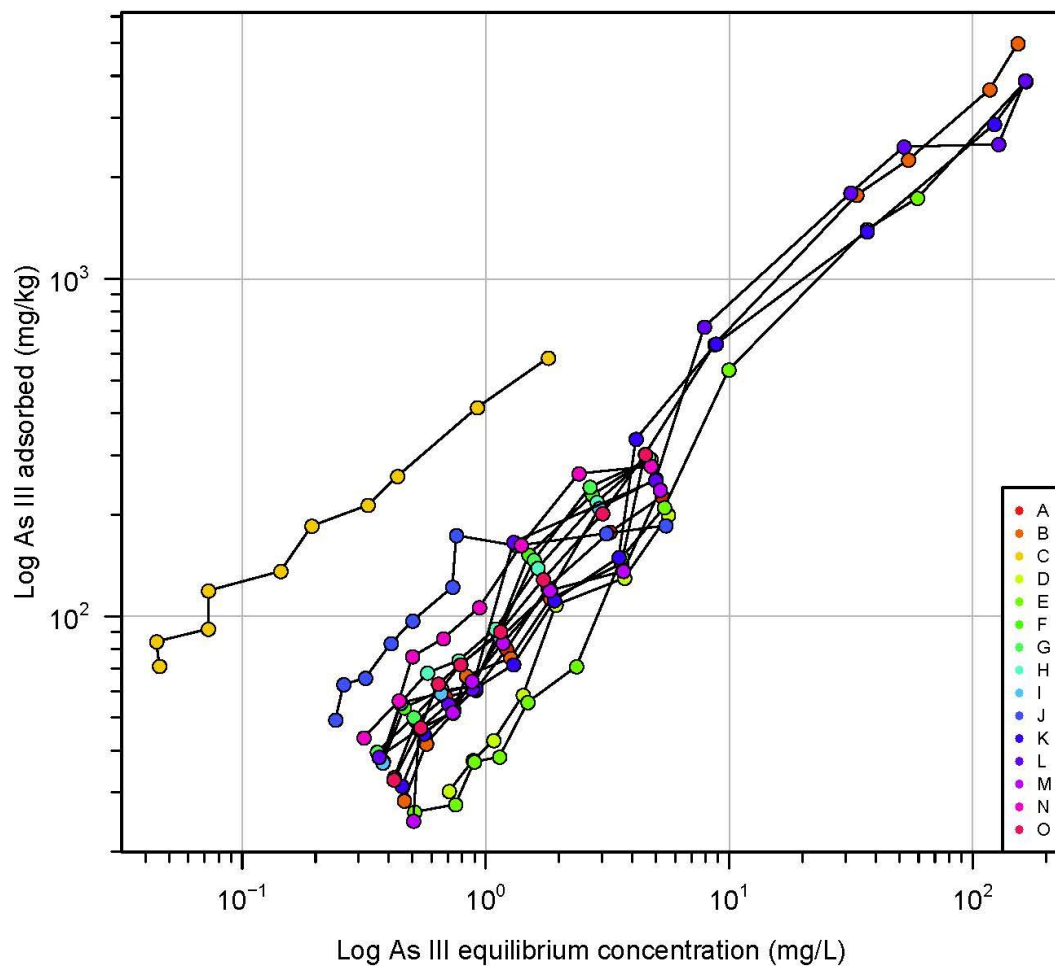
Figure 3. Arsenite adsorption isotherm on a log-log scale. Soils identified in legend and Table 1.

The Freundlich isotherm can be expressed as

$$q = K_f c^v \tag{1}$$

or

$$\log q = \log K_f + v \log c \tag{2}$$

where q = concentration of arsenite on the soil (mg/kg), c = concentration of arsenite in the liquid phase (mg/L), and K_f and v are system specific sorption constants. In order to relate the soil characteristics to the isotherm parameters, the Freundlich isotherm constants, K_f and v, were fit to the data for each of the fifteen soils for arsenite sorption. The results are reported in Table 2. In order to evaluate a relationship between the isotherm parameters and the soil characteristics, K_f and v are plotted against organic carbon and metal oxide content in Figure 4. There appear to be no strong correlations between the soil parameters and the isotherm coefficients, indicating little the relationship of the isotherm parameters and the soil properties.

Table 2. Freundlich Paramters for Individual Soil Fits. Units are given in Table 1.

Soil	Intercept log K_f	Slope, v	Multiple R^2	RMSE
A	1.853	0.739	0.981	0.038
B	1.826	0.877	0.989	0.081
C	2.623	0.542	0.984	0.037
D	1.635	0.915	0.961	0.056
E	1.613	0.934	0.987	0.082
F	1.943	0.843	0.959	0.061
G	1.954	0.835	0.981	0.042
H	1.968	0.774	0.984	0.036
I	1.908	0.734	0.966	0.051
J	2.079	0.411	0.774	0.096
K	1.839	0.813	0.982	0.094
L	1.944	0.781	0.970	0.130
M	1.803	0.777	0.908	0.087
N	2.053	0.726	0.955	0.059
O	1.908	0.869	0.986	0.035
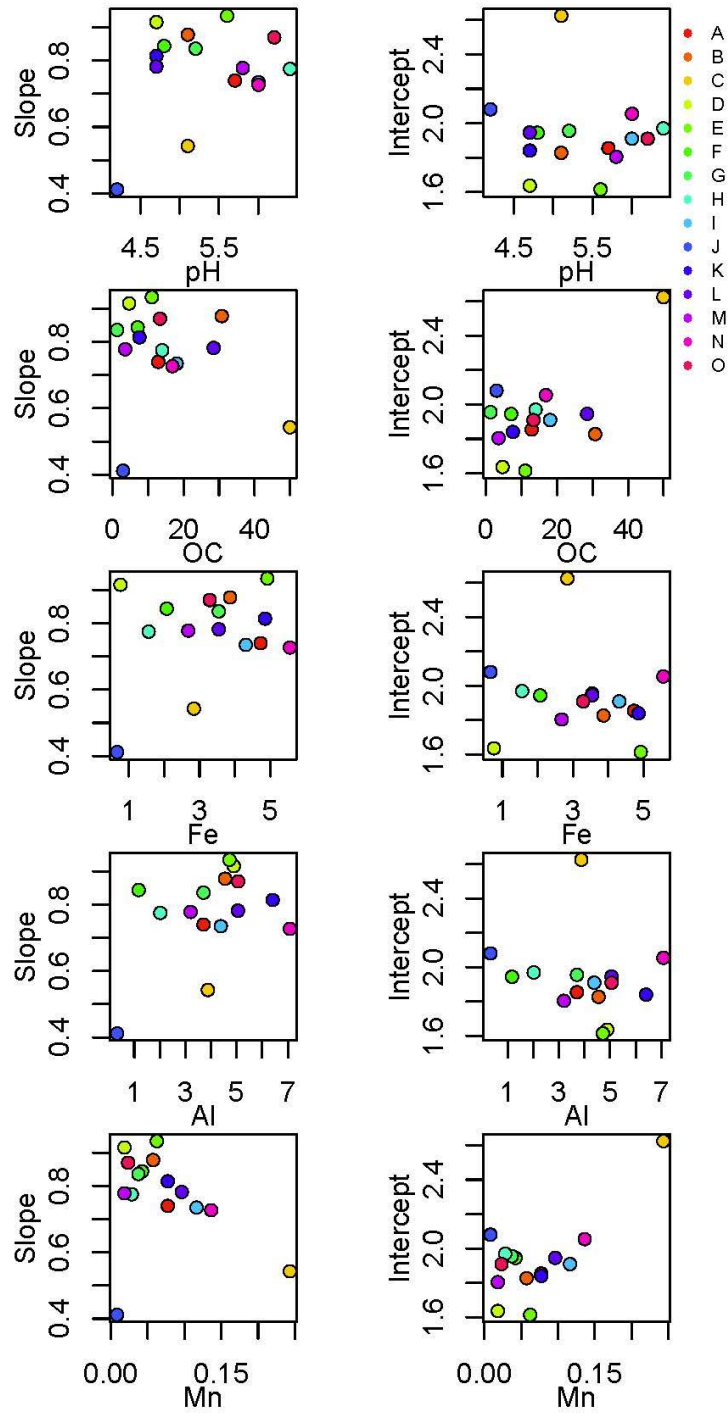
Figure 4. Fitted Freundlich parameters, K_f and v, plotted against pH, organic carbon, and iron, aluminum, and manganese oxide concentration of each soil. Soils and units identified in legend and Table 1.

A more refined fitting procedure is to include the soil characters into the Freundlich parameters directly. The isotherm parameters can be expressed in terms of the soil characteristics as follows. Let b_0 be the constant isotherm parameter and b_i be the coefficients for the relationship between $\log K_f$ and the soil characteristic W_i. Let m_0 be the constant isotherm parameter and m_i be the coefficients for the relationship between v_i and soil characteristic W_i. Then,

$$\log K_f = b_0 + \sum b_i W_i \tag{3}$$

and

$$v = m_0 + \sum m_i W_i \tag{4}$$

The isotherm Equation (2) becomes

$$\log q = b_0 + \sum b_i W_i + \log c \, (m_0 + \sum m_i W_i) \tag{5}$$

The fitting procedure is to use the entire soil data set and fit the sorbed concentrations q using Equation (5). The parameters are estimated jointly using multiple linear regression. For a fit using two soil parameters, e.g. W_1 = total soil iron concentration and W_2 = total soil manganese concentration, there would be six isotherm parameters to estimate: b_0, b_1, b_2, and m_0, and m_1, and m_2 for the $\log K_f$ and v equations, respectively.

Metal oxides and organic matter have been identified as soil characteristics that influence the strength of arsenite sorption (Carbonell-Barrachina et al., 2000; Goldberg and Glaubig, 1988; Goldberg, 2002; Smith et al., 1999; Sparks, 2003). There appears to be only small correlation between the other soil characteristics, pH, clay, and cation exchange capacity, and isotherm parameters despite their reported importance in many other models (Carbonell-Barrachina et al., 2000). In order to make a complete evaluation, all 22 combinations of organic matter and total iron, aluminum, and manganese oxides or amorphous oxides were used to fit the isotherm

parameters (Equation 5). The root mean square error of the residuals, RMSE = log predicted q – log observed q, of each combination was less than 0.25. Figure 5 compares the residuals for each soil type for the three models with the smallest RSME <0.145. The equations for these models are in Table 4 and Appendix B.
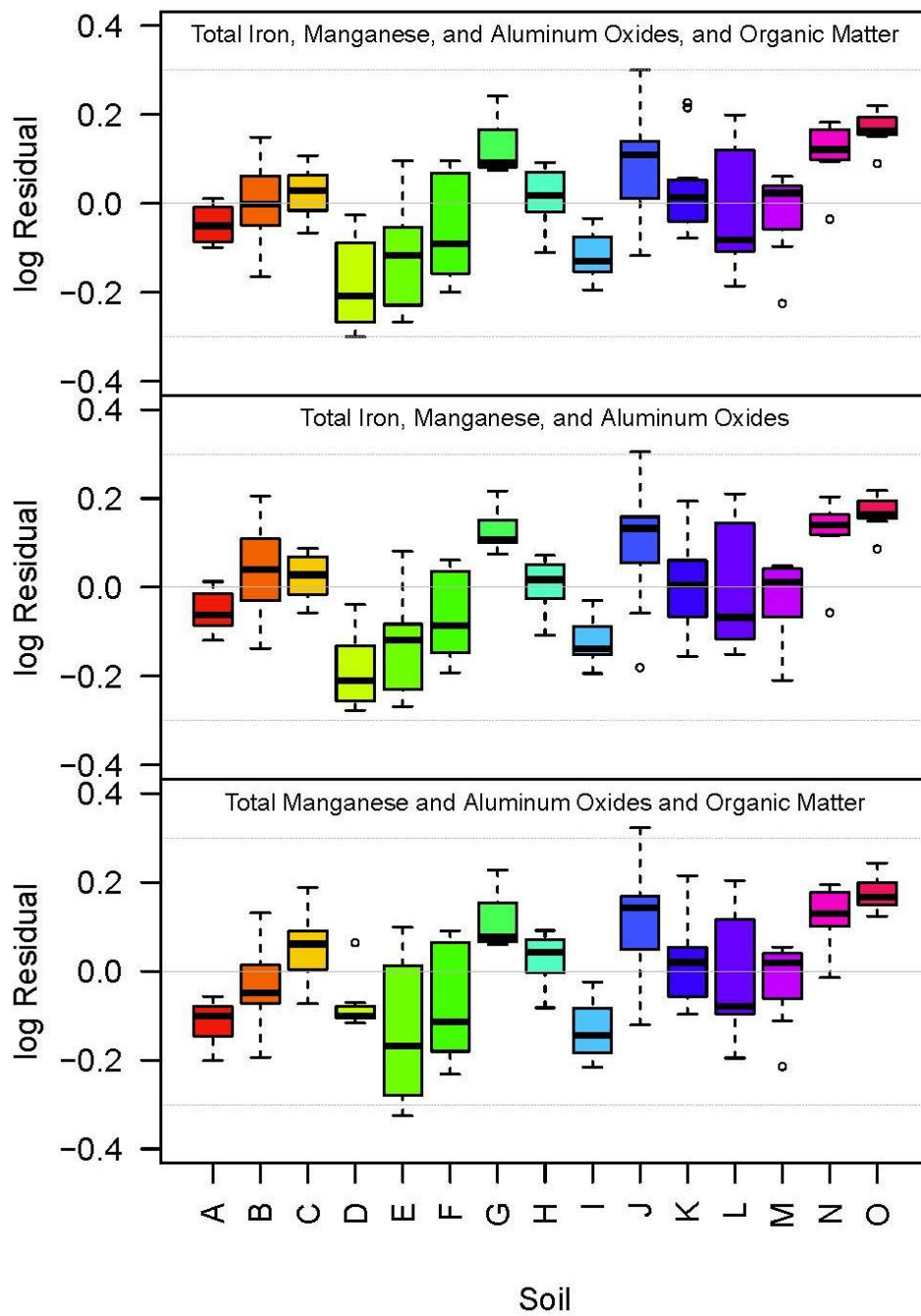
Figure 5. Boxplots of log residuals of fits with the lowest RMSE. Variables included in the regressions are identified at the top of each plot. See Appendix B for the isotherm equations.

The models with the smallest RMSE involved combinations of total metal oxides and organic matter concentrations. Figures 6 and 7 show the predicted versus observed sorbed concentration of arsenite for all fifteen soils. Figure 6 includes total metal oxides and organic matter concentrations. Figure 7 includes only the total metal oxides. Both models are able to predict the amount of arsenite sorbed to within approximately a factor of 2. Table 3 summarizes the statistics of these two models. The isotherm parameter equations for both models are in Table 4. The model that includes all three metal oxides and organic matter concentrations (Figure 6) is not significantly different from the model using only the metal oxide concentrations (Figure 7), and it requires two fewer parameters.

Table 3. Summary Statistics of Model Options

Soil Parameters	Multiple R^2	RMSE
$TotalFe_2O_3$+$TotalAl_2O_3$+$TotalMnO_2$+OC	0.932	0.140
$TotalFe_2O_3$+$TotalAl_2O_3$+$TotalMnO_2$	0.928	0.143

Table 4. Linearized Freundlich Parameter Equations

Metal Oxides & Organic Matter	Log $K_f =$	2.02 – 0.0546 [TotFe] – 0.0338 [TotAl] + 3.47 [TotMn] – 0.000825[OC]
	$v=$	0.470 + 0.0651 [TotFe] + 0.0271 [TotAl] – 2.32 [TotMn] + 0.00533[OC]
Metal Oxides	Log $K_f =$	2.03 – 0.0546 [TotFe] – 0.0374 [TotAl] + 3.38 [TotMn]
	$v=$	0.570 + 0.0473 [TotFe] + 0.275 [TotAl] – 1.36 [TotMn]
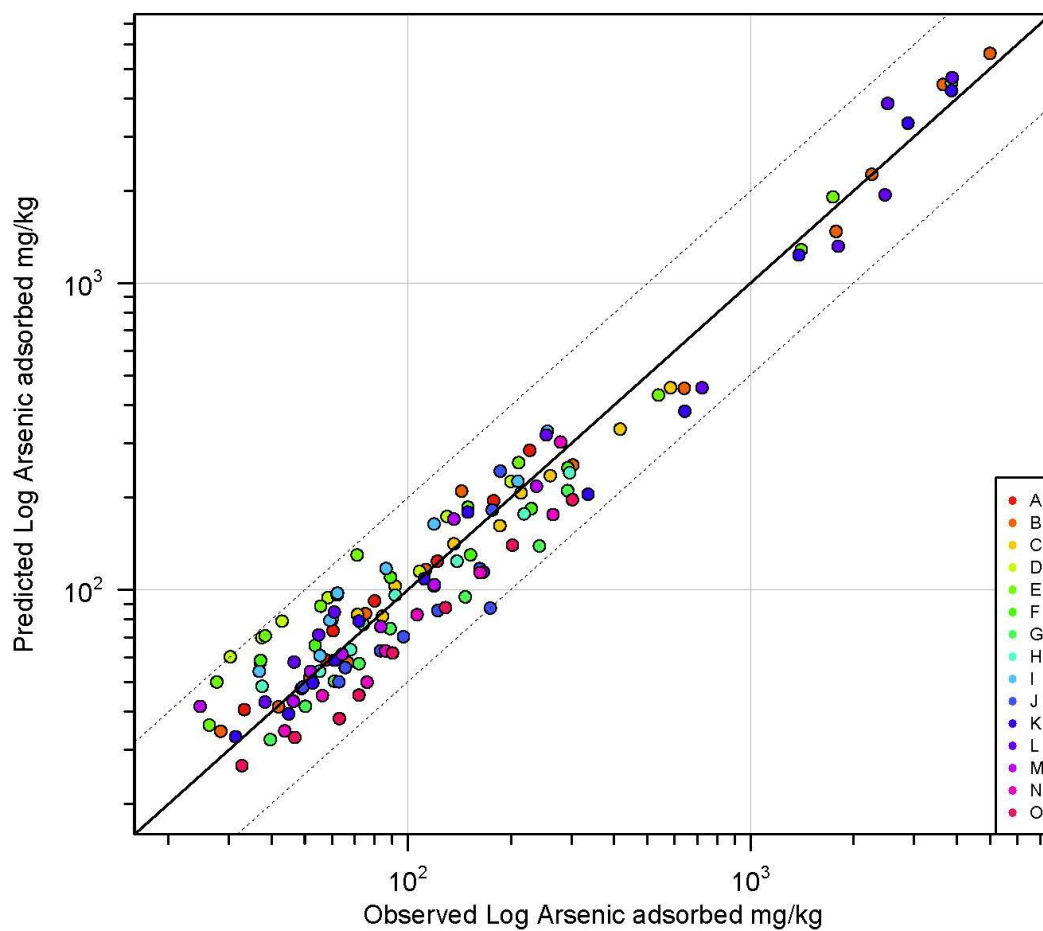
Figure 6a. Predicted versus observed soil arsenite concentration. Freundlich model
parameters fit with total metal oxide and organic matter concentrations.
See Table 4 for the isotherm equation.
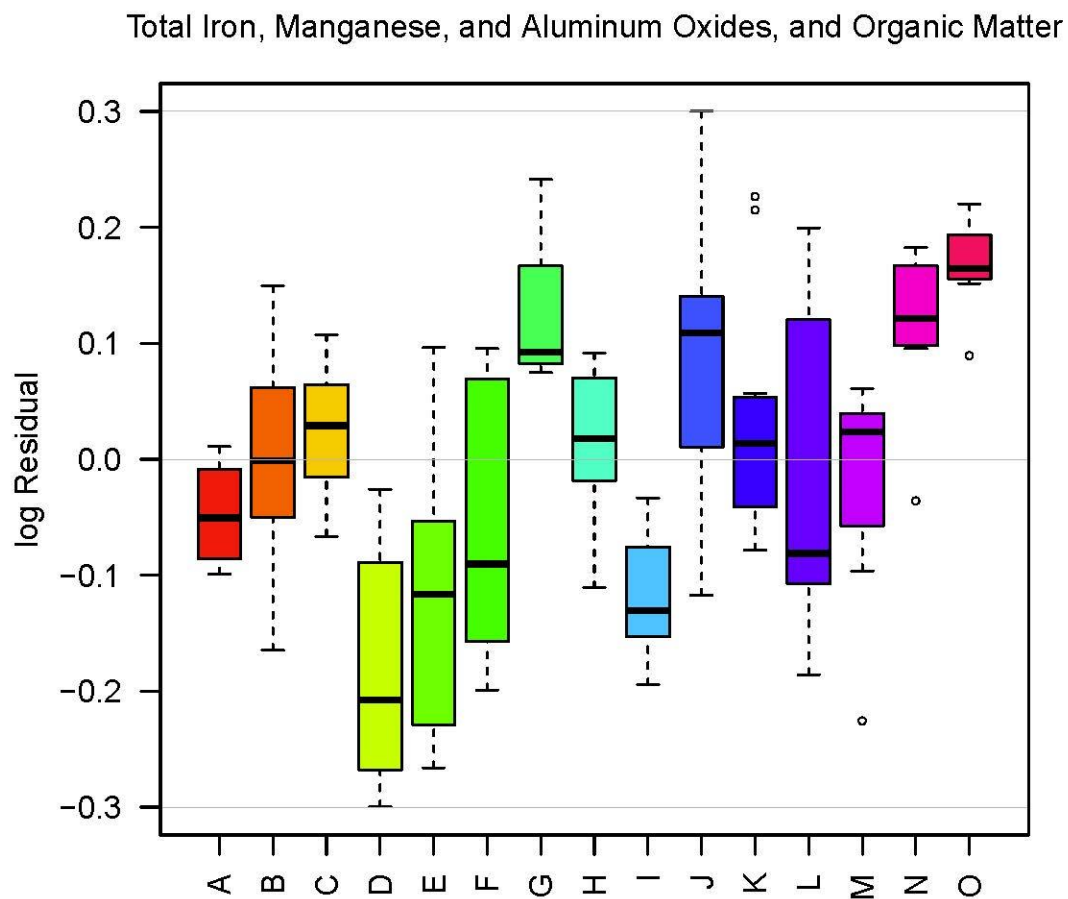
Figure 6b. Box plot of residuals = log predicted – log observed concentrations. Freundlich isotherm parameters fit with total metal oxide and organic matter concentrations. See Table 4 for the isotherm equation.
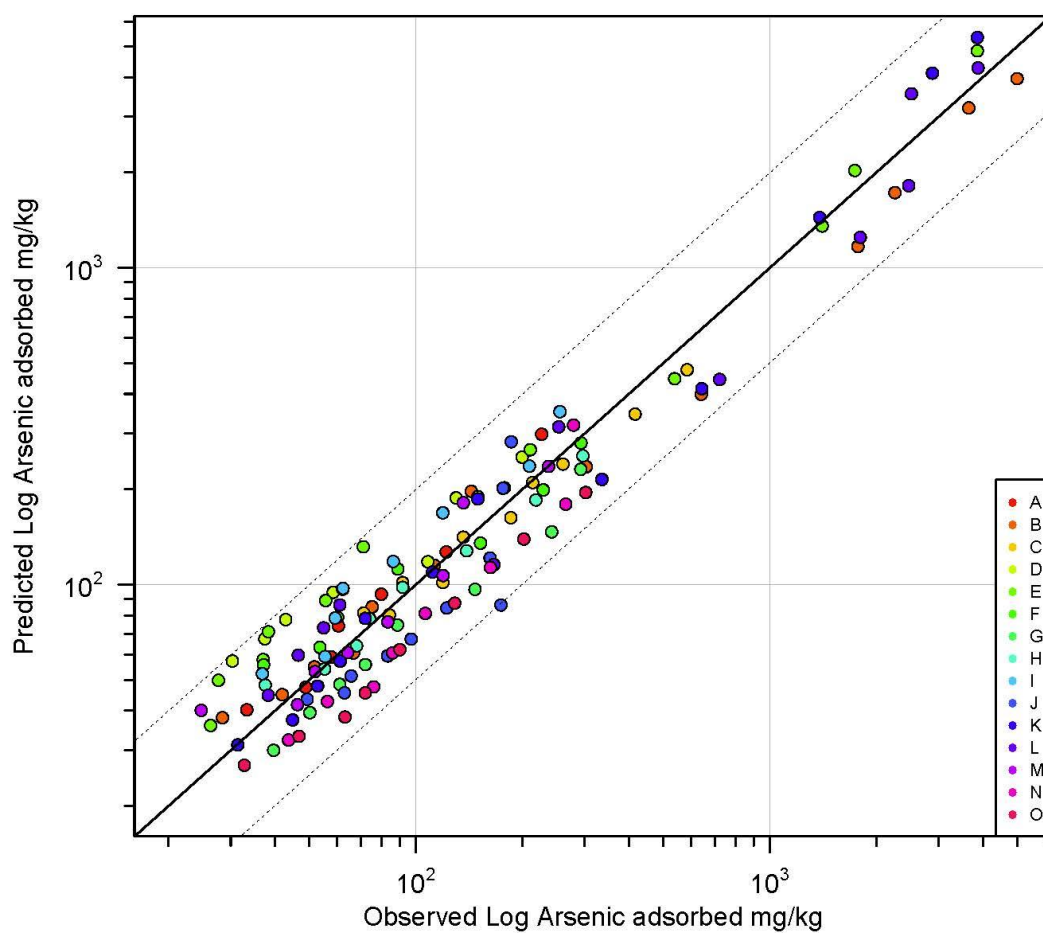
Figure 7a. Predicted versus observed soil arsenite concentration. Freundlich model
parameters fit with total metal oxide concentrations. See Table 4 for the
isotherm equation.

Total Iron, Manganese, and Aluminum Oxides
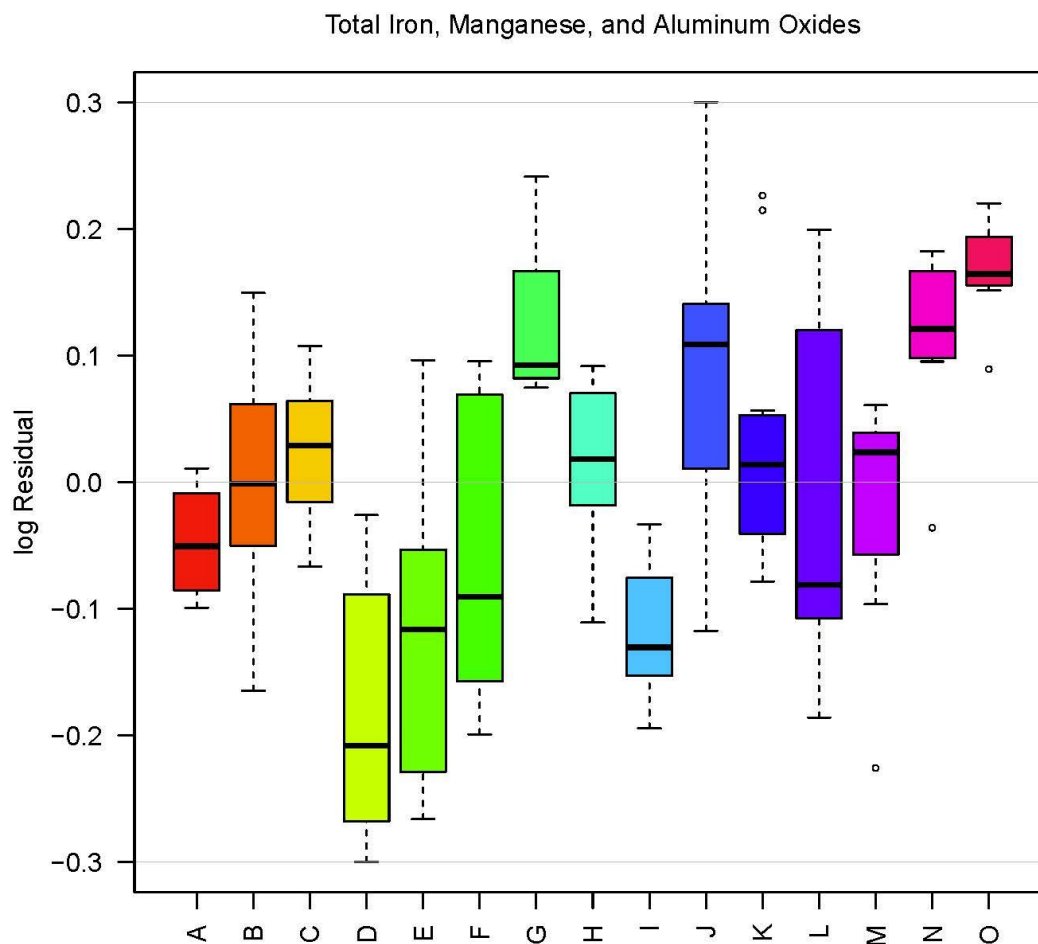
Figure 7b. Box plot of residuals = log predicted – log observed concentrations.
Freundlich isotherm parameters fit with total metal oxide concentrations.
See Table 4 for the isotherm equation.

Comparing the residual boxplots of these models, Figures 6b and 7b, indicates that the model using all the total metal oxides (Figure 7) predicts arsenite sorption onto soil D with less error, without compromising the fit of another soil. Therefore,it is the model of choice.

It has been suggested that the sorptive metal oxide phase in soils is the amorphous metal oxides rather than the total metal oxides (Dzombak and Morel, 1990). However, the models containing total metal oxides performed better than those using amorphous oxide content. Figure 8 shows the fit using all three amorphous oxides and organic carbon, which is the model that includes amorphous oxides and has the smallest RMSE value. Although the model using amorphous metal oxides concentrations was expected to better represent the sorption of arsenite, Figure 8 shows that the sorption on two soils were not as well predicted as by total metal oxide content. Figure 9 summarizes the correlation between the total and amorphous oxide concentrations. The only strong correlation between total and amorphous concentrations are for manganese oxide.

In order to evaluate the influence of each soil characteristic on the isotherm parameters, the isotherm parameters have been calculated as a function of a single metal oxide concentration while holding all other metal oxide concentrations at the median values, indicated by the points on Figure 10. Log K_f and v have an inverse relationship with respect to soil concentrations, as previously seen by fitting the values independently for each soil type in Figure 4. The coefficients of the metal oxide concentrations that determine the slopes of the lines in each plot are listed in Table 4. All three metal oxide concentrations in the model equation for log K_f indicate a negative relationship between this parameter and increasing metal oxide content. Iron

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and aluminum oxide have a negative relationship with log K_f while manganese oxide has a strong positive relationship. For the v model, iron and aluminum oxides concentrations have positive coefficients while manganese oxide has a large negative coefficient. Note that the slopes of the manganese oxide parameter models are much larger than either iron or aluminum oxide. However, the content of manganese oxide in the soils is a fraction of that of either iron or aluminum oxide, which accounts for the larger slopes.
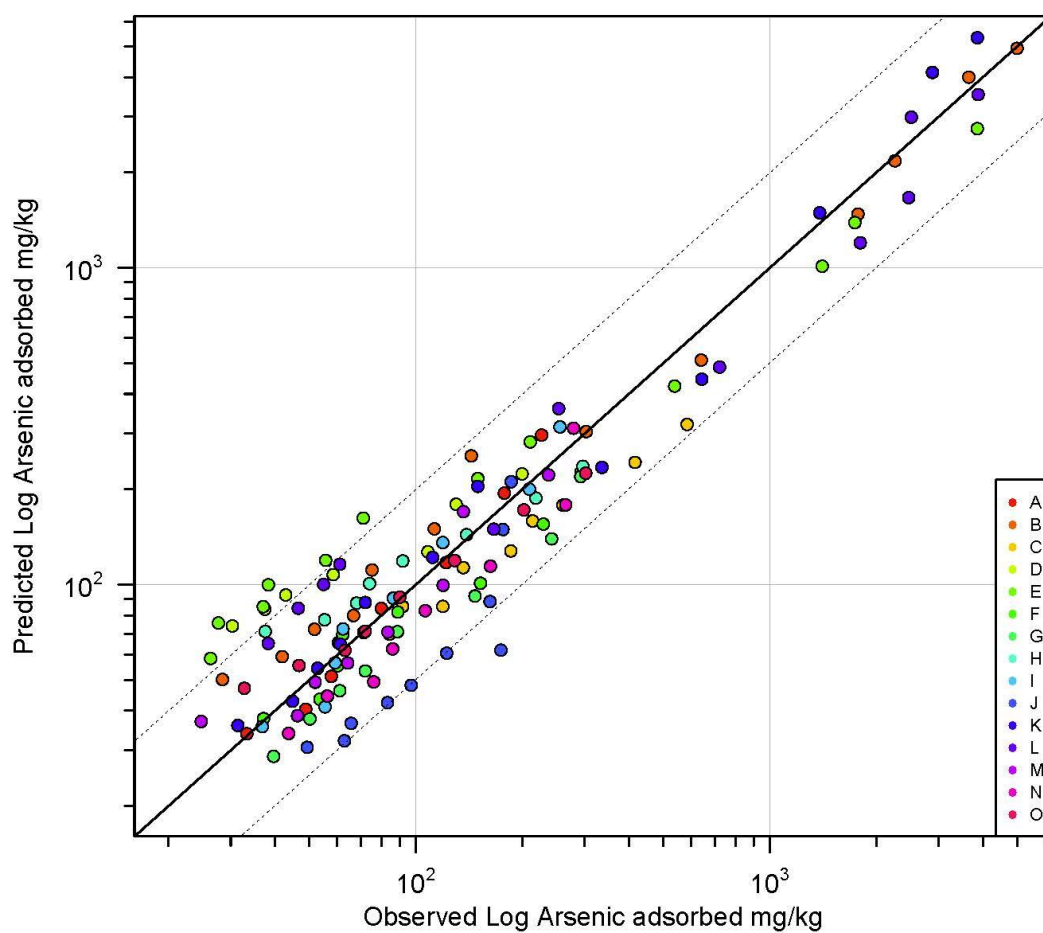
Figure 8a. Predicted versus observed soil arsenite concentration. Freundlich model parameters fit with amorphous metal oxide and organic matter concentrations. See Appendix B for the isotherm equation.
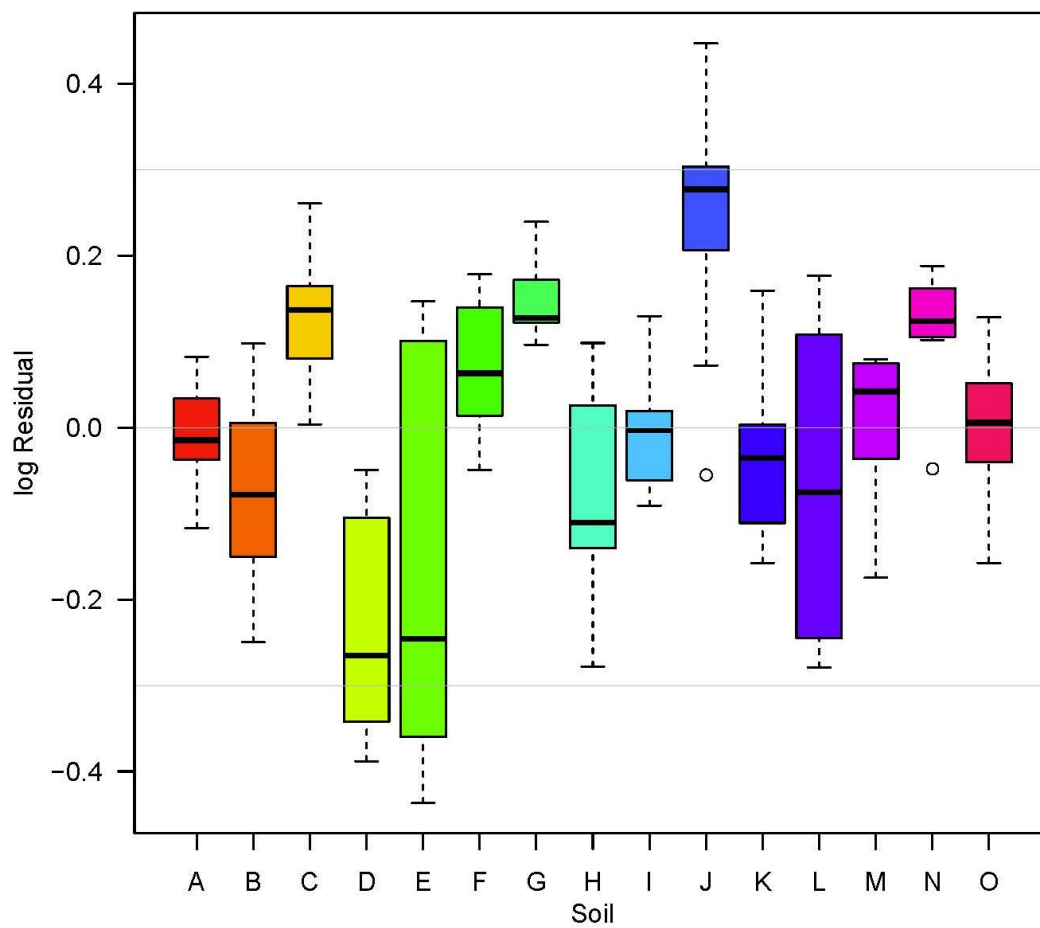
Figure 8b. Box plot of residuals = log predicted – log observed concentrations. Freundlich isotherm parameters fit with total metal oxide concentrations. See Appendix B for the isotherm equation.
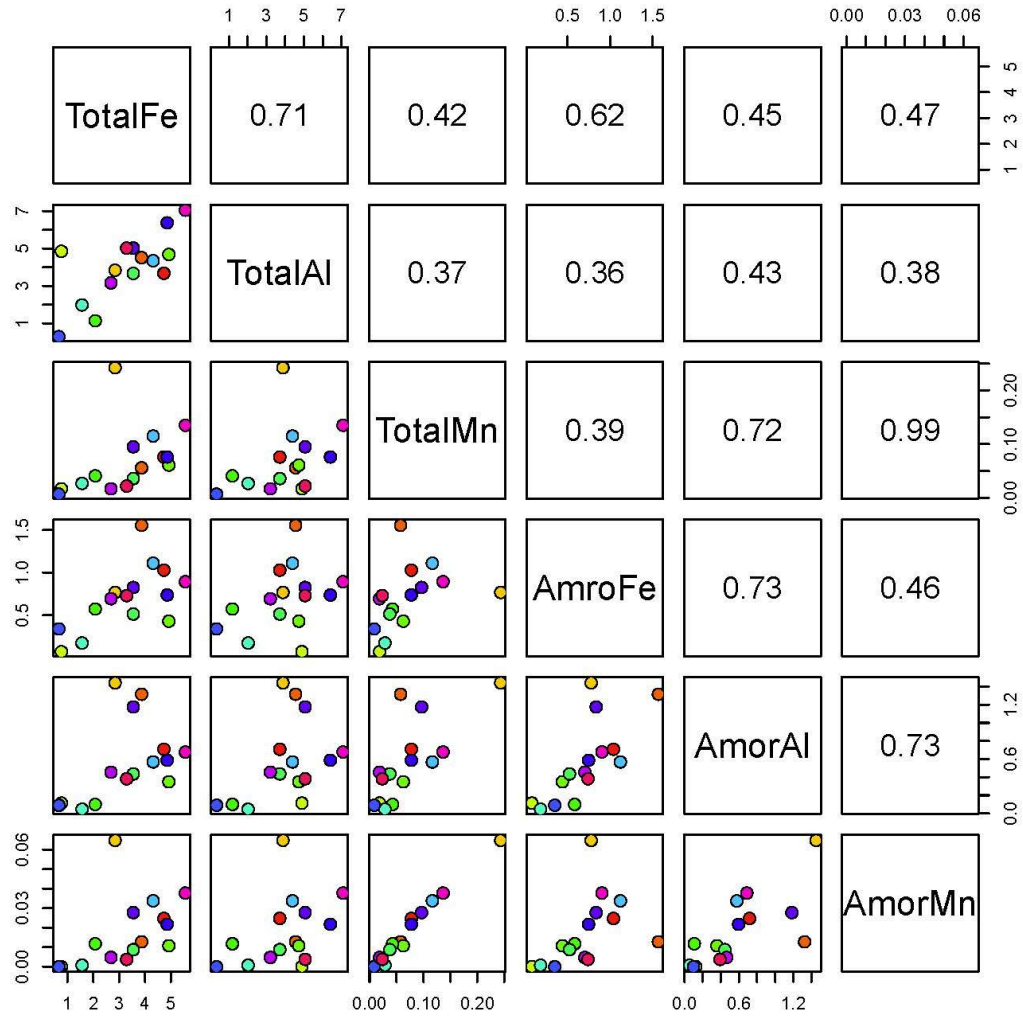
Figure 9. Pairs plot of total metal oxides and amorphous metal oxide content of NJ soils. Data and units in Table 1. X axis label at the head of the column, Y axis label at the right of the row. Upper panels contain the Pearson's correlation coefficient. If the lower panel has row index i and column index j, the correlation coefficient is in the j,i panel.
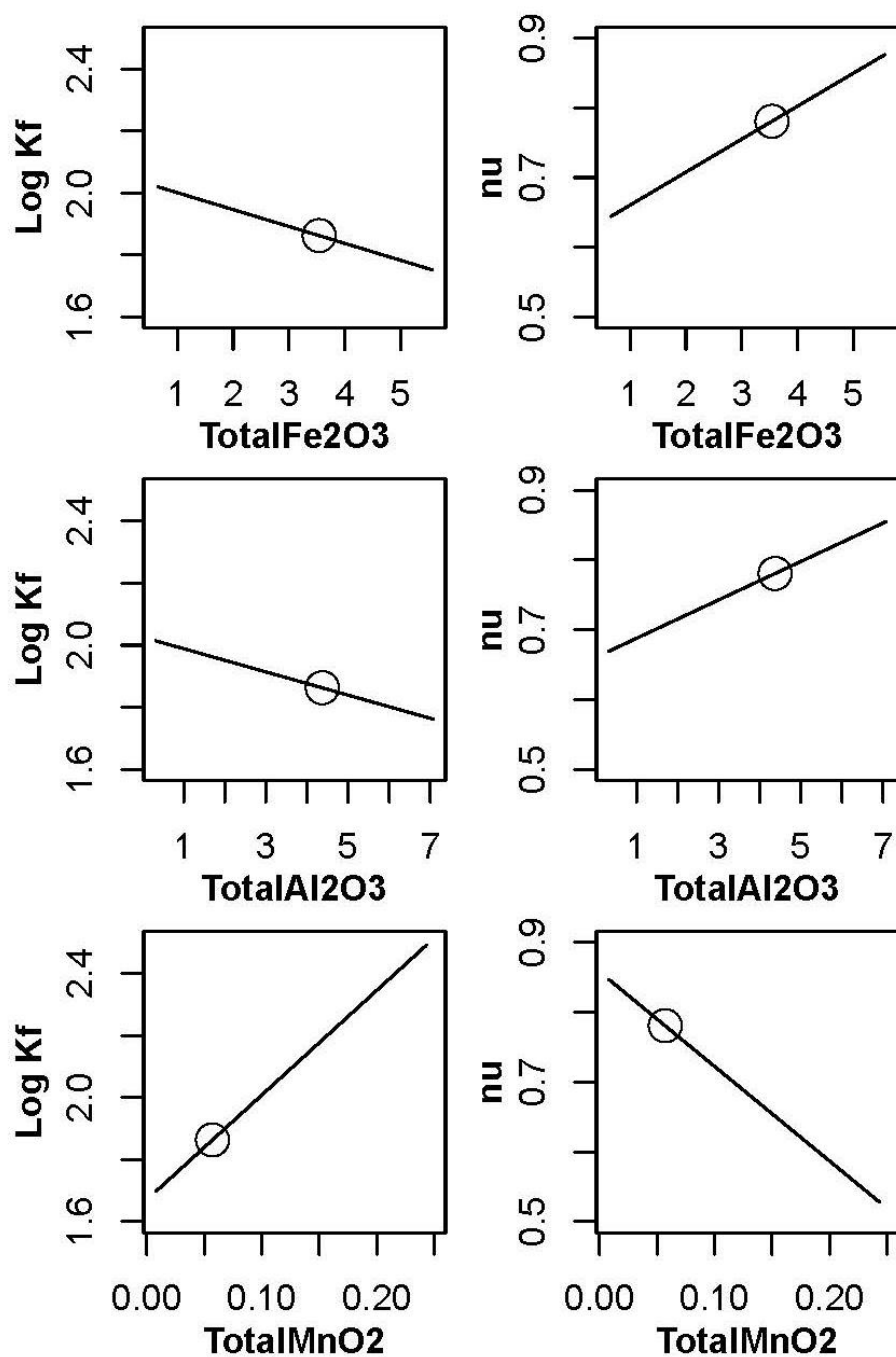
Figure 10. Variation of Freundlich parameters, log K_f and v, as a function of the iron, aluminum, and manganese oxide soil concentration. The parameters not varied are at their median soil concentrations, indicated by the points. See Table 4 for the isotherm parameter equations.

Using the same method employed in Figure 10, the isotherm parameter coefficients were calculated for the minimum, first quartile, median, third quartile, and maximum concentration of each metal oxide, holding the other oxides at their median value. These parameters were used to illustrate the variation of the sorption isotherms as a function of the soil parameter are shown in Figure 11. Increasing the concentration of each soil oxide increases the sorbed concentration. This is as expected since increasing the quantity of sorption sites should increase the sorbed concentration. The log-log plots in Figure 11 shows that there is a threshold of arsenite in the equilibrium concentration before the soil with the highest concentration of iron and aluminum oxide see an increase of sorbed arsenite. This must be the case since continuous increase would be possible only it the isotherms were parallel on the log-log scale.
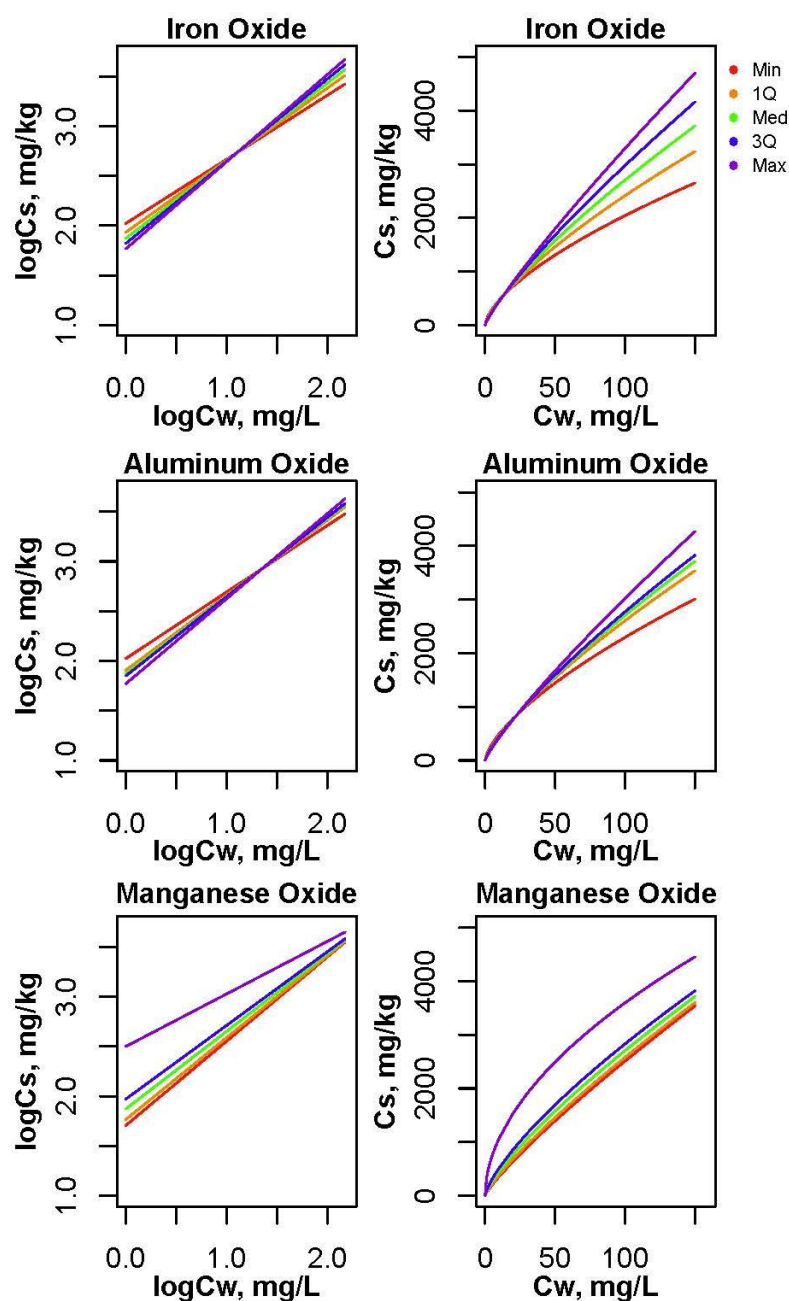
Figure 11. Fitted Freundlich isotherms, linear and log-log, calculated by iron, aluminum, and manganese oxide concentration of each soil. Red indicates the minimum concentration, orange the first quartile, green the median, blue the third quartile and purple the maximum. See Table 4 for the isotherm parameter equations.

Chapter 4

DISCUSSION AND CONCLUSIONS

The traditional method of fitting individual Freundlich isotherm parameters for each soil and comparing the relationship to soil parameters did not expose any relationships between the soil characteristics and the isotherm parameters. By including the soil characteristics directly into the isotherm parameters and fitting the isotherm to the data from all the soils simultaneously resulted estimates that were statistically significant. This method differs from those previously employed by incorporating the soil characteristics directly into the isotherm model rather than identifying relationships between isotherm parameters and soil characteristics.

The importance of iron and aluminum oxides to arsenite sorption has been established by many studies, some of which also include the importance of organic matter. However, including it in the isotherm model did not significantly improve the fit. While most studies do not include manganese oxide as contributing to the sorption of arsenite, they often indicate the oxidation of As^{III} to As^{V} occurs within 48 hours, allowing more than 90% of arsenic sorption to take place within 72 hours (Sparks, 2003). Smith et al. (1999) indicates that iron, in addition to manganese, oxides could participate in the sorption of arsenite through oxidation; however, after conducting recovery tests of the four soils at equilibrium for 24 hours, there is little support that the arsenite oxidation had occurred in that time frame. A different conclusion was reached using desorption, in which arsenate desorbed from soils on which arsenite was originally adsorbed (Dias et al., 2009). The added arsenite was equilibrated with these soils for 48 hours. The importance of manganese oxides in this model could be

attributed to the oxidation of arsenite to arsenate. In addition, there is a strong correlation between organic matter and total manganese oxide content, as seen in Figure 1. This relationship could account for the insignificance between the model containing all three total oxide and organic matter concentrations and the model utilizing only the total mental oxide concentrations. Figure 9 shows little correlation between the amorphous and total oxide concentrations for the soils, except for the case of manganese oxide, explaining the weaker fit for the amorphous oxide model.

The resulting isotherm model predicted the sorption of arsenite on these fifteen New Jersey soils to within approximately a factor of 2. The model that best represented the sorption of arsenite on these New Jersey soils uses the concentration of total iron, aluminum, and manganese oxides.

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Appendix A

ADDITIONAL SOIL CHARACTERISTICS

Table A1. Soil Names

Soil Name	Soil Letter
Birdsboro Silt Loam	A
Boonton Loam (BC)	B
Boonton Loam (UC)	C
Downer Loamy Sand	D
Dunellen Sandy Loam	E
Fill Materials (DE Riv)	F
Freehold Sandy Loam (B Horizon)	G
Freehold Sandy Loam (A Horizon)	H
Hazen Gravelly Loam	I
Lakewood Sand	J
Penn Silt Loam	K
Rockaway Stony Loam	L
Sassafras Sandy Loam	M
Washington Loam	N
Whippany Silty Clay Loam	O

Table A2. Soil Properties as Reported by Lee et al., 1996

Soil	pH in $CaCl_2$	pH zpc	Amorphous and Crystalline Metal Oxide (mg/kg)			Particle Size Distribution (%)			Soil Cation Exchange Capacity (meq/100g)				A_{sp}^2 * (m^2/g)
			Fe	Al	Mn	Sand	Silt	Clay	K	Mg	Ca	ECEC	
A	5.24	3.4	1.6	0.744	0.119	50	32	18	0.47	1.37	3.3	5.3	7.06
B	4.31	3	2.08	0.661	0.076	60	27	13	0.02	0.6	2.59	4.2	7.2
C	4.7	2.8	2.027	2.027	0.195	49	35	16	0.19	0.42	1.58	4.2	6.45
D	3.74	3.4	0.641	0.25	0.014	87	5	8	0.21	0.43	1.1	2.3	1.15
E	4.93	3.2	1.281	0.562	0.057	56	30	14	0.13	1.06	2.71	4.2	5.21
F	4.09	3.2	2.559	0.118	0.077	85	5	10	0.21	0.43	1.1	2.3	2.37
G	4.87	2.9	1.813	0.382	0.053	92	2	6	0.07	0.21	0.46	0.8	2.04
H	5.72	3.6	1.281	0.053	0.017	37	42	21	0.33	0.91	2.84	4.3	9.01
I	5.77	3.1	1.493	0.958	0.189	39	38	23	0.81	1.9	6.53	9.3	5.9
J	3.65	3.6	1.6	0.201	0.016	91	3	6	0.01	0.02	0.05	0.9	1.1
K	4.13	3.3	1.654	0.546	0.142	25	48	27	0.14	0.42	1.25	3.8	8.04
L	4.23	2.8	3.839	1.731	0.179	54	30	16	0.15	0.14	0.59	2.7	8.62
M	5.31	3.4	1.973	0.365	0.032	45	37	18	0.14	0.69	2.06	3.1	5.31
N	5.8	2.85	1.813	0.415	0.236	20	49	31	0.65	1.63	6.59	8.9	11.59
O	5.72	3	1.494	0.184	0.025	49	16	37	0.05	2.52	6.87	9.5	5.98

* BET specific surface area

FREUNDLICH ISOTHERM PARAMTER EQUATIONS AND STATISTICS

Table B1a. Total Iron Oxide, Total Aluminum Oxide, Total Manganese Oxide, and
Organic Matter Isotherm Parameter Coefficients and Statistics

	Coefficient	Std. Error	t value	Pr(>|t|)
b_0	2.024E+00	3.206E-02	6.313E+01	< 2E-16
m_0	4.709E-01	7.508E-02	6.271E+00	4.970E-09
TotFe, b_1	-5.457E-02	1.437E-02	-3.798E+00	2.230E-04
TotAl, b_2	-3.377E-02	1.085E-02	-3.112E+00	2.287E-03
TotMn, b_3	3.466E+00	4.179E-01	8.293E+00	1.250E-13
OC, b_4	-8.300E-04	1.726E-03	-4.780E-01	6.333E-01
TotFe, m_1	6.515E-02	2.592E-02	2.514E+00	1.318E-02
TotAl, m_2	2.714E-02	2.096E-02	1.295E+00	1.977E-01
TotMn, m_3	-2.316E+00	5.010E-01	-4.623E+00	9.060E-06
OC, m_4	5.325E-03	1.919E-03	2.776E+00	6.332E-03

Table B1b. Total Iron Oxide, Total Aluminum Oxide, Total Manganese Oxide, and
Organic Matter Isotherm Parameter Statistics

Residual Std. Error*	Multiple R^2	Adjusted R^2	F-Statistic**	RSME
0.1299	0.9453	0.9415	247.7	0.14

* 129 degrees of freedom ** on 9 and 129 degrees of freedom, p-value < 2.2E-16

Table B2a. Total Iron Oxide, Total Aluminum Oxide, and Total Manganese Oxide
Isotherm Parameter Coefficients and Statistics

	Coefficient	Std. Error	t value	Pr(>|t|)
b_0	2.027E+00	3.099E-02	6.540E+01	< 2E-16
m_0	5.700E-01	6.891E-02	8.272E+00	1.300E-13
TotFe, b_1	-5.223E-02	1.423E-02	-3.669E+00	3.520E-04
TotAl, b_2	-3.742E-02	1.100E-02	-3.401E+00	8.890E-04
TotMn, b_3	3.376E+00	2.773E-01	1.217E+01	< 2E-16
TotFe, m_1	4.732E-02	2.573E-02	1.840E+00	6.810E-02
TotAl, m_2	2.753E-02	2.143E-02	1.285E+00	2.012E-01
TotMn, m_3	-1.356E+00	3.599E-01	-3.767E+00	2.490E-04

Table B2b. Total Iron Oxide, Total Aluminum Oxide, and Total Manganese Oxide
Isotherm Parameter Statistics

Residual Std. Error*	Multiple R^2	Adjusted R^2	F-Statistic**	RSME
0.1332	0.9416	0.9385	301.6	0.143

* 131 degrees of freedom **on 7 and 131 degrees of freedom, p-value < 2.2E-16

Table B3a. Total Manganese Oxide, Total Aluminum Oxide, and Organic Matter
Isotherm Parameter Coefficients and Statistics

	Coefficient	Std. Error	t value	Pr(>|t|)
b_0	2.005E+00	3.158E-02	6.350E+01	< 2E-16
m_0	4.360E-01	7.407E-02	5.886E+00	3.130E-08
TotMn, b_1	-8.727E-02	9.857E-03	-8.854E+00	5.120E-15
TotAl, b_2	3.516E+00	4.237E-01	8.299E+00	1.120E-13
OC, b_3	-1.455E-03	1.763E-03	-8.250E-01	4.106E-01
TotMn, m_1	1.016E-01	1.626E-02	6.248E+00	5.390E-09
TotAl, m_2	-2.322E+00	5.025E-01	-4.620E+00	9.040E-06
OC, m_3	6.159E-03	1.954E-03	3.151E+00	2.010E-03

Table B3b. Total Manganese Oxide, Total Aluminum Oxide, and Organic Matter
Isotherm Parameter Statistics

Residual Std. Error*	Multiple R^2	Adjusted R^2	F-Statistic**	RSME
0.1337	0.9412	0.938	299.4	0.143

* 131 degrees of freedom **on 7 and 131 degrees of freedom, p-value < 2.2E-16

Table B4a. Total Amorphous Oxides Isotherm Parameter Coefficients and Statistics

	Coefficient	Std. Error	t value	Pr(>|t|)
b_0	1.902E+00	3.453E-02	5.510E+01	< 2E-16
m_0	6.627E-01	5.994E-02	1.106E+01	< 2E-16
AmorFe, b_1	-1.973E-01	6.782E-02	-2.909E+00	4.270E-03
AmorAl, b_2	-9.884E-02	9.482E-02	-1.042E+00	2.992E-01
AmorMn, b_3	6.276E+00	1.426E+00	4.401E+00	2.240E-05
OC, b_4	8.671E-03	2.698E-03	3.213E+00	1.660E-03
AmorFe, m_1	1.743E-01	1.080E-01	1.614E+00	1.090E-01
AmorAl, m_2	2.214E-01	1.731E-01	1.279E+00	2.032E-01
AmorMn, m_3	1.902E+00	3.453E-02	5.510E+01	< 2E-16
OC, m_4	6.627E-01	5.994E-02	1.106E+01	< 2E-16

Table B4b. Total Amorphous Oxides Isotherm Parameter Statistics

Residual Std. Error*	Multiple R^2	Adjusted R^2	F-Statistic**	RSME
0.1633	0.9136	0.9076	151.6	0.166

* 129 degrees of freedom **on 9 and 129 degrees of freedom, p-value < 2.2E-16

Appendix C

ARSENITE SORPTION DATA

Table C1. Arsenite Sorption Data, by soil, as Reported by Dias et al., 2009

	logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)
A	-3.764E-01	1.522E+00	B	-3.356E-01	1.453E+00	C	-1.341E+00	1.852E+00
A	-2.866E-01	1.688E+00	B	-2.445E-01	1.622E+00	C	-1.353E+00	1.925E+00
A	-1.656E-01	1.761E+00	B	-1.352E-01	1.713E+00	C	-1.140E+00	1.962E+00
A	-4.167E-02	1.781E+00	B	-8.057E-02	1.823E+00	C	-1.140E+00	2.076E+00
A	8.488E-02	1.902E+00	B	1.016E-01	1.876E+00	C	-8.430E-01	2.133E+00
A	2.536E-01	2.085E+00	B	2.656E-01	2.051E+00	C	-7.153E-01	2.268E+00
A	5.096E-01	2.249E+00	B	5.571E-01	2.156E+00	C	-4.848E-01	2.329E+00
A	7.235E-01	2.355E+00	B	6.543E-01	2.481E+00	C	-3.633E-01	2.415E+00
A	-6.399E-01	1.872E+00	B	9.397E-01	2.806E+00	C	-3.543E-02	2.619E+00
A	-6.530E-01	2.014E+00	B	1.523E+00	3.249E+00	C	2.559E-01	2.766E+00
A	-5.199E-01	2.062E+00	B	1.735E+00	3.353E+00	C	4.612E-01	3.105E+00
A	-3.334E-01	2.176E+00	B	2.069E+00	3.561E+00	C	1.156E+00	3.575E+00
A	-6.706E-02	2.338E+00	B	2.185E+00	3.698E+00	C	1.349E+00	3.729E+00
A	1.262E-01	2.547E+00	B	-5.159E-01	2.071E+00	C	1.669E+00	4.044E+00
A	2.662E-01	2.756E+00	B	-2.976E-01	2.177E+00	C	1.826E+00	4.167E+00
A	9.988E-01	3.317E+00	B	-1.523E-01	2.356E+00			
A	1.179E+00	3.544E+00	B	-1.948E-02	2.601E+00			
A	1.532E+00	3.829E+00	B	1.621E-01	2.788E+00			
A	1.632E+00	3.934E+00	B	5.378E-01	3.057E+00			
A	1.878E+00	4.105E+00	B	9.013E-01	3.351E+00			
			B	1.083E+00	3.579E+00			
			B	1.301E+00	3.750E+00			
			B	1.483E+00	3.848E+00			
			B	1.525E+00	3.970E+00			
			B	1.731E+00	4.003E+00			
			B	1.877E+00	4.084E+00			

	logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)
D	-1.510E-01	1.481E+00	E	-2.931E-01	1.420E+00	F	-4.218E-01	1.569E+00
D	-5.258E-02	1.573E+00	E	-1.251E-01	1.442E+00	F	-3.361E-01	1.728E+00
D	3.183E-02	1.632E+00	E	-4.773E-02	1.568E+00	F	-1.882E-01	1.778E+00
D	1.527E-01	1.766E+00	E	5.568E-02	1.583E+00	F	-5.057E-02	1.793E+00
D	2.875E-01	2.033E+00	E	1.717E-01	1.745E+00	F	4.893E-02	1.949E+00
D	5.690E-01	2.113E+00	E	3.720E-01	1.851E+00	F	1.758E-01	2.182E+00
D	7.490E-01	2.300E+00	E	5.588E-01	2.174E+00	F	4.370E-01	2.359E+00
D	-7.465E-01	1.758E+00	E	7.331E-01	2.323E+00	F	6.673E-01	2.466E+00
D	-6.798E-01	1.891E+00	E	9.974E-01	2.731E+00	F	-9.833E-01	1.808E+00
D	-5.400E-01	1.977E+00	E	1.566E+00	3.147E+00	F	-7.888E-01	1.908E+00
D	-5.198E-01	2.090E+00	E	1.772E+00	3.240E+00	F	-7.037E-01	2.015E+00
D	-2.935E-01	2.176E+00	E	2.218E+00	3.586E+00	F	-6.187E-01	2.101E+00
D	-2.067E-01	2.385E+00	E	1.860E-01	2.538E+00	F	-3.694E-01	2.201E+00
D	3.261E-01	2.746E+00	E	1.997E-01	2.793E+00	F	-2.176E-01	2.380E+00
D	6.990E-01	3.032E+00	E	7.057E-01	3.022E+00	F	6.809E-02	2.587E+00
D	1.265E+00	3.497E+00	E	1.265E+00	3.535E+00	F	3.173E-01	2.744E+00
D	1.432E+00	3.706E+00	E	1.438E+00	3.678E+00	F	3.653E-01	3.113E+00
D	1.791E+00	3.972E+00	E	1.745E+00	4.019E+00	F	1.064E+00	3.606E+00
D	1.971E+00	4.039E+00	E	1.858E+00	4.133E+00	F	1.301E+00	3.764E+00

	logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)
G	-4.467E-01	1.598E+00	H	-4.283E-01	1.574E+00	I	-4.234E-01	1.565E+00
G	-2.963E-01	1.700E+00	H	-3.514E-01	1.742E+00	I	-3.498E-01	1.743E+00
G	-1.829E-01	1.785E+00	H	-2.406E-01	1.832E+00	I	-1.849E-01	1.772E+00
G	-1.037E-01	1.857E+00	H	-1.115E-01	1.869E+00	I	-5.784E-02	1.794E+00
G	5.456E-02	1.947E+00	H	3.883E-02	1.962E+00	I	5.751E-02	1.936E+00
G	1.974E-01	2.166E+00	H	2.131E-01	2.143E+00	I	2.658E-01	2.076E+00
G	4.272E-01	2.383E+00	H	4.559E-01	2.338E+00	I	4.660E-01	2.320E+00
G	6.774E-01	2.465E+00	H	6.669E-01	2.471E+00	I	6.990E-01	2.406E+00
G	-1.269E+00	1.982E+00	H	-9.575E-01	1.804E+00	I	-6.119E-01	1.688E+00
G	-1.150E+00	2.073E+00	H	-6.790E-01	1.891E+00	I	-6.245E-01	1.867E+00
G	-1.105E+00	1.826E+00	H	-5.785E-01	1.990E+00	I	-4.912E-01	1.965E+00
G	-8.737E-01	2.139E+00	H	-5.170E-01	2.074E+00	I	-4.125E-01	2.046E+00
G	-5.974E-01	2.251E+00	H	-2.968E-01	2.168E+00	I	-2.550E-01	2.152E+00
G	-3.277E-01	2.414E+00	H	-1.381E-01	2.351E+00	I	-6.730E-02	2.348E+00
G	-1.405E-02	2.610E+00	H	1.761E-01	2.537E+00	I	2.176E-01	2.527E+00
G	2.180E-01	2.773E+00	H	2.927E-01	2.737E+00	I	3.505E-01	2.739E+00
G	5.123E-01	3.080E+00				I	6.898E-01	3.000E+00
G	9.188E-01	3.640E+00				I	1.125E+00	3.563E+00
G	1.226E+00	3.778E+00				I	1.337E+00	3.717E+00
						I	1.756E+00	3.995E+00
						I	1.895E+00	4.092E+00

	logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)
J	-6.176E-01	1.692E+00	K	-3.450E-01	1.496E+00	L	-4.389E-01	1.582E+00
J	-5.844E-01	1.798E+00	K	-2.548E-01	1.651E+00	L	-2.688E-01	1.667E+00
J	-4.952E-01	1.817E+00	K	-1.321E-01	1.722E+00	L	-1.534E-01	1.739E+00
J	-3.897E-01	1.919E+00	K	-4.167E-02	1.785E+00	L	-5.627E-02	1.785E+00
J	-3.010E-01	1.986E+00	K	1.134E-01	1.856E+00	L	-1.413E-01	2.621E+00
J	-1.370E-01	2.087E+00	K	2.811E-01	2.047E+00	L	1.138E-01	2.220E+00
J	-1.215E-01	2.240E+00	K	5.459E-01	2.174E+00	L	6.968E-01	2.403E+00
J	1.266E-01	2.209E+00	K	6.164E-01	2.525E+00	L	8.972E-01	2.858E+00
J	4.944E-01	2.246E+00	K	9.455E-01	2.807E+00	L	1.498E+00	3.255E+00
J	7.385E-01	2.269E+00	K	1.566E+00	3.140E+00	L	1.717E+00	3.392E+00
J	-8.636E-01	1.783E+00	K	2.089E+00	3.459E+00	L	2.106E+00	3.399E+00
J	-7.016E-01	1.845E+00	K	2.218E+00	3.586E+00	L	2.215E+00	3.588E+00
J	-6.919E-01	1.902E+00	K	-1.225E+00	1.832E+00	L	-1.203E+00	2.154E+00
J	-5.364E-01	1.979E+00	K	-8.666E-01	1.944E+00	L	-1.002E+00	2.275E+00
J	-4.198E-01	2.042E+00	K	-7.601E-01	2.035E+00	L	-5.950E-01	2.441E+00
J	-2.773E-01	2.154E+00	K	-7.101E-01	2.120E+00	L	-7.195E-02	2.628E+00
J	-1.899E-01	2.256E+00	K	-4.841E-01	2.225E+00	L	1.899E-02	2.827E+00
J	-8.947E-02	2.337E+00	K	-2.459E-01	2.401E+00	L	5.480E-01	3.086E+00
J	2.215E-01	2.523E+00	K	-7.730E-03	2.603E+00	L	1.150E+00	3.571E+00
J	4.158E-01	2.688E+00	K	2.746E-01	2.754E+00	L	1.350E+00	3.743E+00
			K	4.808E-01	3.093E+00			
			K	1.177E+00	3.556E+00			
			K	1.434E+00	3.694E+00			

	logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)		logCw (mg/L)	logCs (mg/kg)
M	-2.976E-01	1.393E+00	N	-5.011E-01	1.640E+00	O	-3.782E-01	1.515E+00
M	-2.740E-01	1.665E+00	N	-3.568E-01	1.750E+00	O	-2.686E-01	1.669E+00
M	-1.364E-01	1.715E+00	N	-3.020E-01	1.880E+00	O	-1.955E-01	1.799E+00
M	-5.748E-02	1.807E+00	N	-1.747E-01	1.934E+00	O	-1.044E-01	1.856E+00
M	6.963E-02	1.920E+00	N	-2.694E-02	2.026E+00	O	5.953E-02	1.954E+00
M	2.620E-01	2.076E+00	N	1.448E-01	2.210E+00	O	2.358E-01	2.109E+00
M	5.648E-01	2.134E+00	N	3.818E-01	2.423E+00	O	4.788E-01	2.304E+00
M	7.157E-01	2.374E+00	N	6.777E-01	2.445E+00	O	6.550E-01	2.479E+00
M	-7.504E-01	1.859E+00	N	-7.445E-01	1.750E+00	O	-8.250E-01	1.776E+00
M	-7.320E-01	1.998E+00	N	-7.319E-01	2.035E+00	O	-7.769E-01	1.921E+00
M	-5.011E-01	2.050E+00	N	-7.018E-01	1.889E+00	O	-6.803E-01	2.013E+00
M	-3.248E-01	2.197E+00	N	-5.011E-01	2.080E+00	O	-5.412E-01	2.075E+00
M	1.556E-01	2.550E+00	N	-3.248E-01	2.197E+00	O	-3.721E-01	2.199E+00
M	3.624E-01	2.756E+00	N	-1.605E-01	2.366E+00	O	-2.938E-01	2.397E+00
M	5.208E-01	3.020E+00	N	1.556E-01	2.550E+00	O	2.647E-02	2.606E+00
M	1.117E+00	3.541E+00	N	3.624E-01	2.719E+00	O	2.984E-01	2.745E+00
M	1.317E+00	3.725E+00	N	5.269E-01	3.071E+00	O	5.217E-01	3.082E+00
M	1.798E+00	4.004E+00	N	1.123E+00	3.592E+00	O	1.192E+00	3.567E+00
			N	1.324E+00	3.754E+00	O	1.392E+00	3.721E+00
			N	1.798E+00	3.960E+00	O	1.827E+00	3.953E+00