

# Heavy Metals Transport Model in a Sludge-treated Soil<sup>1</sup>

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## ABSTRACT

A transport model was developed to study the movement of Cu, Zn, and Cd in a forest soil. Adsorption of cationic forms of these metals by the soil was described by Freundlich adsorption isotherms. Two slug inputs of heavy metals were introduced during the first 37 days and between the 168th and 196th days of the 17-month simulation period. The transport model based on the adsorption of cationic forms of Cu, Zn, and Cd severely underpredicted the movement of these metals in both the 15- and 120-cm percolate when compared to previously observed field data. With the addition of assumed levels of less highly adsorbed chelated components to the model, percolate metal concentrations were roughly predicted at the 15-cm depth. However, data generated by the partitioned (chelate component) model also underpredicted movement of heavy metals to the 120-cm depth, indicating that transport processes other than accelerated movement of chelated compounds, such as "channelization", may be involved in the field.

**Additional Index Words:** Cu, Zn, Cd, Freundlich adsorption, dispersion coefficient, percolate, chelated metals, cationic metals, channelization.

The ability to predict heavy metal movement in soils would be of considerable value in managing a land disposal system for municipal or industrial wastes. Much research has been done (8, 11, 17, 24) concerning the disposition of potentially toxic heavy metals in land systems treated with sewage. In order to make better utilization of this type of data for the management of heavy metals disposal at other sites, it is desirable to have a predictive model for heavy metals transport in field soils.

The movement of adsorbed constituents through porous media under steady-state soil water conditions can be described by the following differential equation:

$$\frac{\rho}{\theta} \left( \frac{\partial S}{\partial t} \right) + \left( \frac{\partial C}{\partial t} \right) = D \left( \frac{\partial^2 C}{\partial x^2} \right) - v_p \left( \frac{\partial C}{\partial x} \right), \quad [1]$$

where

$C$  = concentration of the heavy metal in solution (mg/liter),

$S$  = amount of heavy metal adsorbed per gram of soil ( $\mu\text{g/g}$ ),

$D$  = dispersion coefficient ( $\text{cm}^2/\text{day}$ ),

$v_p$  = average pore water velocity ( $\text{cm}/\text{day}$ ),

$\rho$  = soil bulk density ( $\text{g}/\text{cm}^3$ ),

$\theta$  = volumetric water content ( $\text{cm}^3/\text{cm}^3$ ),

$x$  = linear distance in direction of flow (cm), and

$t$  = time (days).

The sink term  $(\partial S/\partial t)$  in Eq. [1] describes the adsorp-

tion or desorption of constituents by the soil. Predictive models utilizing Eq. [1] have been described in the literature for phosphorus (3, 15) and pesticide (9, 21) movement in soils. Skopp and Warrick (18) developed a closed mathematical solution for the miscible displacement of reactive solutes in soils. Unlike most models, theirs assumes that the reaction is limited by molecular diffusion into a stationary liquid phase of the soil. A model based on thermodynamic principles has been developed by Jurinak and Santillan-Medrano (5) describing the transport of Pb and Cd in calcareous soils. However, this model requires much more input data than is necessary for the solution of Eq. [1].

The objective of this research was to develop a relatively simple one-dimensional solute transport model for the movement of Cu, Zn, and Cd (applied to the soil via sludge) in a forest soil. The model will then be verified with field data from a previous study (17) to determine its utility as a management tool for heavy metals disposal on land.

## THEORY AND METHODOLOGY

### Adsorption Component

A programmed solution to Eq. [1] using S/360 CSMP simulation (22) was adapted to describe Cu, Zn, and Cd transport in soils. Adsorption of all three metals was described by the following Freundlich equation:

$$S = KC^{1/n}, \quad [2]$$

where  $S$  = amount of heavy metal adsorbed per gram of soil ( $\mu\text{g/g}$ ),  $C$  = equilibrium solution concentration of heavy metal ( $\text{mg}/\text{liter}$ ), and  $K$  and  $1/n$  = empirical constants. Freundlich constants utilized in the transport model (Table 1) were obtained in a previously described (16) batch equilibrium study using cationic forms of Cu, Zn, and Cd. The pH levels of the initial batch equilibration solutions were adjusted to approximately the levels found in the sludge (7.2). After equilibration pH levels approached that of the soil. Constants for Cu were based on 0.3-hour batch equilibrium data, while those for Zn and Cd were obtained from 3.0-hour batch equilibrium data.

### Dispersion Component

The dispersion coefficient,  $D$ , in Eq. [1] was evaluated by the miscible displacement of  $\text{Cl}^-$  through undisturbed soil cores 7.5 cm in length with a volume of  $441.5 \text{ cm}^3$ . Cores were taken using brass rings at depths of 0-7.5 cm and 7.5-15 cm at a forest site adjacent

Table 1—Freundlich constants ( $K$  and  $1/n$ ) utilized at the 0- to 7.5-cm and >7.5-cm depths in the CSMP cationic transport model for heavy metals

Transport model	0-7.5 cm depth		>7.5 cm depth	
	$K$	$1/n$	$K$	$1/n$
Cu†	386.4	1.00	158.1	1.00
Zn‡	138.0	0.94	30.9	0.69
Cd‡	32.4	0.82	7.4	0.71

† Based on 0.3-hour batch equilibrium data.

‡ Based on 3.0-hour batch equilibrium data.

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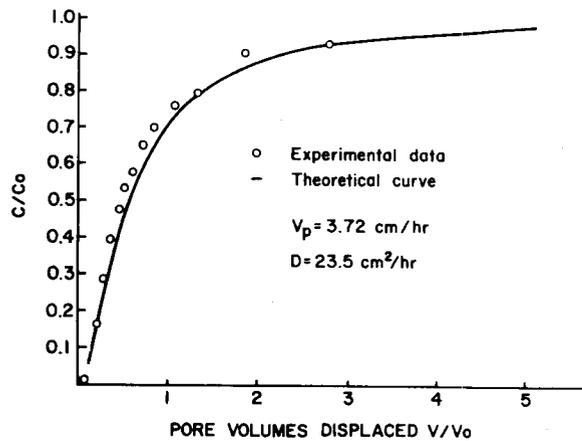


Fig. 1—Chloride breakthrough curve for 0- to 7.5-cm undisturbed soil core.

to the sludge disposal area (16). The cores were exposed to chloroform fumes to kill any macroinvertebrates, such as earthworms, prior to being placed in Tempe cells, where the cores were saturated with 0.01N CaSO<sub>4</sub>. Once saturated, the influent solution was changed to 0.006N CaCl<sub>2</sub>, containing 425 mg/liter Cl<sup>-</sup>. Flow rate of Cl<sup>-</sup> solution into the horizontal cores was controlled by means of a Mariott bottle. The Cl<sup>-</sup> concentrations in the effluent were monitored with an Orion specific ion electrode until they approached input concentrations. Chloride breakthrough curves for the 0- to 7.5-cm core were obtained for pore velocities of 7.77, 3.72, 2.87, and 1.01 cm/hour, while curves for the 7.5- to 15-cm core were obtained for pore velocities of 2.95, 2.06, and 0.75 cm/hour. The experimental breakthrough data were subsequently compared with theoretical curves based on the following solution (12) to the convective-dispersive equation:

$$C/C_0 = \frac{1}{2} \operatorname{erfc} \left[ \frac{L - v_p t}{(4Dt)^{1/2}} \right] + \frac{1}{2} \exp(v_p L/D) \operatorname{erfc} \left[ \frac{L + v_p t}{(4Dt)^{1/2}} \right] \quad [3]$$

where  $C_0$  is the influent concentration (mg/liter) and  $L$  is the core length (cm). The best least squares fit between theoretical curves generated by Eq. [3] and the experimental data determined the value of  $D$  to be used. Figure 1 shows the results obtained for one experiment on the 0- to 7.5-cm core. The relative concentration,  $C/C_0$ , is plotted vs. the number of pore volumes displaced by the feed solution,  $V/V_0$  ( $v_p t/L$ ), where  $V$  is the volume of effluent displaced and  $V_0$  is the volume of water in the core. It can be seen from Fig. 1 that Eq. [3] does not require that  $C/C_0 = 0.5$  at 1 pore volume displaced. The nonsigmoid appearance of the calculated curve in Fig. 1, in which the value of  $C/C_0$  at one pore volume was approximately 0.7, is the consequence of the small value of the dimensionless group,  $v_p L/D$ , known as the Peclet number. When different analytical solutions are used, such as those given by Lindstrom et al. (9) and Danckwerts (2), the breakthrough curve passes much closer to 0.5 after one pore volume. Lindstrom's solution gave a value of  $C/C_0$  equal to 0.43 after one pore volume when using the same  $D$  as in Fig. 1, while the solution given by Danckwerts always passes through 0.5 after one pore volume. These two analytical solutions were unable to describe the experimental data and, thus, Eq. [3] was utilized since it fit the data best. A theoretically more acceptable comparison between Eq. [3] and the experimental data could have been made by increasing the length,  $L$ , of the undisturbed core. However, preliminary experimentation with the transport model indicated that the value of  $D$  had very little influence on the propagation of the concentration front of heavy metals due to the large adsorption component.

Dispersion data (cm<sup>2</sup>/hour) for the 0- to 7.5-cm and 7.5- to 15-cm cores are plotted vs. their respective  $v_p$  (cm/hour) in Fig. 2. The regression relationship describing the data is as follows:

$$D = 1.09 + 7.16 v_p \quad (r = 0.938) \quad [4]$$

For low values of  $v_p$  (i.e., <0.03 cm/hour),  $D$  was set equal to the

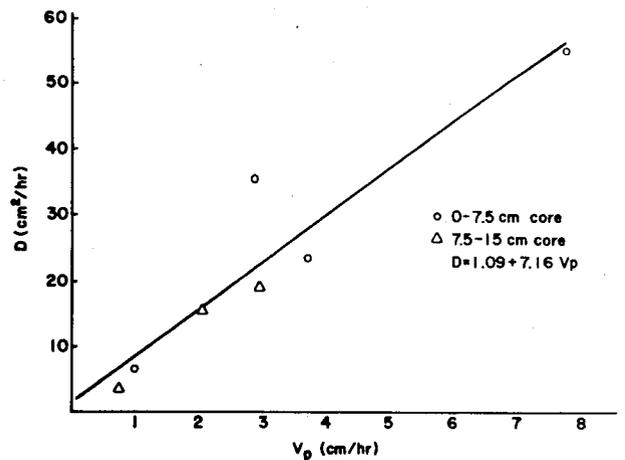


Fig. 2—Values of calculated dispersion coefficients vs. pore water velocities. The solid line represents the least squares linear regression equation.

molecular diffusion coefficient of Cl<sup>-</sup> in free solution, multiplied by a tortuosity factor of 0.6, yielding a  $D$  value of 0.026 cm<sup>2</sup>/hour (6). Equation [4] was derived for saturated conditions and, for modeling purposes, is assumed to also hold for unsaturated conditions.

#### Soil Physical-Chemical Data

Soil physical parameters used in the model included volumetric soil water content, 0.25, and bulk density of both the 0- to 7.5-cm depth, 1.08 g/cm<sup>3</sup>, and the 7.5- to 15-cm depth, 1.30 g/cm<sup>3</sup>. Net monthly soil water fluxes used in the model were evaluated by a water budget method depicted in Table 2. Precipitation data were obtained from a gaging station located approximately 150 m from the sludge disposal site. Net interception losses were estimated using guidelines given by Lull (10) for northern hardwoods. Hydraulic inputs associated with liquid sludge applications were measured in the field by bail testing. Evapotranspiration (ET) was estimated by the Thornthwaite (20) method and losses were distributed throughout the soil profile based on an estimate of root mass distribution (14). No heavy metal uptake by the roots was considered, since vegetative cycling in forest ecosystems is a dynamic process and difficult to model.

The initial solution levels of Cu (4.9 ppb), Zn (44.5 ppb), and Cd (0.25 ppb) used in the model were based on average percolate concentrations found in a field control plot from a previous study (17). Soil at the forested sludge disposal site was a Hublersburg clay loam (Typic Hapludult) with an average pH of 5.0 in the upper 7.5 cm, and a pH of 4.7 in the 7.5- to 15-cm depth. The upper 7.5-cm depth interval of soil had a much higher organic matter content (10.6%) and CEC (18.4 meq/100 g soil) than the 7.5- to 15-cm depth interval (organic matter = 5.4%, CEC = 8.4 meq/100 g soil).

#### Heavy Metals Transport Model

The mechanics and theory of the transport model used are presented in some detail by van Genuchten and Wierenga (22). Equation [1] is evaluated in the model in terms of one dependent variable,  $C$ , by incorporating Eq. [2] into the transport equation as follows:

$$\partial C/\partial t = D'(\partial^2 C/\partial x^2) - v'(\partial C/\partial x) \quad [5]$$

where

$$D' = D/R_f$$

$$v' = v_p/R_f \text{ and}$$

$$R_f = 1 + \rho K C^{(1/n-1)}/n\theta$$

The retardation factor,  $R_f$ , was assumed to be independent of the adsorption direction.

A 17-month simulation period was used, with sludge inputs occurring during the first 37 days and again between the 168th and 196th days of the period, as in the field study (17). Input concen-

Table 2—Field water budget data for sludge disposal plots

Characteristic	1974		1975											1976			
	Nov.†	Dec.	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.
	cm																
Precipitation	4.93	11.38	7.82	7.95	6.27	5.49	12.93	13.34	4.95	11.13	18.29	9.04	5.99	5.74	4.93	5.61	6.22
Net interception	0.36	0.79	0.56	0.56	0.43	0.38	1.93	2.01	0.74	1.68	2.74	1.35	0.43	0.41	0.36	0.38	0.43
Adjusted potential ET	1.62	0.0	0.0	0.0	0.40	2.41	9.24	11.50	13.59	12.27	6.81	5.23	2.53	0.0	0.0	0.32	2.01
Net precipitation	2.95	10.59	7.26	7.39	5.44	2.70	1.76	-0.17	-9.38	-2.82	8.74	2.46	3.03	5.33	4.57	4.91	3.78
Liquid sludge loadings																	
High	17.33	0.41				4.21	2.70										
Low	7.32	1.02				3.24	1.67										
Total hydraulic loadings																	
High	20.28	11.00	7.26	7.39	5.44	6.91	4.46	-0.17	-9.38	-2.82	8.74	2.46	3.03	5.33	4.57	4.91	3.78
Low	10.27	11.61				5.94	3.43										
Soil water flux																	
High	0.81	0.35	0.23	0.26	0.19	0.23	0.14	-0.01	-0.30	-0.09	0.29	0.08	0.10	0.17	0.15	0.17	0.12
Low	0.41	0.37				0.20	0.11										

† Nov. 1974 data based on last 25 days of the month, all other data based on entire months.

trations of heavy metals shown in Table 3 for the two simulated sludge application periods were calculated using known total concentrations of these metals (17) together with hydraulic loadings (via liquid sludge application) and water budget data (Table 2) for the 17-month period. The two separate sludge applications with concentrations of heavy metals as shown in Table 3 were simulated as pulse inputs of 37 and 28 days.

The simulation of heavy metals transport involved dividing the soil into a number of finite layers. The net flux of heavy metals into each layer was calculated and subsequently integrated for each time step, directly leading to new estimates of both solution and adsorbed concentrations. Printed output of *C* and *S* was obtained for the distribution of heavy metals in both time and space. A detailed description of the CSMP transport program is given by Sidle.<sup>3</sup>

## RESULTS AND DISCUSSION

The transport model predicted virtually no movement of Cu, Zn, and Cd in soil water percolate to either the 15- or 120-cm depths during the 17-month simulation period. This was in spite of the fact that input levels of heavy metals were based on total concentrations in the sludge, rather than on water or weak acid-extractable concentrations. The results predicted were in contrast to field data, which showed average increases in concentrations from the high sludge treatment plots over control plots of 322,

422, and 833% for 15-cm percolate Cu, Zn, and Cd, respectively. Corresponding concentration increases in field percolate samples from the 120-cm depths were 121, 305, and 800% for Cu, Zn, and Cd, respectively.

One of the reasons for the underprediction of metal movement in the soil water phase could be that the sink term in the model (Eq. [2]) was based on the adsorption of cationic forms of Cu, Zn, and Cd. The possibility of chelated forms of these metals existing in soil solution following sludge application may lead to a greater penetration in the soil. Chelated forms of heavy metals are more soluble in soil systems (4) and, thus, would be transported to greater depths than their cationic counterparts. Lagerwerff et al. (7) indicated that > 60% of the total Cu, and > 12% of the total Zn, in an initial sewage sludge aqueous leachate were in noncationic forms (anionic, amphoteric, or uncharged). This indicates the potential presence of chelated metals in sewage sludge.

In order to see if chelation could generate the magnitude of heavy metals transport encountered in field percolate samples at the 15-cm depth, it was assumed that the input concentrations of metals were partitioned between cationic and chelated components. Freundlich adsorption constants (*K*), as well as amounts of the chelated component, were adjusted in an attempt to simulate the breakthrough of heavy metals in the 15-cm percolate from the high sludge treatment area.

By partitioning only 0.7% of the total applied Cu in a chelated form (and the remaining 99.3% in cationic form), and assuming Freundlich constants for the chelated component of *K* = 3.0 for the 0- to 7.5-cm depth and *K* = 1.3 for the > 7.5-cm depth, the general magnitude of Cu movement at the 15-cm depth in the field was simulated fairly accurately (Fig. 3). Higher levels of Cu in field percolate samples than predicted by simulation during May and June of 1975 may reflect the accelerated movement of Cu associated with the hydraulic applications in the spring sludge treatment. Coefficients of variability (C.V.) for field percolate Cu data ranged from 25-82%, with only the months of Apr., Sept., and Dec. of 1975 having C.V.'s > 70%. The levels of Cu in 120-cm soil water percolate predicted by this partitioned model showed almost no increase, indicating that processes other than chelation may have been involved in the movement of Cu to greater depths in the soil profile.

<sup>3</sup>R. C. Sidle. 1976. Transport of heavy metals in a sludge-treated forested area. Ph.D. Dissertation. Dep. of Agronomy, The Pennsylvania State Univ., University Park, PA 16802.

Table 3—Input loading and concentration of heavy metals used in the CSMP model for the two sludge application periods

Heavy metal	Average metal loading†	Sludge hydraulic loading	Net precipitation hydraulic loading	Average concentration in input sludge solution
	µg/cm <sup>2</sup> per day		cm/day	mg/liter
1st sludge application (7 Nov.-12 Dec. 1974)				
Cu	3.51	0.48	0.24	4.89
Zn	4.29	0.48	0.24	5.96
Cd	0.036	0.48	0.24	0.051
2nd sludge application (22 Apr.-19 May 1975)				
Cu	4.12	0.25	0.10	11.84
Zn	4.50	0.25	0.10	10.94
Cd	0.042	0.25	0.10	0.120

† Based on data from Sidle and Kardos (17) for the high treatment sludge disposal area.

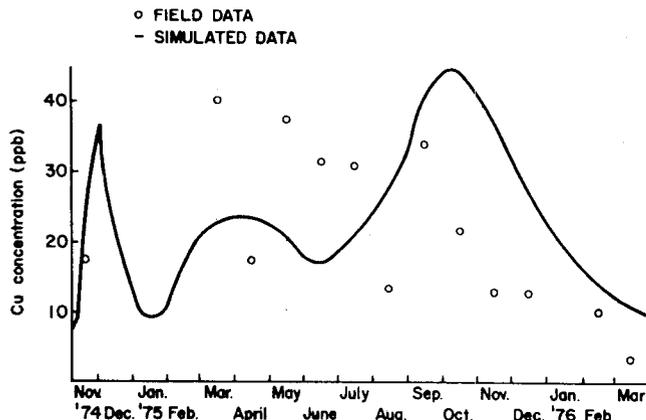


Fig. 3—Simulated and field percolate Cu data (15-cm depth) from high treatment sludge disposal site.

Simulation of Zn movement in soil water percolate (Fig. 4) at the 15-cm depth was generated by partitioning 9.0% of the total applied Zn in a chelated form (91.0% in cationic form) with Freundlich constants of  $K = 5.0$  for the 0- to 7.5-cm depth and  $K = 1.0$  for the  $> 7.5$ -cm depth. As with Cu, the higher levels of Zn in percolate samples taken shortly after the spring 1975 sludge applications were not predicted by the partitioned model. Also, levels of Zn in the 120-cm simulated percolate increased only slightly ( $\sim 6$  ppb) over the 17-month period, again indicating that other transport processes may have been involved.

Simulation of Cd movement in the 15-cm percolate (Fig. 5) was achieved by partitioning 10.0% of the total applied Cd in a chelated form (90.0% in cationic form) with Freundlich constants of  $K = 5.0$  for the 0- to 7.5-cm depth and  $K = 1.0$  for the  $> 7.5$ -cm depth. Again, high levels of Cd in 15-cm field percolate samples taken after the spring 1975 sludge application were not simulated by the partitioned model. Higher C.V.'s were found for field percolate Zn (16–113%) and Cd (9–123%) than for Cu, with only the months of Mar., June, and Aug. of 1975 having C.V.'s  $< 55\%$ . Virtually no Cd movement to the 120-cm depth was predicted by the partitioned model.

Net changes in total amounts of soil Cu, Zn, and Cd sampled before and after sludge applications are compared to net changes predicted by both the cationic and partitioned transport models (Table 4). Net increases in total soil Cu predicted by both models in the 0- to 7.5-cm

Table 4—Net changes in total amounts of soil Cu, Zn, and Cd sampled before (Oct. 1974) and after (Nov. 1975) sludge applications compared to net changes predicted by the cationic and partitioned models

Heavy metal	Depth cm	Net increase		In field samples
		Predicted by cationic model	Predicted by partitioned model	
		$\mu\text{g/g}$		
Cu	0-7.5	32.54	32.28	21.88
	7.5-15	$< 0.01$	0.04	0.14
Zn	0-7.5	34.06	31.36	2.24
	7.5-15	0.15	0.37	-19.21
Cd	0-7.5	0.326	0.301	0.448
	7.5-15	0.002	0.005	0.374

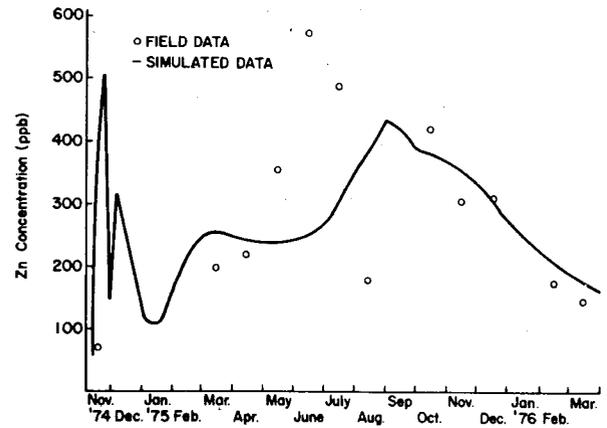


Fig. 4—Simulated and field percolate Zn data (15-cm depth) from high treatment sludge disposal site.

depth were similar in magnitude to the increases found in the field soil. A slight increase ( $0.14 \mu\text{g/g}$ ) in field soil Cu was found at the 7.5- to 15-cm depth, compared with virtually no increases predicted by either model. Due to the high variability found for total soil Zn in field samples, predicted values did not compare favorably with the experimental data. Simulated Zn movement into the 7.5- to 15-cm depth of soil in both models was greater than the corresponding Cu movement. Net increases in total soil Cd in field samples were slightly higher than predicted levels at the 7.5- to 15-cm depth.

The inability of the model to describe the transport of heavy metals in the soil water percolate to depths where movement was observed in the field casts some doubt on the suitability of the model for simulating transport processes in the field. A possible explanation for the severely underpredicted migration of heavy metals in the soil water could be the accelerated movement of metals through interconnected soil channels at times of high hydraulic loadings. Since the soil water fluxes used in the model are based on a monthly water budget (Table 2), individual rainfall events and hydraulic loadings of sludge tend to be averaged over fairly long time periods. Leaching studies with chloride (13) and nitrate (19, 25) under natural field conditions have shown that a large fraction of the percolating soil water tends to move in large pores between aggregates, especially during moist soil water condi-

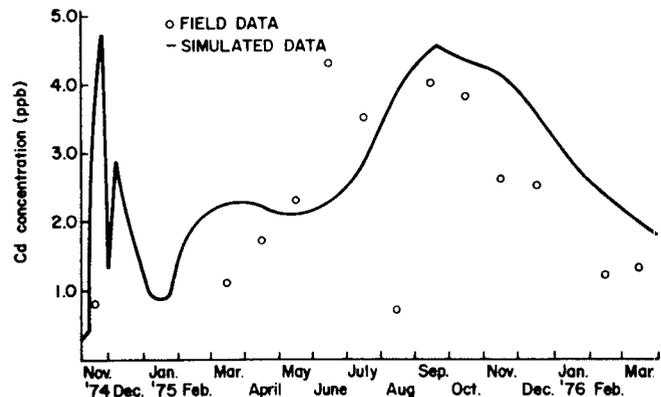


Fig. 5—Simulated and field percolate Cd data (15-cm depth) from high treatment sludge disposal site.

tions. Quisenberry and Phillips (13) observed that rapid and deep percolation of applied water occurred in a field soil in which the volumetric soil water content was below the upper limit of the water-holding capacity of the soil. This suggested the influence of large soil pores or cracks in water conduction. The forest soil environment in our study seems particularly endowed with such water-conducting pores, including root channels, macroinvertebrate passageways, and freeze-thaw cracks, as well as inter-aggregate channels. Evidence of the importance of these macropores in the channelization of soil water in forest soils has been demonstrated by Aubertin (1). Assuming that channelization did occur in our field experiment, the actual adsorbing mass of soil contacted by the percolating heavy metals would have been greatly reduced, leading to a faster and greater migration of metals in the percolate than would be predicted by the model.

### SUMMARY AND CONCLUSIONS

Results from adsorption and dispersion studies have been incorporated into a predictive transport model for Cu, Zn, and Cd in a forest soil. The dispersion study showed that  $D$  is related to  $v_p$  by Eq. [4]. Freundlich adsorption isotherms were used to characterize the adsorption of Cu, Zn, and Cd by a forest soil.

The transport model using sink terms based only on adsorption of cations underpredicted the movement of percolate Cu, Zn, and Cd at the 15- and 120-cm depths. Even with the addition of less highly adsorbed chelated components to the models, which roughly predicted field percolate Cu, Zn, and Cd levels at the 15-cm depth, virtually no penetration of these metals was predicted at the 120-cm depth. Data generated by the partitioned transport model indicates that transport processes other than chelation may be involved in the leaching of these metals to the 120-cm depth, as observed in field percolate samples. One such transport mechanism could be the "channelization" of percolating solution preferentially through the larger interconnected soil pores at times of high hydraulic loadings. This would result in a greatly reduced active adsorbing mass of soil during periods of high soil water fluxes, since the percolating solution would not move through all soil pores and, hence, would be unable to react with all sorption sites in the soil.

### RECOMMENDATIONS

It was evident that the type of simple transport model containing a cationic sink term advocated in the scope of this study would not be a valuable management tool for disposal of heavy metal-laden sludge on land systems. Some recommendations for further research that may provide data needed to obtain better predictions of heavy metal transport in sludge-treated soils include:

- 1) Extracting samples of the applied sewage sludge with water (and 0.1N HCl) and then determining the proportions of cationic vs. noncationic metals in aqueous extracts. This would provide a better idea of the distribution of heavy metals between aqueous and solid phases and, also, the distribution of cationic and complexed forms of metals in the aque-

ous phase. A laboratory study now in progress indicates that substantial levels of heavy metals may be in noncationic forms, and that proportions vary from sludge to sludge.

- 2) Conducting adsorption studies with various chelated forms of heavy metals to better describe the chelated portion of the partitioned model.
- 3) Performing the dispersion study with longer soil columns. In this study, we were restricted to using the 0- to 7.5-cm and 7.5- to 15-cm depths of soil due to great differences in their physical and chemical properties.
- 4) Utilizing a transport model that would allow for the previously mentioned "channeling" phenomena and diminished active adsorbing soil mass. This model should take into account the aggregated nature of field soil and the limited accessibility of heavy metal adsorption sites away from the larger channels. Such a model has been proposed very recently (23) and should be further investigated.

Since the ultimate goal of this heavy metals transport model is to establish a management tool for sludge disposal, the additional studies could provide necessary information to develop more attractive transport models. However, individual sludges vary greatly in their heavy metal composition and, thus, adsorption and extraction studies may still have to be conducted for each sludge.

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