Predicting Soil—Water Partition Coefficients for Cadmium

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Equilibrium batch measurements were made of the adsorption of Cd(II) on 15 New Jersey soils. The soils were characterized in terms of surface properties and chemical composition, particularly organic matter and metal oxides. The adsorption of Cd(II) followed the Langmuir relation and increased with increasing pH between pH 3 and pH 10. At constant pH and metal concentration, greater adsorption was observed for soils with higher organic matter content. To better understand the mechanism of adsorption, the experimental results for the adsorption of Cd by the 15 soils were tested using a partition coefficient model that related the adsorption of the Cd(II) to the soil components: organic matter, iron oxides, aluminum oxides, and manganese oxides. This model was not successful when applied to measurements at the natural soil pH because of the competition of protons with Cd(II) for available sites. However, at constant pH, partition coefficients obtained from experimental data were highly correlated with those calculated for a partition coefficient between Cd(II) and organic matter alone. Normalization of the partition coefficients, \( K_{om} \), for the organic matter content of the soils, greatly improved the relationship between the partition coefficient and pH (\( R^2 \) increased from 0.799 to 0.927). This suggests that the surficial adsorption sites are principally composed of organic matter. For the 24-h equilibration period employed, diffusion of Cd(II) through this superficial organic matter coating to underlying sorptive materials, including metal oxides, is unimportant in the partitioning of Cd(II).

Introduction

The mobility and fate of metals in the soil environment are directly related to their partitioning between soil and soil solution (1). The presence of trace metals in the solid phase is a result of precipitation and adsorption to components of the soil, processes that are highly pH dependent. For trace metals, adsorption is normally of greater importance than is precipitation. Varying contents of organic matter and metal oxides may result in varying capacity for metal sorption onto soils.

The interaction of metal ions with natural soil particles is complex, involving multiple mechanisms. Much of the work relating to trace metal sorption by soils in natural systems has involved the use of well-defined “model” surfaces such as clay, aluminum oxides, iron oxides, and manganese oxides. These substances are present in soils, and including them is necessary to develop an understanding of mechanisms for the adsorption of metals.

Boekhold et al. (2), Boekhold and Van der Zee (3), and Van der Zee and Van Reimsdijk (4) have used equations in which the Cd(II) sorption by soil is proportional to the hydrogen ion concentration raised to the \(-0.5\) power. Anderson and Christensen (5) studied 38 soils with samples being taken at several depths. For their 117 sorption measurements made at very low Cd(II) concentrations, from 0.7 to 12.6 \( \mu \)g/L, regression of log of the Cd(II) sorption on the pH gave a slope of 0.64. The regression had an \( R^2 \) value of 0.776, indicating that the proton concentration is the principal factor affecting the partitioning process. More recently, Boekhold et al. (6) have found an exponent for the hydrogen ion concentration of \(-0.69\) for sodium electrolytes and \(-0.77\) for calcium electrolytes, values considerably more negative than \(-0.5\). Apparently, the value is a function of the type of soil considered. A different approach has been taken by Schulte and Beese (7), who showed that, over specified pH intervals, the sorption of Cd(II) onto different soils could be related to their specific surface areas. Both Luoma and Davis (8) and Jenne et al. (9) have considered the modeling of metal sorption by incorporating the contributions of different types of binding phases, such as metal oxides and organic matter.

In this study, we investigated the sorption of Cd(II) on 15 soils collected in New Jersey. We conducted both adsorption isotherm and adsorption edge measurements on the soils. We developed a relationship to describe the sorption of Cd(II) by the different soils that predicts sorption by other soils. We considered the influence of solution pH, of the concentrations of iron, manganese, and aluminum oxides, and of organic matter in the soil on Cd sorption.

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Materials and Methods
The 15 New Jersey soil samples are listed in Table 1. They are representative of the major soil types present in New Jersey.

The soils were air dried, and agglomerates were broken by hand and by using a wooden mallet. Particles larger than 2 mm were removed by sieving. All further tests were performed on the less than 2-mm size fraction of the soils. For the soil analyses presented in Table 1, organic matter was measured by the Walkley–Black wet combustion method; soil pH was determined in water; and soil particle size distribution was determined by a sedimentation (hydrometer) method (10). Soil metal oxides were determined by the ammonium oxalate extraction method of Iyengar et al. (11), using a Perkin Elmer Model 5000 atomic absorption spectrophotometer to analyze the solubilized metal. The natural Cd concentrations of the soils were negligible.

Samples for the determination of adsorption isotherms were prepared by adding 1.0 g of soil to 100 mL of 0.01 M NaNO₃. The pH was adjusted by the addition of small volumes of HNO₃ or NaOH, as necessary, to attain the desired pHs of 4.0, 5.5, and 7.0. The initial Cd(II) concentrations ranged from 1.0 × 10⁻⁶ to 5.0 × 10⁻³ M, and the equilibration time was 24 h at room temperature (25 ± 2 °C). Preliminary testing of Sassafras sandy loam and Washington loam at pH 4 and pH 7 established the adequacy of a 6- to 12-h equilibration time for these soils. After 24 h, the pH was readjusted to the initial value, and the samples were filtered through 0.45-µm membrane filters. The Cd(II) concentrations, [Cd]s, at each pH, for the New Jersey soils varied widely, from 3.419 to 6469 µg/g; [Cd]w is the cadmium concentration in water (µg/L); Γₚ is the maximum sorbed cadmium concentration (µg/g); and the highest concentration of organic matter, 8.6%, while the Freehold sandy loam (A horizon) had the smallest concentration of organic matter, 0.2%.

Adsorption edge measurements were conducted for Cd(II) for each of the 15 soils. The soil suspensions were 1.00 ± 0.01 g/100 mL. Ionic strength was maintained at 0.01 M with NaNO₃. We added Cd(NO₃)₂ at a concentration of 1.0 × 10⁻⁴ M. Fifteen pH values covering the range from 3 to 10 were employed. The pH values of the samples were adjusted by adding NaOH or HNO₃ as required. Samples were shaken at 150 strokes/min on a reciprocating shaker (Lab-Line Instruments, Melrose Park, IL) for 24 h at room temperature (25 ± 2 °C). The pH values were determined again after the 24 h of shaking. The samples were then filtered through 25-mm diameter, 0.45-µm membrane filters (Gelman Sciences, Ann Arbor, MI). The Cd(II) concentration in the filtered solution was determined by atomic absorption spectrophotometry.

Results and Discussion
Soil Characteristics. A summary of soil characteristics is presented in Table 1. The soils ranged from sand to silty clay loam; the range in clay content is 6 to 37%. Soil pH ranged from 4.18 to 6.44. Organic matter ranged from 0.2 to 8.6%. The Boonton Union County soil had the highest concentration of aluminum and manganese oxides, whereas the Washington loam had the highest iron oxide content among the soils.

Adsorption Isotherms. The adsorption data obtained from isotherm experiments were fitted by nonlinear regression (12) to Langmuir adsorption isotherms:

$$[\text{Cd}]_w = \frac{\Gamma_m \cdot [\text{Cd}]_w \times 1000}{1 + K \cdot [\text{Cd}]_w \times 1000}$$

where [Cd]w is the cadmium concentration in soil (µg/g); [Cd]w is the cadmium concentration in water (µg/L); Γₚ is the maximum sorbed cadmium concentration (µg/g); and K is the constant (L/mg).

Table 2 presents the values of the Langmuir Γₚ and K parameters from the nonlinear regression of [Cd]w vs [Cd]w. At a given pH, for example pH 7.0, the maximum adsorption concentrations, Γₚ, for the New Jersey soils varied widely, from 714 to 6469 µg/g, but correlated with the soil organic matter content (R² = 0.812). At pH 7, the Boonton Union County soil had the greatest Γₚ, 6469 µg/g, and the highest concentration of organic matter, 8.6%, while the Freehold sandy loam (A horizon) had the smallest Γₚ, 714 µg/g, and the smallest concentration of organic matter, 0.2%. There was a much weaker relationship between Γₚ and soil iron, aluminum, or manganese oxides (R² = 0.229, 0.599, and 0.564, respectively at pH 7). Results for the other pH values were similar in that sorption correlated to organic matter much better than it did to the metal oxides.
Adsorption Edge. The most important factor controlling the partitioning of a metal to soil is the solution pH \((pH)\). In general, the percentage of a cationic metal adsorbed increases as pH increases with a transition range of 2–3 pH units in which the percentage of the metal that is adsorbed increases from nearly 0 to almost 100%. This transition range does not occur at the same value of pH for different soils. In this study, the extremes were represented by Boonton Union County soil and Freehold sandy loam (A horizon), which reach nearly 100% Cd(II) adsorption at pH 6 and 9, respectively (Figure 1). The high dependence of Cd(II) adsorption on pH is explained by the fact that hydrogen ions affect the surface charge of the adsorbent and the degree of ionization and the speciation of the adsorbate \((14)\).

Partition Coefficients. The ratio of the Cd concentration in the soil, \([Cd]_s\), to the concentration of Cd(II) in the solution phase, \([Cd]_w\), may be described by a partition coefficient \((K_d, mL/g)\), which is a function of pH, metal concentration, and other properties of the solution and the solid matrix:

\[
K_d = \frac{[Cd]_s}{[Cd]_w} \quad (2)
\]

We transformed the partition data from the adsorption edges for the 15 soils into \(K_d\) values using the added and equilibrium concentrations of Cd(II). The Cd in the soil was assumed to be solely to the adsorption of cadmium from the solution. The \(K_d\) values varied greatly from soil to soil. Figure 2 shows the large \(K_d\) data set of Anderson and Christensen \((5)\) for Danish agricultural soils and that from our laboratory for 15 New Jersey soils at their natural pH. Although our solution concentrations (97.8–11124 \(\mu g/L\)) are much higher than those from Anderson and Christensen (0.7–12.6 \(\mu g/L\)), our log \(K_d\) values are still within the envelope of those of Anderson and Christensen. The slope of the regression for our soils (0.804) is about one-third higher than the value of 0.603 reported by Anderson and Christensen. Figure 3 shows all the adsorption edge data, transformed into \(K_d\) values, plotted versus pH for all 15 of our soils. Although it is clear from Figures 2 and 3 that pH is the major factor controlling partitioning, the variation in \(K_d\) is still substantial. This indicates that although pH is the most important parameter, it is not the sole one.

A portion of the removal of cadmium may have been a result of precipitation of CdCO\(_3\), but we believe that this is not likely as the media was NaNO\(_3\), and these soils contained little inorganic carbon. We evaluated whether

### Table 2

Parameters for Nonlinear Regression of \(C_s vs C_w\) of Langmuir Equation for Adsorption of Cadmium on 15 New Jersey Soils at Three pH Values

<table>
<thead>
<tr>
<th>Soil Type</th>
<th>(\Gamma_m) ((\mu g/g))</th>
<th>(K) (L/mg)</th>
<th>(R^2)</th>
<th>(\Gamma_m) ((\mu g/g))</th>
<th>(K) (L/mg)</th>
<th>(R^2)</th>
<th>(\Gamma_m) ((\mu g/g))</th>
<th>(K) (L/mg)</th>
<th>(R^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Birdsboro silt loam</td>
<td>1123</td>
<td>0.016</td>
<td>0.999</td>
<td>4113</td>
<td>0.027</td>
<td>0.998</td>
<td>3300</td>
<td>0.310</td>
<td>0.996</td>
</tr>
<tr>
<td>Boonton Bergen County</td>
<td>930</td>
<td>0.043</td>
<td>0.995</td>
<td>2718</td>
<td>0.121</td>
<td>0.983</td>
<td>3977</td>
<td>0.357</td>
<td>0.956</td>
</tr>
<tr>
<td>Boonton Union County</td>
<td>1864</td>
<td>0.037</td>
<td>0.996</td>
<td>6285</td>
<td>0.049</td>
<td>0.994</td>
<td>6469</td>
<td>0.847</td>
<td>0.998</td>
</tr>
<tr>
<td>Downer loamy sand</td>
<td>2644</td>
<td>0.002</td>
<td>0.996</td>
<td>1208</td>
<td>0.075</td>
<td>0.996</td>
<td>2580</td>
<td>0.120</td>
<td>0.997</td>
</tr>
<tr>
<td>Dunellen sandy loam</td>
<td>1123</td>
<td>0.017</td>
<td>0.998</td>
<td>3021</td>
<td>0.040</td>
<td>0.995</td>
<td>2897</td>
<td>0.245</td>
<td>0.998</td>
</tr>
<tr>
<td>Delaware River fill material</td>
<td>886</td>
<td>0.014</td>
<td>0.997</td>
<td>1987</td>
<td>0.054</td>
<td>0.997</td>
<td>2161</td>
<td>0.210</td>
<td>0.999</td>
</tr>
<tr>
<td>Freehold sandy loam (A horizon)</td>
<td>293</td>
<td>0.019</td>
<td>0.999</td>
<td>740</td>
<td>0.030</td>
<td>0.997</td>
<td>714</td>
<td>0.171</td>
<td>0.995</td>
</tr>
<tr>
<td>Freehold sandy loam (B horizon)</td>
<td>942</td>
<td>0.004</td>
<td>0.996</td>
<td>2286</td>
<td>0.115</td>
<td>0.991</td>
<td>3170</td>
<td>0.277</td>
<td>0.981</td>
</tr>
<tr>
<td>Hazen gravelly loam</td>
<td>1973</td>
<td>0.014</td>
<td>0.995</td>
<td>5110</td>
<td>0.050</td>
<td>0.997</td>
<td>4333</td>
<td>0.602</td>
<td>0.980</td>
</tr>
<tr>
<td>Lakewood sand</td>
<td>161</td>
<td>0.034</td>
<td>0.985</td>
<td>323</td>
<td>0.215</td>
<td>0.999</td>
<td>826</td>
<td>0.378</td>
<td>0.998</td>
</tr>
<tr>
<td>Penn silt loam</td>
<td>154</td>
<td>0.175</td>
<td>0.994</td>
<td>1174</td>
<td>0.095</td>
<td>0.996</td>
<td>1524</td>
<td>0.549</td>
<td>0.998</td>
</tr>
<tr>
<td>Rockaway stony loam</td>
<td>1180</td>
<td>0.012</td>
<td>0.997</td>
<td>3370</td>
<td>0.048</td>
<td>0.995</td>
<td>3939</td>
<td>0.210</td>
<td>0.970</td>
</tr>
<tr>
<td>Sassafras sandy loam</td>
<td>116</td>
<td>0.289</td>
<td>0.983</td>
<td>984</td>
<td>0.081</td>
<td>0.997</td>
<td>1124</td>
<td>0.297</td>
<td>0.998</td>
</tr>
<tr>
<td>Washington loam</td>
<td>826</td>
<td>0.119</td>
<td>0.997</td>
<td>3038</td>
<td>0.075</td>
<td>0.998</td>
<td>2050</td>
<td>1.550</td>
<td>0.985</td>
</tr>
<tr>
<td>Whippany silty clay loam</td>
<td>1273</td>
<td>0.075</td>
<td>0.997</td>
<td>1418</td>
<td>0.358</td>
<td>0.998</td>
<td>1968</td>
<td>0.821</td>
<td>0.996</td>
</tr>
</tbody>
</table>
The total quantity of the Cd sorbed by the soil, \( [\text{Cd}]_{\text{tot}} \), is the sum of the quantities of the Cd sorbed by the individual sorption phases:

\[
[\text{Cd}]_{\text{tot}} = [\text{Cd}]_{\text{org}} + [\text{Cd}]_{\text{Al}} + [\text{Cd}]_{\text{Mn}}
\]

The equilibrium Cd(II) concentration present in solution then can be obtained by consideration of the binding of Cd(II) by all binding sites:

\[
\frac{[\text{Cd}]}{[\text{Cd}]_{\text{tot}}} = \frac{1}{\sum_{\text{s}} K_{\text{s}-\text{Cd}}[S_{\text{s}}]} = \frac{1}{K_d}
\]

Therefore, the partition coefficient is

\[
K_d = \sum_{\text{s}} K_{\text{s}-\text{Cd}}[S_{\text{s}}]
\]
TABLE 4
Comparison of $R^2$ for Regression of Experimental Partition Coefficients against Predicted Values Obtained from Conditional Stability Constants

<table>
<thead>
<tr>
<th>soil component considered</th>
<th>natural pH</th>
<th>pH 4.0</th>
<th>pH 5.0</th>
<th>pH 6.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>OM$^a$</td>
<td>0.104</td>
<td>0.928</td>
<td>0.958</td>
<td>0.966</td>
</tr>
<tr>
<td>Fe</td>
<td>0.087</td>
<td>0.275</td>
<td>0.357</td>
<td>0.263</td>
</tr>
<tr>
<td>Al</td>
<td>0.040</td>
<td>0.722</td>
<td>0.789</td>
<td>0.720</td>
</tr>
<tr>
<td>Mn</td>
<td>0.105</td>
<td>0.705</td>
<td>0.684</td>
<td>0.593</td>
</tr>
<tr>
<td>OM and Al</td>
<td>0.149</td>
<td>0.931</td>
<td>0.968</td>
<td>0.973</td>
</tr>
<tr>
<td>OM, Al, and Mn</td>
<td>0.172</td>
<td>0.958</td>
<td>0.973</td>
<td>0.975</td>
</tr>
</tbody>
</table>

$^a$The regressions were performed at three different pH values and at the natural pH of each of the soils by considering one, two, and three soil components in the model (eq 8). $^b$OM, organic matter content.

![Figure 4](image_url)

**FIGURE 4.** Correlation between experimental and predicted partition coefficient by considering organic matter only at three pH values. Line of equality is shown. $R^2 = 0.982$; $1 \times 10^{-4}$ M cadmium; soil/water = 1 g/100 mL; $I = 0.01$ M NaNO$_3$; $T = 25 \pm 2^\circ$C.

![Figure 5](image_url)

**FIGURE 5.** Log $K_{om}$ as a function of pH for adsorption of cadmium onto 15 New Jersey soils. Soil/water = 1 g/100 mL; $I = 0.01$ M NaNO$_3$; $T = 25 \pm 2^\circ$C. Regression equation: log $K_{om} = 1.084 + 0.477$ pH; $R^2 = 0.927$.

**TABLE 5**
Regression Coefficients for Log $K_d$ vs pH (Figure 3) and Log $K_{om}$ vs pH (Figure 5)

<table>
<thead>
<tr>
<th>regression</th>
<th>$n$</th>
<th>slope</th>
<th>intercept</th>
<th>$R^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>log $K_d$ vs pH</td>
<td>194$^b$</td>
<td>0.455</td>
<td>-0.552</td>
<td>0.799</td>
</tr>
<tr>
<td>log $K_{om}$ vs pH</td>
<td>194$^b$</td>
<td>0.477</td>
<td>1.084</td>
<td>0.927</td>
</tr>
<tr>
<td>log $K_d$ vs pH</td>
<td>136$^c$</td>
<td>0.478</td>
<td>-0.582</td>
<td>0.858</td>
</tr>
<tr>
<td>log $K_{om}$ vs pH</td>
<td>136$^c$</td>
<td>0.491</td>
<td>0.996</td>
<td>0.928</td>
</tr>
<tr>
<td>log $K_d$ vs pH</td>
<td>146$^d$</td>
<td>0.488</td>
<td>-0.666</td>
<td>0.698</td>
</tr>
<tr>
<td>log $K_{om}$ vs pH</td>
<td>146$^d$</td>
<td>0.522</td>
<td>0.892</td>
<td>0.951</td>
</tr>
</tbody>
</table>

$^a$Number of samples. $^b$All data sets. $^c$Samples with organic matter content less than 0.5% have not been included. $^d$Samples with pH less than 3.5 and greater than 8.5 have not been included.

...soil components in the model (eq 8). at the natural pH of each of the soils by considering one, two, and three organic matter to the binding of Cd(II). experimentally obtain. Figure 4 shows the importance of the form of required additional information that is difficult to gains only a marginal improvement at great cost in the of Cd(II) by the soils. Adding a second phase to the model considered, iron oxide was the least important to the binding of Cd(II) by the soils. Furthermore, consistent with eq 8, we have forced our regressions through the origin. This implies that in the absence of the sorption phases considered, there will be no sorption of Cd(II).

To reduce the variance in the pH-dependent $K_d$'s that are shown in Figure 3, the $K_d$ values were normalized to the organic matter content of the soils. Normalization of $K_d$ to the amount of organic carbon is commonly done for hydrophobic organic compounds (15, 16). The organic matter normalized partition coefficients, $K_{om}$, for the 15 soils are plotted versus pH for the 15 soils in Figure 5. The data collapse to form a single curve in which the data scatter is far less than was present in Figure 3. The regression parameters for Figures 3 and 5 are given in Table 5. When $K_{om}$ rather than $K_d$ is considered, the $R^2$ value increases from 0.799 to 0.927. Table 5 also shows the effect of truncating the data set. If soils have low concentrations of organic matter, the contribution of other binding sites, such as metal oxides and ion exchange sites on clay minerals, might be expected to become more important in the partitioning of Cd(II). When only samples with an organic matter content greater than 0.5% were included in the regressions, the $R^2$ value for the $K_d$ vs pH regression increased from 0.799 to 0.858. The $R^2$ values for the regressions of $K_{om}$ on pH were almost the same. The regression in Figure 5 deviates at both low and high pH. We have recently demonstrated that significant concentrations of organic matter dissolve from these soils at high pH (17). These organics would be expected to form soluble com-
plexes with Cd that would decrease the measured partition coefficient. Furthermore, in the \( K_{om} \) values we have not applied any correction for the solubilization of organic matter at higher pH.

**Application to Field Soils.** In our 24-h adsorption experiments, it was found that primarily the organic matter and not the metal oxides were involved in the sorption of the added Cd(II). This conclusion regarding the partitioning of Cd(II) in our work is in contradiction to that obtained from sequential fractionation data which show a distribution of Cd among various sorption phases (see, for example, ref 18 and references cited therein). The fractionations are of soils that are field contaminated and therefore aged. We believe that the results of our experiments demonstrate that organic matter is the surficial coating of the soil and, in the 24-h period that we used to equilibrate the samples, diffusion of the Cd to underlying metal oxide and other sorption phases was not important in controlling the partitioning of the Cd(II) between the soil and the solution phases. In a like manner, the desorption will be controlled by the equilibrium of Cd between the soil organic matter and soil water. The underlying oxide materials serve as a reservoir of Cd but do not affect the soil water concentration of cadmium. The uptake and release of Cd by these underlying metal oxide and other sorptive phases appears to be a diffusion controlled process.

We have applied the model for Cd(II) partitioning that is shown in Figure 5 to data for 13 soils that were collected in The Netherlands (see Table 6) (19). The reported \( K_d \) values were computed from Cd released from the soil by nitric acid digestion and from measurements of \( K_{om} \) in pore water. We predicted the log \( K_{om} \) using the equation presented in Figure 5. All predicted \( K_{om} \) values were within one order of magnitude of the measured \( K_{om} \) values. The mean deviation of log \( K_{om} \) was 0.241.

**Summary and Conclusions**

The results of both adsorption edge and adsorption isotherm studies showed that Cd(II) adsorption is highly pH dependent. The different soils have very different adsorption abilities. Among all soil properties, the organic matter plays the most important role in controlling Cd(II) sorption by soils.

The adsorption coefficient (\( K_d \)) increases with increasing pH values. We have developed a model for the sorption of metal by soil surface substances at fixed pH. The results show that the conditional partition coefficients were highly correlated to the organic matter content at a fixed pH. The \( R^2 \) of the regression line was improved from 0.799 to 0.927 when log \( K_{om} \) vs pH rather than log \( K_d \) vs pH was considered for the 194 measurements for the 15 soils that we investigated. We tested the model using data for soils from The Netherlands and were able to predict the partition coefficient within 1 order of magnitude for soils to which Cd(II) had not been added in the laboratory.

For the 24-h equilibration period employed, diffusion of Cd through the superficial organic matter coating to underlying sorptive materials, including metal oxides, appears to be unimportant in the partitioning of Cd(II).

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**Literature Cited**


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