



## DEVELOPMENT OF SOIL METAL CRITERIA TO PRESERVE GROUNDWATER QUALITY

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### ABSTRACT

The principal impediment in the remediation of contaminated sites and in the protection of groundwater quality is the lack of appropriate and reasonable standards for heavy metals in soils. There are no standards applicable to predict the potential for groundwater contamination by heavy metals in Taiwan. Lack of these soil standards may result in subjective judgment regarding the remediation needed. The migration of heavy metals through the unsaturated zone to groundwater is controlled by sorption to the soil, a highly pH-dependent process, and the hydrological regime. Soil sorption behavior is the criterion upon which to establish a standard based on a maximum permissible concentration in groundwater. The maximum level of metal in soil for which the equilibrium soluble metal does not exceed the Drinking Water Standard can be computed, at any pH, from the measured adsorption coefficient for any metal and soil. These metal criteria can be used as soil standards that will be protective of groundwater quality. Criteria for soil remediation are based on specific soil types and the effect of pH on metal sorption because the partitioning of trace metals is highly dependent on the solution pH and the chemical nature of the soil. © 1998 IAWQ. Published by Elsevier Science Ltd. All rights reserved

### KEYWORDS

Adsorption coefficient; groundwater; heavy metal; soil; soil standard.

### INTRODUCTION

The rapid increase in the disposal of municipal and industrial waste in landfill sites and the application of sewage sludge to land, has resulted in significant pollution of the environment. There are many routes for human exposure to contaminants in soil. A major route for exposure is via groundwater. The lack of suitable heavy metals soil standards has hampered the design of soil clean-up strategies in site remediation. The partitioning of metals between soil and soil solution can be directly utilized to evaluate the environmental impact and the fate of metals in the environment. Metal adsorption by soil is highly pH dependent. (Kuo and Baker, 1980; Harter, 1983; Elliott *et al.*, 1986). Christensen (1989) found that the  $K_d$  values of cadmium sorption for 63 samples of Danish agricultural soils correlated very well ( $r^2 = 0.72$ ) with soil pH. Soil organic matter content, as a second parameter, improved the correlation ( $r^2 = 0.79$ ). The transport of metals through

the soil column is controlled by a host of parameters with pH being the most sensitive. Without considering the influence of geochemical background, influence of the land utilization, long term behavior, this study only focus on the leaching to groundwater. Empirical expressions have been developed to predict the metal adsorption behavior of these soils. Based upon these expressions, it is possible to establish standards for the clean-up of metal contaminated soils and incorporate the influence of pH which is the most important factor in controlling the sorption of metals.

In this study the metal adsorption characteristics of five typical Tainan County soils in Taiwan, exemplified by Cd(II), Cu(II), Zn(II) and Cr(VI) as a function of pH, were evaluated. Soil adsorption behavior is the criterion on which to establish a standard based on a maximum permissible concentration in groundwater. This maximum level of metal in soil for which the equilibrium soluble metal does not exceed the drinking water standard can be computed from the measured partition coefficient for Cd(II), Cu(II), Zn(II) and Cr(VI) as a function of pH for five Tainan soils.

Based on these sorption data, we can predict the maximum concentration of metal which would be expected to be in a solution that was in contact with a soil having a given metal concentration and pH. It is possible that this predicted concentration is greater than that which would actually be observed. The overprediction could result from such phenomena as slow desorption. In that case, the overprediction would provide a measure of insurance when used as a soil standard. However, this insurance would be at the cost of greater levels of soil remediation or lower levels of introduction of metals. It is important to ascertain the magnitude of the difference between the sorption and desorption values. Column measurements allow approximation of flow conditions. If similar partition coefficients are found from batch and column methods one can utilize partition coefficients obtained directly from batch methods.

## METHODS

### Soil samples

The five soil samples, Wan-li loam; Kuei-jen loam; Hu-tou-pi sandy loam; Niu-chou-pu sandy loam, Hu-shan farm sand, represent a selection of the major soil types present in Tainan County. 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. For the soil analyses presented in Table 1, organic matter was measured by the Walkley-Black wet combustion method (Sims and Heckendorn, 1991b); soil pH was measured in 0.01 M CaCl<sub>2</sub> (1:1 soil:solution ratio) (Storer, 1991); soil particle size distribution was determined by a sedimentation (hydrometer) method (Sims and Heckendorn, 1991a); soil metal oxides were measured by the ammonium oxalate extraction method (Iyengar *et al.*, 1981).

Table 1. Soil properties

| Soil name              | Clay | pH                   | Organic Matter | ECEC       |
|------------------------|------|----------------------|----------------|------------|
|                        | %    | In CaCl <sub>2</sub> | %              | (meq/100g) |
| Wan-li loam            | 11.5 | 6.51                 | 3.8            | 1.0        |
| Kuei-jen loam          | 9.0  | 6.38                 | 0.9            | 8.1        |
| Hu-tou-pi sandy loam   | 10.0 | 3.96                 | 0.8            | 8.1        |
| Niu-chou-pu sandy loam | 12.0 | 4.81                 | 0.3            | 8.5        |
| Hu-shan farm sand      | 6.5  | 6.89                 | 1.1            | 7.3        |

### Metal adsorption studies

Batch equilibration studies were conducted for Cd(II), Cu(II), Zn(II) and Cr(VI) for the five Tainan soils. The soil samples were 1.00±0.01 g per 100 mL solution. Ionic strength was maintained at 0.01N with NaNO<sub>3</sub>. We added cadmium, zinc, copper, and chromium nitrate and potassium chromate at concentrations of 1x10<sup>-4</sup> M. Fifteen pH values covering the range from 3 to 10 were used and the temperature was

maintained at room temperature ( $25 \pm 2^\circ\text{C}$ ). The pH values of the samples were adjusted by adding NaOH or  $\text{HNO}_3$ , as required. Samples were shaken at 150 rpm for 24 hours. The pH values were measured again after 24 hours shaking. These values were taken to be the reaction values. The samples were then filtered through 25 mm diameter,  $0.45 \mu\text{m}$  membrane filters. The cadmium, zinc, and copper concentration in the filtered solution were determined by atomic absorption spectrophotometry using a Atomic Absorption Spectrophotometer.

The procedures for hexavalent chromium determination follow those of ASTM D 1687-86, Standard Test Methods for Chromium in Water (Storer 1990). The hexavalent chromium concentration in the filtered solution was determined by reddish-purple color complex developed between 1,5-diphenylcarbohydrazide and chromate ions in an acid solution. Absorbance was then measured at 540 nm with a UV spectrophotometer.

Samples for the determination of adsorption isotherms for cadmium were prepared by adding 1.0 g of soil to 100 ml of 0.01 M  $\text{NaNO}_3$ . The pH was adjusted by the addition of small volumes of  $\text{HNO}_3$  or NaOH, as necessary, to attain the desired pHs of 4.0, 5.5, and 7.0. The initial Cd(II) concentrations ranged from  $1.0 \times 10^{-6}$  to  $5 \times 10^{-3}$  M, and the equilibration time was 24 hours at room temperature ( $25 \pm 2^\circ\text{C}$ ). After 24 hours, the pH was readjusted to the initial value, and the samples were filtered through  $0.45 \mu\text{m}$  membrane filters. The changes in pH after 24 hours were small (all pH values were within  $\pm 0.3$  pH). The amount of adsorbed Cd was calculated as the difference between the amount added initially and that remaining in solution.

#### Calculation of partition coefficient

The batch adsorption data were transformed to conditional partition coefficients ( $K_d$ ) which are a function of pH. To compute  $K_d$ , we must first convert the measurements of initial ( $C^0$ ) and filtered concentrations ( $C'$ ).

$$K_d = \frac{C_s}{C_w} (\text{mL/g}) \quad (1)$$

$C_s$  = metal concentration in soil (mg/g)

$C_w$  = metal concentration in water (mg/mL)

$$K_d = \frac{\frac{(r) \times (\text{molecular weight of metal})}{S}}{(1-r) \times (\text{molecular weight of metal})} = \frac{V \times r}{S \times (1-r)} \left( \frac{\text{mL}}{\text{g}} \right) \quad (2)$$

$r$  = fraction adsorbed:

$S$  = amount of soil used (g)

$V$  = volume of solution (mL)

#### Computation of maximum permissible amount of metal in soil

To apply sorption data in the decision making process, it is necessary to use the  $K_d$  and appropriate soil-groundwater conditions in the environment. The calculation determines the maximum concentration of metal which will not exceed Water Quality Standards. This criterion can be utilized as a soil standard which will protect groundwater quality.

The  $K_d$  values are not directly useful, because the soil-to-water ratio in the laboratory experiments does not equal that in the field. We can convert the laboratory-derived partitioning values to that in the field by accounting for the amount of moisture in field soils.

The maximum permissible metal concentration in soil,  $[M]_s$ , is established by multiplying the Drink Water Standard,  $D$ , by the partition coefficient,  $K_d$ .

$$[M]_s = K_d \times D \quad (3)$$

$D$  = drinking water standard (in Taiwan : Cd: 0.01; Cu: 1; Zn: 5; Cr(VI): 0.05  $\mu\text{g/mL}$ )

The mass balance of metal in soil including solid phase and soil water:

$$Y_t = Y_s + Y_w \quad (4)$$

$Y_t$  = total contaminants in soil

$Y_s$  = contaminants bound by dry soil solids

$Y_w$  = contaminants dissolved in soil water

Total amount of metal in the soil is the concentration of metal in the soil times the mass of the soil ( $W_s$ ):

$$Y_s = (K_d \times D) \times W_s \quad \text{or} \quad (5)$$

$$Y_s = (K_d \times D) \times (d_s \times V_s) \quad (6)$$

$d_s$  = density of soil particle (commonly, 2.65  $\text{g/mL}$ )

$V_s$  = volume of soil particle ( $\text{mL}$ ) =  $(1-p) \times V$

$p$  = porosity of soil =  $V_v/V$

$V$  = volume of soil ( $V_s + V_v$ )

$V_v$  = void volume of soil

The total amount of metal in the pore water is the concentration of metal in the pore water times the volume of the pore water ( $V_p$ ). The maximum permissible concentration of metal in the pore water was assumed to be the drinking water standard:

$$Y_w = D \times V_p \quad (7)$$

Total amount of metal in the soil is the sum of the mass of metal on the particles and that in the pore water.

$$Y_t = \{K_d \times D \times d_s \times (1-p) \times V\} + (D \times V_p) \quad (8)$$

We can use this expression for the total amount of contaminants to calculate the Soil Metal Criteria,  $SMC$

$$SMC = (\text{Total contaminants}) / \text{Total weight of soil} \quad (9)$$

Since

$$\text{total weight of soil} = d_s \times (1-p) \times V$$

We can now write the allowed soil concentration as:

$$SMC = \frac{\{K_d \times D \times d_s \times (1-p) \times V\} + (D \times V_p)}{d_s \times (1-p) \times V} \quad (10)$$

$$\text{SMC} = (\text{Drinking Water Standard}) \times \left\{ K_d + \frac{p \times f}{d_s \times (1 - p)} \right\} \quad (11)$$

$f$  = degree of water saturation in soil =  $V_p/V_v$

Equation 11 permits us to evaluate the maximum allowable soil concentration that will not cause a violation of the Drinking Water Standard. This maximum allowable soil concentration is dependent on the partition coefficient, the soil porosity, the moisture content of the soil and the density of the soil particles. In most situations, the value of  $K_d$  is much greater than that of the second term in equation 11.

## RESULTS AND DISCUSSION

The sorption data have been evaluated to provide maximum soil contamination levels that will not exceed drinking water standards. Different soils exhibit different absorption ability, as is shown in Fig. 1, which compares the adsorption of cadmium by Wan-li loam; Kuei-jen loam; Hu-tou-pi sandy loam; Niu-chou-pu sandy loam, Hu-shan farm sand. The soils vary in their strength and capacity for metal binding. Metal adsorption was highly pH dependent. The Wan-li loam displayed the greatest extent of adsorption. It adsorbed metals much more than did the other four soils. Niu-chou-pu sandy loam had the poorest adsorption capacity.

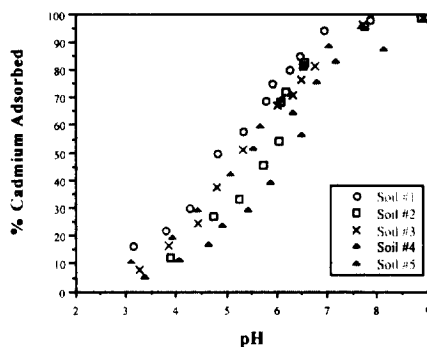


Figure 1. Adsorption of  $1 \times 10^{-4}$  M cadmium onto five soils (soil #1: Wan-li loam; soil #2: Kuei-jen sandy loam; soil #3: Hu-tou-pi sandy loam; soil #4: Niu-chou-pu loam; soil #5: Hu-shan farm sand).

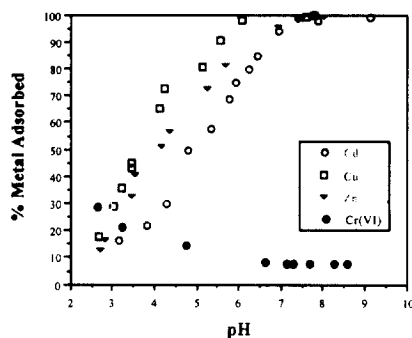


Figure 2. Adsorption of  $1 \times 10^{-4}$  M cadmium, copper, zinc and hexavalent chromium onto Wan-li loam. Soil:water = 1g/100 mL; I = 0.01 M NaNO<sub>3</sub>; T = 25°C.

All cationic metals had similar adsorption behavior, but copper had the strongest adsorption (Fig. 2). Figure 2 shows data from our study for the adsorption of cadmium, copper, zinc and hexavalent chromium by Wan-

li loam from Tainan. At higher pH, virtually all the added cadmium, copper and zinc was adsorbed. As the pH decreases, the concentration of soluble cationic metals increased. For hexavalent chromium, a reverse trend, in which adsorption is of a maximum at low pH and decreases with increasing pH, is found.

We evaluated whether the high concentrations of metal used in the experiments invalidated the use of the linear partitioning relationship of equation 1 to describe the distribution between soluble and sorbed metal. We compared the partition coefficient obtained for the linear portion of the Langmuir isotherm using the constants at pH 5.5 in Table 2 to the experimental values of soluble and sorbed cadmium using equation 1. The mean deviation between these values for the five soils was 43 percent and the maximum deviation was a factor of 3. This difference is a small contribution to the scatter of our cadmium data in Table 3.

Table 2. Parameters for the nonlinear regression of  $C_s$  vs.  $C_w$  of the Langmuir equation for the adsorption of cadmium onto five Tainan soils at three pH values.

| Soil name              | pH 4.0              |        |       | pH 5.5              |        |       | pH 7.0              |        |       |
|------------------------|---------------------|--------|-------|---------------------|--------|-------|---------------------|--------|-------|
|                        | $\Gamma_m$          | K      | $R^2$ | $\Gamma_m$          | K      | $R^2$ | $\Gamma_m$          | K      | $R^2$ |
|                        | ( $\mu\text{g/g}$ ) | (L/mg) |       | ( $\mu\text{g/g}$ ) | (L/mg) |       | ( $\mu\text{g/g}$ ) | (L/mg) |       |
| Wan-li loam            | 2465                | 0.003  | 0.995 | 5420                | 0.084  | 0.992 | 6397                | 0.126  | 0.996 |
| Kuei-jen loam          | 163                 | 0.006  | 0.006 | 845                 | 0.037  | 0.994 | 1036                | 1.121  | 0.989 |
| Hu-tou-pi sandy loam   | 1231                | 0.017  | 0.996 | 2851                | 0.115  | 0.997 | 3762                | 0.213  | 0.991 |
| Niu-chou-pu sandy loam | 134                 | 0.174  | 0.998 | 372                 | 0.213  | 0.996 | 756                 | 0.227  | 0.993 |
| Hu-shan farm sand      | 1724                | 0.073  | 0.997 | 3124                | 0.048  | 0.998 | 4763                | 0.112  | 0.985 |

Table 3. Partition coefficient of cadmium and zinc (mL/g) for five Tainan soils at different pH values

| Soil name              | pH 4 |      | pH 5  |       | pH 6  |        | Soil pH |        |
|------------------------|------|------|-------|-------|-------|--------|---------|--------|
|                        | Cd   | Zn   | Cd    | Zn    | Cd    | Zn     | Cd      | Zn     |
| Wan-li loam            | 36.2 | 93.1 | 169.5 | 218.5 | 365.1 | 594.6  | 1594.9  | 5667.0 |
| Kuei-jen loam          | 17.4 | 39.5 | 41.4  | 128.1 | 116.0 | 373.6  | 303.2   | 994.9  |
| Hu-tou-pi sandy loam   | 22.7 | 50.6 | 69.8  | 133.0 | 199.4 | 1080.2 | 22.1    | 33.8   |
| Niu-chou-pu sandy loam | 12.9 | 28.4 | 33.0  | 94.6  | 69.8  | 258.5  | 23.9    | 72.5   |
| Hu-shan farm sand      | 25.5 | 38.9 | 74.2  | 123.2 | 220.5 | 334.6  | 461.8   | 1989.0 |

The higher the partition coefficient values, the higher the ratio of metal bound by soil to that remaining in solution. Different soils have very different absorption abilities. For example, at a given pH Wan-li loam could have a relatively high cadmium and zinc concentration, whereas Niu-chou-pu loam could have only relatively low concentrations (Fig. 3).

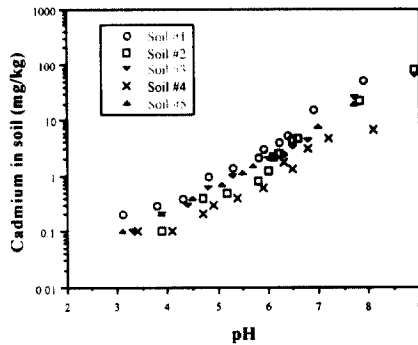


Figure 3. The maximum permissible level of five cadmium soils (soil #1: Wan-li loam; soil #2: Kuei-jen sandy loam; soil #3: Hu-tou-pi sandy loam; soil #4: Niu-chou-pu loam; soil #5: Hu-shan farm sand) for  $1 \times 10^{-4}$  M cadmium.

Because partition coefficients can be obtained easily by batch adsorption experiments, it is desirable to utilize them. However, the conditions in these experiments are very different from those found in the field where the soil is stationary and the question frequently is the prediction of desorption behavior. Environmental conditions, such as chemicals loading and water flow rate, change the rate and extent of chemical reaction on soils. Allen *et al.* (1995) investigated the desorption behavior of cadmium from five soils. Desorption was studied as a function of cadmium loading onto the soil and the flow rate of water through the soil column. They found that the partition coefficients ( $K_d$ ) determined from column flow processes using the double extrapolation technique were very close to those obtained from batch measurements ( $R^2 = 0.98$ ) (Table 4). In order to better understand the mechanism of adsorption, Lee *et al.* (1996) proposed 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 for the adsorption of Cd by the 15 soils. They found that the 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. They developed the means to relate the sorption of Cd by the different soils to enable the results to be predictive of sorption to other soils. Therefore, the batch equilibrium methods provide a good and more convenient estimation of the value obtained for desorption of cadmium contaminant in the dynamic systems.

Table 4. Comparison of  $K_d$  from batch and column flow measurements; Data from Allen *et al.* (1993)

| Soil Name                     | Batch $K_d$ | Column $K_d$ | $R^2$ |
|-------------------------------|-------------|--------------|-------|
|                               | (mL/g)      | (mL/g)       |       |
| Boonton Union County          | 300         | 280.2        | 0.98  |
| Downer loamy sand             | 48          | 44.7         |       |
| Fill materials from Delaware  | 110         | 76.5         |       |
| Freehold sandy loam (surface) | 40          | 35.6         |       |
| Rockaway stony loam           | 130         | 129.7        |       |

The values range from 0.2 to 16.0 mg/kg for cadmium and 0.17 to 28.0 g/kg for zinc at the natural pHs of the soils (Table 5). The values of the SMC for cadmium can be compared to those that have been proposed or are in use as standards. Wu *et al.* (1991) reported proposed maximum allowable concentration of cadmium in the soils of a number of countries (Table 6). Comparison of Table 5 and Table 6 indicates that the proposed standards may be inadequate for some soils. The proposed standard for France would be unnecessarily low for Wan-li loam, Kuei-jen loam and Hu-shan farm sand at the soil pH but would not protect groundwater in contact with sandy soils such as Hu-tou-pi sandy loam and Niu-chou-pu sandy loam. The USSR standard would fail to protect groundwater in most of the Tainan soils listed in Table 6 at their natural pH values. This approach presented here offers the ability to provide SMC values for varying environmental soil pH conditions that could result from acid rain or from soil treatment.

Table 5. Maximum permissible level of cadmium and zinc (mg/kg) for five Tainan soils at different pH values

| Soil name              | pH 4 |       | pH 5 |        | pH 6 |        | Soil pH |         |
|------------------------|------|-------|------|--------|------|--------|---------|---------|
|                        | Cd   | Zn    | Cd   | Zn     | Cd   | Zn     | Cd      | Zn      |
| Wan-li loam            | 0.4  | 466.1 | 1.7  | 1093.4 | 3.7  | 2973.7 | 16.0    | 28335.9 |
| Kuei-jen loam          | 0.2  | 198.2 | 0.4  | 641.3  | 1.2  | 1869.0 | 3.0     | 4975.5  |
| Hu-tou-pi sandy loam   | 0.2  | 253.8 | 0.7  | 665.6  | 2.0  | 5401.9 | 0.2     | 169.9   |
| Niu-chou-pu sandy loam | 0.1  | 142.6 | 0.3  | 473.8  | 0.7  | 1293.2 | 0.2     | 363.5   |
| Hu-shan farm sand      | 0.3  | 195.2 | 0.7  | 616.8  | 2.2  | 1673.9 | 4.6     | 9945.8  |

Table 6. Maximum allowable concentrations of cadmium in soils (Wu *et al.* 1991)

| Country | China | EEC | Canada | Scotland | France | F.R.<br>Germany | Italy | England | USSR |
|---------|-------|-----|--------|----------|--------|-----------------|-------|---------|------|
| *MAC    | 0.3   | 1-3 | 1.6    | 1.6      | 2      | 3               | 3     | 3.5     | 5    |

\*MAC: Maximum Allowable Concentrations (mg/kg)

The SMC value for cadmium in Kuei-jen at its native pH of 6.38 was 3.0 mg/kg. If the pH were raised to 7 the SMC would be approximately 9.5 mg/kg (Fig. 3) and if the pH were lowered to 4, the SMC would have to be lowered to approximately 0.2 mg/kg (Fig. 3; Table 5).

At any pH, for a given soil, zinc could have the highest soil concentration without causing a violation of the Drinking Water Standard because among all metals in this study (1) zinc has stronger adsorption ability and (2) zinc has the highest Drinking Water Standard. (Fig. 4).

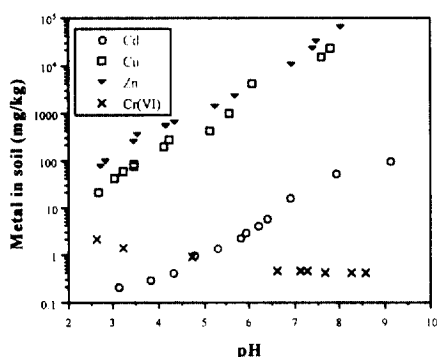


Figure 4. The maximum permissible level of cadmium, copper, zinc and hexavalent chromium onto Wan-li loam. Soil:water = 1g/100 mL; I = 0.01 M NaNO<sub>3</sub>; T = 25°C.

At the natural soil pH (Table 5), Wan-li loam has the highest allowed cadmium and zinc concentrations, Niu-chou-pu sandy loam has the lowest cadmium and Hu-tou-pi sandy loam has the lowest zinc concentration without violating the Drinking Water Standard because Wan-li loam has higher soil pH, organic matter and finer particle size, whereas Niu-chou-pu sandy loam has the lowest organic matter content and Hu-tou-pi sandy loam has the lowest soil pH of the soils studied.

## CONCLUSIONS

Metal adsorption is highly pH dependent. Different soils have very different adsorption abilities. The adsorption coefficient ( $K_d$ ), which is related to the percentage metal adsorbed, increases with rising pH values.

Data for partition coefficients can be used to compute maximum permissible metal concentrations in soil which will not result in a violation of the Drinking Water Standard.

These maximum permissible metal concentrations can be used as soil clean-up criteria and to predict the effect of metal addition to soil.

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