

LAND DISPOSAL OF DRINKING WATER SLUDGE

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SUMMARY: Sludges produced during purification of drinking water can be successfully disposed of by land spreading. Heavy metal content depends on coagulant type and purity and N content is generally <1%. Soil additions of highly reactive Al and Fe hydrous oxide compounds in water plant sludges can fix P, potentially inducing P deficiencies in crops.

KEYWORDS: Sludge, heavy metals, phosphorus, availability, land application

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INTRODUCTION

The major difference between water treatment and wastewater treatment is the intended use of the treated water. Whereas wastewater is treated and discharged back into the natural water system, municipal water treatment must provide a potable water supply for human consumption. Assuring a safe drinking water supply requires the use of various processes and chemicals to remove undesirable constituents such as dissolved gases and minerals.

Sludges and wash water produced by water treatment are highly variable in composition, containing materials removed from the raw water as well as chemicals (alum or FeCl_3) added as coagulants during treatment. Direct discharge of these wastes to a watercourse was once standard practice, until federal and state regulations made this method of disposal unacceptable. Under the federal Water Pollution Control Act Amendments of 1972, water plant sludges are categorized as industrial wastes, which have stringent disposal requirements (AWWA, 1978).

Currently, the most common methods of disposal of water plant sludges include landfilling and discharge to a municipal sewage system. However, dewatering, a costly process, is required before landfilling, and landfill space is becoming more and more limited. An alternative to landfill disposal is land application of the water plant sludge. Although land application of wastewater sludges has been widely investigated, limited documentation exists on land treatment of water plant sludges. Barring a grossly contaminated water source, water treatment residuals have low potential for adverse environmental effects compared to wastewater sludges.

In a previous study, Elliott and Singer (1988) performed a greenhouse investigation using a FeCl_3 sludge and reached two major conclusions. First, the naturally alkaline condition of the sludge helped immobilize toxic heavy metals and reduce the potential for plant uptake. Second, although a possible beneficial use of the sludge would be as a lime substitute for agricultural purposes, overly alkaline conditions must be avoided to insure that precipitation-induced nutrient (such as phosphorus) deficiencies do not occur.

A major objective of the present study was to characterize several sludges gathered from water utilities using a range of coagulant chemicals and dosages. The few relevant studies reported in the literature provide analysis of a single sludge; information on sludge variability was therefore warranted. Using a chemical fractionation procedure, we attempted to document the distribution of the sludge metals among fractions of varying reactivity. A second purpose was to examine the effect of sludge amendments on the phosphorus retention by soils. The overall intention was to provide insight into the use of land application as an environmentally and agronomically sound method for disposal of water plant sludges.

MATERIALS AND METHODS

Drinking water sludges were collected from nine different locations in Pennsylvania. Three were alum sludges, five used FeCl_3 as the primary coagulant and one plant used synthetic polymers alone.

Two A-horizon soils with markedly different properties, Evesboro sand (Typic Quartzipsamment) and Vergennes silt loam (Glossaquic Hapludalf) were used in this study. The mineralogical properties of these soils has been extensively characterized (Johnson and Chu, 1983). Selected properties of the soils are given in Table 1.

To assess the mobility of the metals in the sludge, a five-step sequential fractionation was performed on the sludges. This procedure, based on similar schemes by Tessier et al. (1979); Pickering, (1986); and Gibson and Farmer, (1986), divides metals into the following fractions, exchangeable, acid soluble, Fe-Mn oxide bound, organically bound, and residual. The reagents and conditions of this method are given in Table 2.

The effect of water sludge application on P availability of the soil was investigated by developing an adsorption-desorption curve for sludge-soil mixtures (White, 1981). To prepare sludge soil mixtures, various percentages of an air-dried alum sludge were mixed with air-dried soil. One gram of soil or sludge-soil mixture was placed in 25 ml of solution containing various levels of soluble P. The samples were allowed to equilibrate for 24 hours. The suspensions were filtered and the filtrate P analysis performed by the ascorbic acid method (APHA, 1985). The quantity of soluble P gained or lost per gm of soil was plotted against the final equilibrium P concentration in solution.

RESULTS AND DISCUSSION

SLUDGE CHARACTERIZATION

Heavy Metal Content

Transfer of toxic heavy metals to the food chain or water supplies is the primary long-term environmental concern in land application of waste materials. Heavy metal buildup in soils is the factor determining the useful lifetime of a site used for sludge disposal. While metal contamination of wastewater sludges has been extensively studied, analyses of water plant sludges are quite limited. The few pertinent studies provide data for a single sludge (Schmitt and Hall, 1975; Elliott and Singer, 1988).

The analysis of these sludges (Table 3) included concentrations for the six metals normally regarded as most important for land applied wastes. For comparison, the maximum allowable metal levels are also provided, these concentrations often used by state regulatory agencies as upper limits for sludges suitable for application to agricultural land. The metal content of the water plant sludges was generally well below these maximum allowable levels (Table 3).

One notable exception was the relatively high Ni levels in three sludges, two FeCl_3 sludges were at or over the 200 ppm maximum (189 and 218 ppm), and the cationic polymer (CP) sludge contained 643 ppm Ni, greatly exceeding the suggested maximum level. In a previous study (Elliott and Singer, 1988), another FeCl_3 sludge was determined to have a relatively high Ni content (307 ppm). The data suggests that type and purity of the coagulant chemicals may be largely responsible for the metal contamination in the sludges. Although the relative proportions of coagulant chemical and removed colloidal materials impacts on the metal content of the composite sludge, the data reveals some general trends. The FeCl_3 sludges contained relatively high concentrations of Cu, Cr, and Ni. The alum sludges were not markedly high in any of the metals, but had substantially lower levels of Cr and Ni than the FeCl_3 or CP sludge. The CP sludge contained 2015 ppm Zn, 10 ppm Cd, and 643 ppm Ni, levels that were extremely high relative to the median sludge values (Table 3). Since this sludge was the only one generated using CP alone, generalization about the metal content of such sludges is not justified. Pinpointing the cause of these high levels would require a metals mass balance for that particular water treatment plant.

Total metal concentrations are often of limited value because they do not necessarily reflect the environmentally relevant metal forms. In order to assess the potential mobility and bioavailability of the sludge-borne metals, a five-step chemical extraction scheme was performed on the sludges. The exchangeable, acid soluble, and possibly, the organically bound metals can be considered as labile, or potentially bioavailable or leachable. The oxide bound and residual metals are essentially inert under conditions encountered in natural environments. Figures 1 and 2 show the distribution of Cr and Cd, respectively, in the first four fractions as a function of the total metal content represented by these fractions. From Fig. 1, it can be seen that the majority of the Cr is bound in the Fe-Mn oxide fraction. This feature, coupled with the relatively small contribution by the exchangeable and acid soluble fractions, indicates a relatively low Cr availability associated with these sludges. The dominance of the oxide bound fraction in the distribution of metals was also characteristic for Cu, Ni, and Zn.

The behavior of Cd was quite different (Fig. 2). While the oxide components retained little of the Cd, the acid soluble and organically bound fractions represented the major share of the nonresidual metal. The data in Figs. 1 and 2 is consistent with the adsorption properties of these heavy metals on various soil components. Kinniburgh et al. (1976) reported that metal adsorption onto amorphous Fe and Al oxides followed the sequence $\text{Cu} > \text{Pb} > \text{Zn} > \text{Cd}$. Elliott et al. (1986) found this same adsorption sequence for mineral soils but for soils with substantial organic matter content, Cd was preferentially absorbed over Zn. Consistent with this strong association of Cd with organic function groups, a sizeable portion of the Cd was bound by organic matter (Fig. 2). Based on this behavior, it would be expected that among the six metals, Cd would likely have the greatest mobility in soils amended with water plant sludges. Fortunately, the level of Cd in the sludges was consistently low (Table 3).

Sludge Nitrogen Content

While wastewater sludges typically contain about 4% total N (EPA, 1983), it was not expected that water plant sludges would have any appreciable N content. Bugbee and Frink (1985) analyzed an alum sludge and concluded it contained "few if any plant nutrients". The total Kjeldahl N (TKN) of a FeCl_3 sludge was reported to be a scant 554 ppm (0.05%) (Elliott and Singer, 1988). Surprisingly, the TKN of the water plant sludges of this study ranged from 0.39 to 1.00% with a mean value of 0.54%. Similarly, Lin (1988) reported that an alum sludge had 0.47% total N.

We have not gathered sufficient data in this study to pinpoint the source of this N, but two possibilities exist. The N could originate from the water source, perhaps as soluble N compounds or as a component of nitrogenous organic matter. If the water source contains 0.5 ppm NO_3^- , the sludge would contain roughly 0.8% N assuming complete NO_3^- removal and 500 lbs. of sludge produced per million gallons of water treated. The organic matter content of water sludges can also contribute N. Reed et al. (1988) quote a source listing such sludges having from 15-25% organic matter. Bugbee and Frink (1985) found an alum sludge to contain from 25-35% organic matter based on loss on ignition. Since the C/N is about 10/1 in most humus (Fuller and Warrick, 1985), it is plausible that the N is a constituent of the sludge organic matter.

A second possible source of N is the organic polyelectrolytes added to aid coagulation. The polymers may be N-containing polyacrylamide compounds. Since some direct filtration plants may use polymer dosages as high as 3-5 mg/l, these polymers may represent a substantial contribution to the final sludge. The common cationic polymer, Cat-floc, contains about 11% N. If dosed at 1 mg l^{-1} , the resulting sludge would contain 0.2% N, assuming 500 lbs. sludge produced per million gallons of water treated. This theory does not neatly fit our data, however, as the sludge with the highest N value does not contain polymer and the plant using polymer only had a lower than average N content (0.42%).

SLUDGE EFFECTS OF SOIL P STATUS

The most well documented use of water plant residues on cropland involves the substitution of lime softening sludges for commercial lime to modify soil pH. The benefits of such alkaline sludges has long been recognized (Fleming, 1957). Even when FeCl_3 is the primary coagulant and lime was used solely for pH adjustment, the resulting sludge had a 53% calcium carbonate equivalence (Elliott and Singer, 1988). Similarly, Bugbee and Frink (1985) found that an alum sludge acted as a liming material.

A few recent studies have investigated the effect of water plant sludges on soils and crops. These have been both greenhouse studies (Bugbee and Frink, 1985; Elliott and Singer, 1988) and field investigations (Grabarek and Krug, 1987; Lin, 1988). The consensus of much of this work is that water plant sludges, when applied at rates of a few tons per acres, have little adverse environmental impact. Heavy metal uptake by crops seems limited with proper soil pH adjustment (Elliott and Singer, 1988; Lin, 1988). Large doses of alum sludges do not produce Al phytotoxicity (Grabarek and Krug, 1987). Lin (1988) concluded that alum sludge application had "neither beneficial nor adverse effects on soils and crops."

One potential negative impact when these materials are land applied is a reduction in plant available P. When grown in potting media amended with water sludges, tomato (Elliott and Singer, 1988) and lettuce (Bugbee and Frink, 1985) leaves showed purple venation, characteristic of P deficiency. Tissue analysis confirmed significantly lower P levels for FeCl_3 sludge amended trials (Table 4). The data in this table are for 6% sludge (60 tons/acre), which is a high sludge loading rate, at least in the context of wastewater sludges. Extremely high sludge loading rates (33-67% of the potting media) induced P deficiencies in lettuce in the Bugbee and Frink (1985) study. Rengasamy et al. (1980) found significant reduction in P uptake by maize when alum sludge was applied at 20t ha^{-1} .

Quantitative assessment of the effect of sludge application on the P status of a soil can be made by developing an adsorption-desorption curve (more simply adsorption curve) for the soil (White, 1981). Figure 3 shows a typical diagrammatic form of the P adsorption curve. Two important parameters can be derived from such a plot. If a soil is added to a solution containing a given P level and P is neither adsorbed or desorbed from the soil, the P concentration is called the Equilibrium Phosphorus Concentration (EPC). The slope of the curve (β) at the EPC indicates the amount of P taken up for every unit of P that is released into the solution. This β value is called the Phosphate Buffer Capacity. The higher the β , the poorer the expected response to fertilization.

Figure 4 shows the P adsorption curve for the Vergennes soil as affected by various levels of alum sludge. It can be seen that sludge amendment had a dramatic effect on the soil's P retention properties. Even at the 2% rate, the slope of the adsorption curve was increased sharply. The EPC and β values for this data are plotted in Fig. 5 as well as data for the Evesboro soil. For both soils, the EPC value decreases with sludge loading, indicating less available P. The EPC values seem to be quite similar over a range of sludge loading rates for the two soils. The β values increased for both soils, with a larger increase noted for the Evesboro soil.

For low sludge loading rates, both the EPC and β parameters seemed to be more markedly impacted for the Evesboro soil. The soils are quite different in their physical and chemical makeup (Table 1). The Evesboro soil is very coarse textured, low in organic matter, and has a small Fe-oxide content. All of these properties seem to be associated with weak P adsorption capacity. In a review of P retention and release by soils, White (1981) cites research finding documenting P adsorption capacity being correlated with clay content, extractable Fe and Al, and organic C content. From the textural analysis (Table 1), the surface area of the Evesboro soils should be considerably lower than that of the Vergennes soil. Freshly-precipitated Fe and Al hydrous oxides produced in the coagulation process, have extremely high surface areas and are quite reactive as P sorbing surfaces. Freshly precipitated Fe and Al gels sorb 1-2 orders of magnitude more inorganic P than their crystalline analogues (McLaughlin et al., 1981). Addition of sludge to the Evesboro soil would be expected to dramatically enhance P sorption capacity. This effect was apparently less for the Vergennes soil which has substantial P adsorbing constituents even in an unamended form.

CONCLUSIONS

1. Mean total metal levels in sludges produced from treatment of water for potable uses are generally within limits specified for sewage sludges applied to agricultural land. Three sludges had Ni levels at or above the maximum allowable levels. Management of certain sludges for land application may require a use restriction for crops sensitive to Ni phytotoxicity.
2. Chemical fractionation studies revealed that Cr, Cu, Ni, and Zn in sludges are retained primarily in the Fe-Mn oxide occluded fraction. This fraction represents a relatively nonreactive metal pool and mobilization of these metals in sludge-amended soils would occur only under extreme environmental conditions. However, the acid soluble and organically bound fractions contained the majority of the total nonresidual Cd. It is fortunate that Cd levels tend to be low in water plant sludges, for its partitioning behavior suggests potentially significant mobility in the soil profile.
3. Water plant sludges contained an average of 0.54% TKN. This may lead some state regulatory agencies to permit land application of water plant sludges based on N loading in a manner similar to wastewater sludges. The availability of N in water plant sludges is expected to be quite low and the use of mineralization rates typical for wastewater sludges may unnecessarily restrict loading rates. The dynamics of N transformations in soils amended with water plant sludges is a research topic that warrants investigation.
4. A reduction in P availability results from the strong fixation of PO_4^{3-} by hydrous Fe and Al oxide compounds in water plant sludges. The effect seems to be enhanced in coarse-textured soils with limited amounts of P-sorbing constituents. Use of water plant sludges in agricultural production will require management strategies (appropriate pH control, judicious crop selection, supplemental fertilization) to circumvent potential P deficiencies in crops.

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Table 1. Selected Properties of the Soils

<u>Soil</u>	<u>Horizon</u>	<u>Texture</u>	<u>Sand/Silt/Clay (%)</u>	<u>OM (%)</u>	<u>Fe Oxides (%)</u>
Evesboro	Ap	Sand	89.6/8.2/2.3	0.67	0.2
Vergennes	Ap	Silt loam - Silty clay loam	8.6/65.1/26.2	2.83	1.6

Table 2. Chemical Fractionation Procedure.

<u>Step</u>	<u>Reagent</u>	<u>Shaking Time (hr)</u>
Exchangeable	1M MgCl ₂ (pH 7.0)	1.0
Acid Soluble	1M Na-acetate adjusted to pH5 with acetic acid	5.0
Fe-Mn Oxide Bound	0.175M ammonium oxalate and 0.1M oxalic acid	4.0
Organically Bound	0.1M Na-pyrophosphate	24.0
Residual	HNO ₃ /HClO ₄ /HF heated at 140°C	3.5

Table 3. Total Metal Levels (ppm) in Water Plant Sludges.

<u>Metal</u>	<u>Mean^a</u>	<u>Median</u>	<u>Range</u>	<u>Maximum^b Allowable</u>
Cd	2	2	BD ^c -10	25
Cu	224	220	135-485	1000
Cr	170	64	40-406	1000
Ni	162	122	26-643	200
Pb	231	127	18-840	1000
Zn	719	580	195-2015	2500

^aMean of nine sludges

^bRecommended maximum allowable metal levels for municipal sludges used in agricultural production

^cBD = below detection.

Table 4. Effect of Liming and FeCl₃ Sludge Addition on Tomato Shoot Weight and P Content.

<u>Description</u>	<u>pH</u>	<u>Fresh Weight (gm)</u>	<u>Tissue P (%)</u>
Control	4.99 ^{a*}	8.38 ^a	0.50 ^a
Limed	7.51 ^b	6.63 ^a	0.44 ^a
6% Sludge	7.68 ^b	3.40 ^b	0.24 ^b

Values followed by same notation within a column do not differ at the 0.05 significance level, according to Duncan's Multiple Range Test. Data from Elliott and Singer (1988).

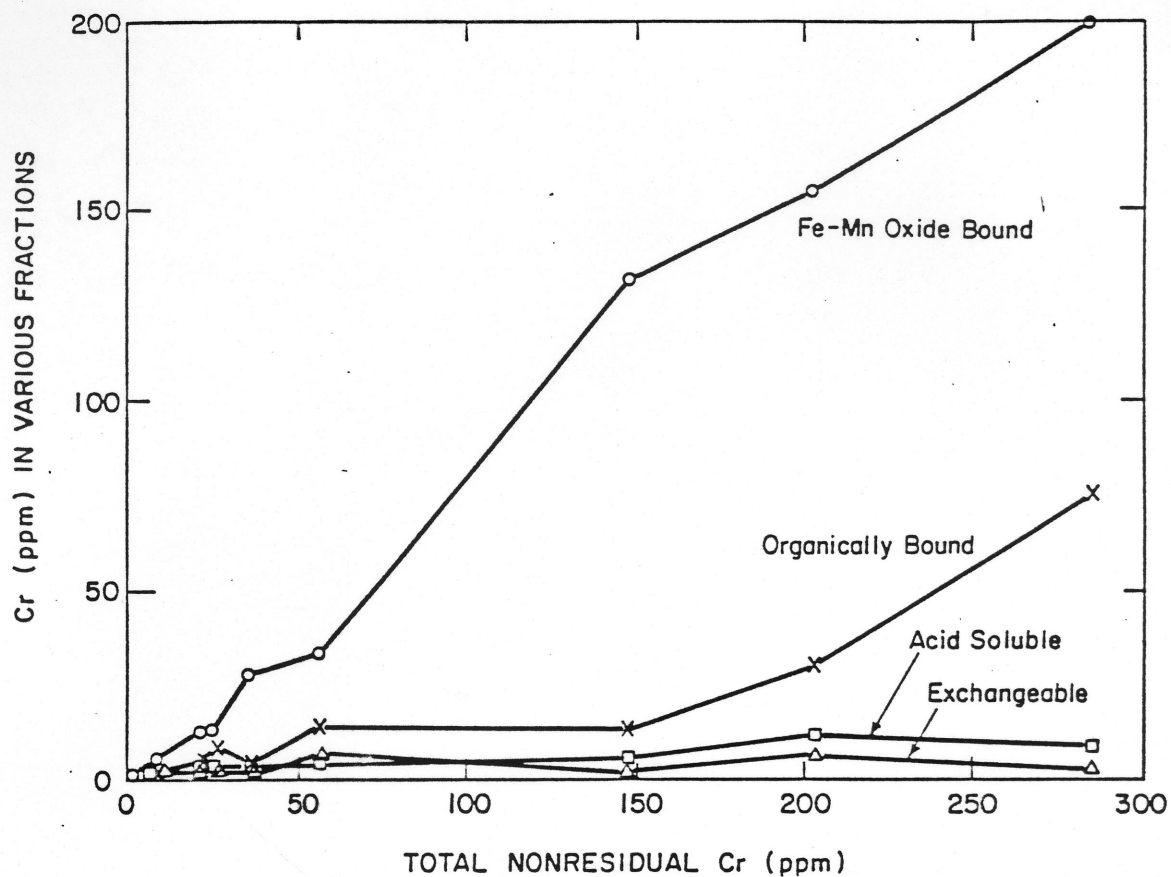


Figure 1. Distribution of Nonresidual Sludge Cr Among Various Fractions.

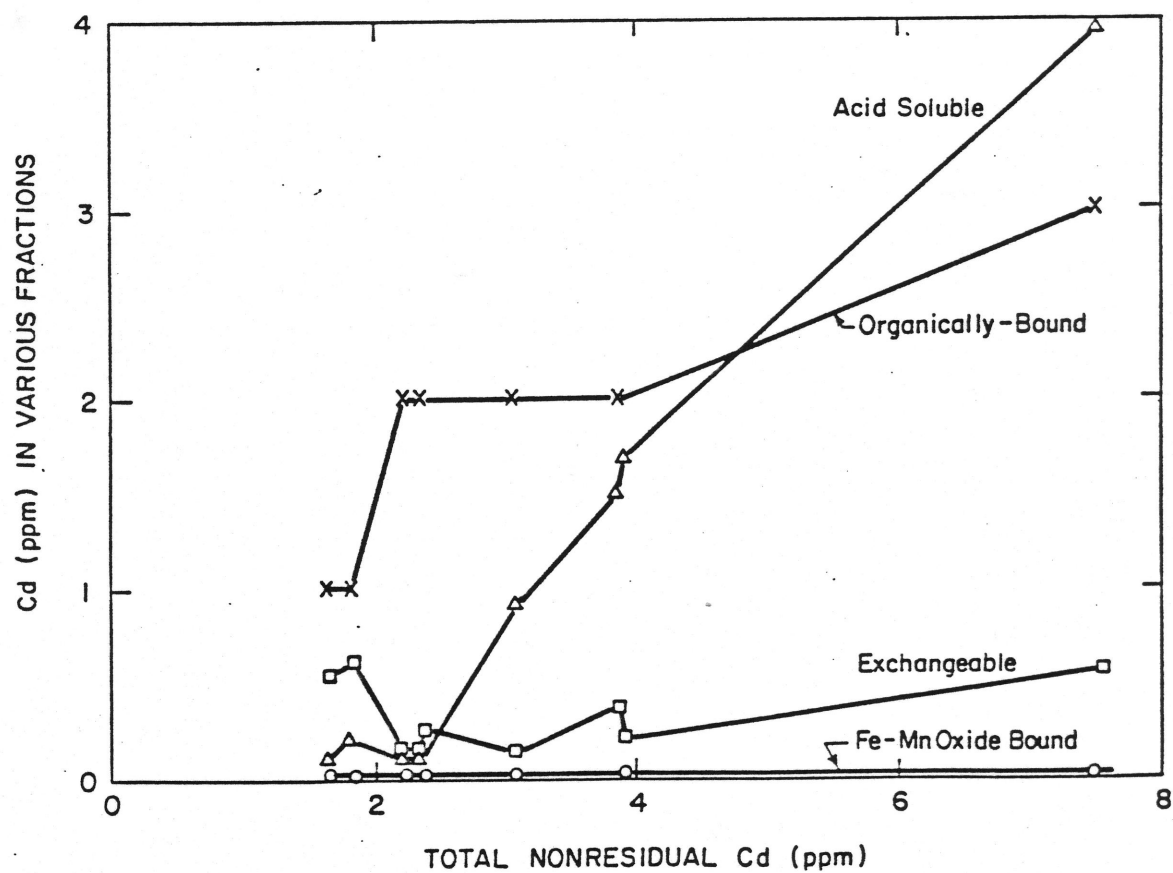


Figure 2. Distribution of Nonresidual Sludge Cd Among Various Fractions.

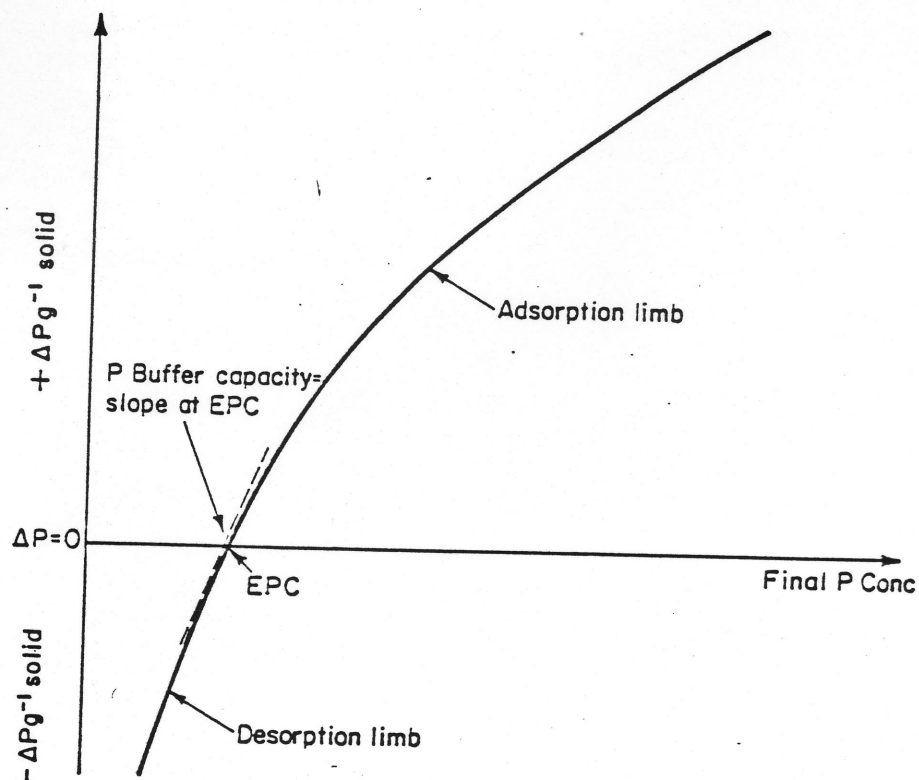


Figure 3. Soil P Adsorption-desorption Curve (From White, 1981).

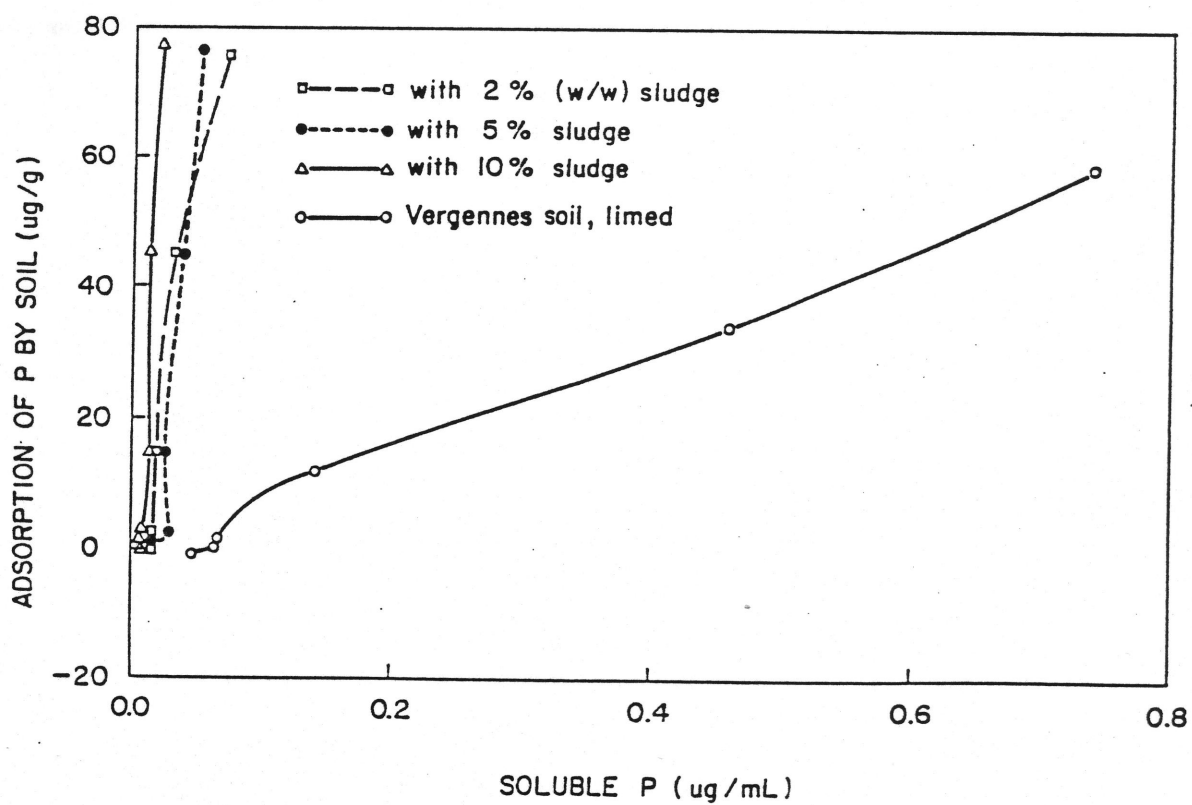


Figure 4. Effect of Sludge Addition on P Adsorption by Vergennes Soil.

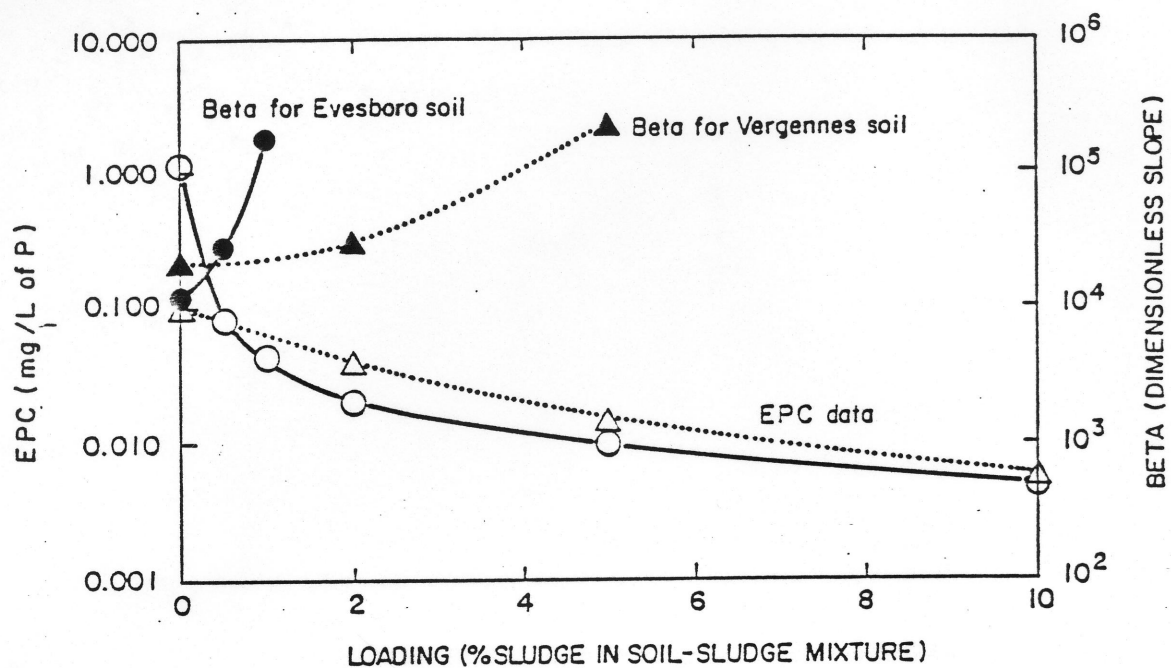


Figure 5. Equilibrium P Concentration (EPC) and Phosphorus Buffer Capacity (β) as a Function of Sludge Loading.